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Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes

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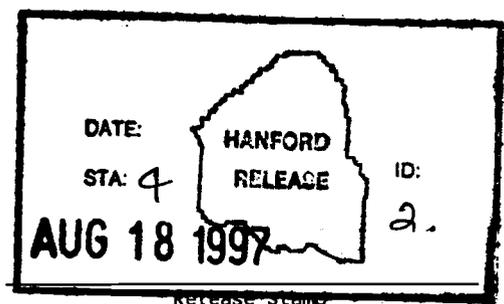
Key Words: global, inventory, best-basis, waste

Abstract: This report presents work performed to date to establish standard global and tank-by-tank component inventory estimates. This work includes: global inventory reconciliations for 21 nonradioactive components, that account for approximately 80 percent of the total tank waste inventory (hydroxide accounts for an additional 15 of the remaining 20 percent), and tank-by-tank reconciliations for 41 tanks. To ensure that inventory values resulting from these evaluations are technically defensible and reproducible, every attempt is being made to document the bases, i.e., originating sources of assumptions, data, and background information, that support these values. Further refinement of the global inventory data, will be performed as necessary.

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Revision 0

**STANDARD INVENTORIES OF
CHEMICALS AND RADIONUCLIDES
IN HANFORD SITE TANK WASTES**

August 1997

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EXECUTIVE SUMMARY

This report presents global best-basis inventory estimates for chemical and radionuclide components in the 177 Hanford Site underground storage tanks. Tank-by-tank best-basis inventories have also been developed as part of the Standard Inventory task and will be published in the respective tank characterization report for each tank. In instances where a tank characterization report has not been issued, the best-basis inventory is issued as a preliminary tank characterization report. The methodology for deriving global chemical and radionuclide inventories, as well as individual tank inventories, is also presented in this report.

Tank-by-tank waste inventories include 26 chemical and 46 radionuclide components for each of the 177 single- and double-shell underground waste storage tanks on the Hanford Site. The global waste inventories reported herein represent the sum of each chemical or radionuclide component (e.g., metric tons of sodium, nitrate, aluminum, etc., and curies of $^{239/240}\text{Pu}$, ^{137}Cs , ^{90}Sr , etc.) presently stored in the tanks. The chemical analytes selected represent over 99 weight percent of the tank contents, and the radionuclides represent over 99 percent of the activity. Global and tank-by-tank inventories now serve as waste composition data for Tank Waste Remediation System process flowsheet modeling work, safety analyses, risk assessments, and waste retrieval, treatment, and disposal system design.

Information used to establish global inventories originated from key historical records (e.g., essential material purchase records), from various chemical flowsheets used in

reprocessing of irradiated Hanford Site reactor fuels, and from calculations of radionuclide isotope generation and decay. Tank-by-tank inventories are most often based on sample analysis results.

In recent years, Los Alamos National Laboratory has revisited some of the historical information for the development of the Hanford Defined Waste model.^{1, 2} This model generates chemical and radionuclide inventory estimates for each of the underground tanks as well as global inventories for both single- and double-shell tanks. These Hanford Defined Waste model inventories are often inconsistent with the currently used Tank Waste Remediation System inventories.

A major objective of the Standard Inventory task was to resolve these inconsistencies and to establish standard global and tank-by-tank inventories that benefit a multitude of waste management and disposal activities. Evaluations performed to resolve differences among reported values involved reviewing the bases, i.e., assumptions, data, background information, etc., that support these values. To ensure that inventory values resulting from

¹Agnew, S. F., J. Boyer, R. A. Corbin, T. B. Duran, J. R. FitzPatrick, K. A. Jurgensen, T. P. Ortiz, and B. L. Young, 1996, Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 3, LA-UR-96-858, Los Alamos National Laboratory, Los Alamos, New Mexico.

²Agnew, S. F., J. Boyer, R. A. Corbin, T. B. Duran, J. R. FitzPatrick, K. A. Jurgensen, T. P. Ortiz, and B. L. Young, 1997, Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4, LA-UR-96-3860, Los Alamos National Laboratory, Los Alamos, New Mexico.

these evaluations are technically defensible and reproducible, every attempt was made to document all bases that support these values.

Global inventory values resulting from nonradioactive component evaluations are summarized in Table ES-1 along with the corresponding document section containing the evaluation discussion. Global inventory values resulting from the radionuclide component evaluations are summarized in Table ES-2. The global inventories, developed primarily by purchase records and flowsheets, have yet to be reconciled with global inventories that can be calculated by summation of the individual tank-by-tank inventories. This work is planned for fiscal year 1998.

*Table ES-1. Summary of Global Best-Basis Inventories for Nonradioactive Components.
(2 Sheets)*

<i>Component</i>	<i>Best-basis inventory (MT)</i>	<i>Corresponding document section</i>
<i>Aluminum</i>	<i>7,845</i>	<i>5.1</i>
<i>Bismuth</i>	<i>580</i>	<i>5.2</i>
<i>Calcium</i>	<i>214</i>	<i>5.3</i>
<i>Carbonate</i>	<i>4,830</i>	<i>5.4</i>
<i>Cerium</i>	<i>8.8</i>	<i>5.5</i>
<i>Chloride</i>	<i>500</i>	<i>5.6</i>
<i>Chromium</i>	<i>785</i>	<i>5.7</i>
<i>Fluoride</i>	<i>1,360</i>	<i>5.8</i>
<i>Hydroxide</i>	<i>23,000</i>	<i>5.9</i>
<i>Iron</i>	<i>1,230</i>	<i>5.10</i>
<i>Lanthanum</i>	<i>51</i>	<i>5.11</i>
<i>Lead</i>	<i>279</i>	<i>5.12</i>
<i>Manganese</i>	<i>105</i>	<i>5.13</i>

*Table ES-1. Summary of Global Best-Basis Inventories for Nonradioactive Components.
(2 Sheets)*

<i>Component</i>	<i>Best-basis inventory (MT)</i>	<i>Corresponding document section</i>
<i>Aluminum</i>	<i>7,845</i>	<i>5.1</i>
<i>Mercury</i>	<i>2.1</i>	<i>5.14</i>
<i>Nickel</i>	<i>111</i>	<i>5.15</i>
<i>Nitrite and Nitrate</i>	<i>85,700</i>	<i>5.16</i>
<i>Phosphate</i>	<i>6,000</i>	<i>5.17</i>
<i>Potassium</i>	<i>481</i>	<i>5.18</i>
<i>Silicon</i>	<i>570</i>	<i>5.19</i>
<i>Sodium</i>	<i>54,200</i>	<i>5.20</i>
<i>Sulfate</i>	<i>5,000</i>	<i>5.21</i>
<i>Strontium</i>	<i>31.3</i>	<i>5.22</i>
<i>Total organic carbon</i>	<i>4,000</i>	<i>5.23</i>
<i>U_{TOTAL}</i>	<i>965</i>	<i>6.3</i>
<i>Zirconium</i>	<i>440</i>	<i>5.24</i>
<i>Minor components</i>		
<i>Cadmium</i>	<i>8.2</i>	<i>7.3.1</i>
<i>Silver</i>	<i>8.93</i>	<i>7.3.2</i>
<i>Thorium</i>	<i>25.6</i>	<i>7.3.3</i>
<i>Tungsten</i>	<i>15.9</i>	<i>7.3.4</i>

*Table ES-2. Summary of Global Best-Basis Inventories for Radioactive Components.
(3 Sheets)*

<i>Radionuclide</i>	<i>Ci^a</i>	<i>Corresponding document section</i>
<i>³H</i>	<i>3.40 E+04</i>	<i>6.1</i>
<i>¹⁴C</i>	<i>4.81 E+03</i>	<i>6.1</i>
<i>⁵⁹Ni</i>	<i>9.34 E+02</i>	<i>6.1</i>
<i>⁶⁰Co</i>	<i>1.23 E+04</i>	<i>6.1</i>
<i>⁶³Ni</i>	<i>9.20 E+04</i>	<i>6.1</i>
<i>⁷⁹Se</i>	<i>7.73 E+02</i>	<i>6.1</i>

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Table ES-2. Summary of Global Best-Basis Inventories for Radioactive Components.
(3 Sheets)

Radionuclide	Ci ^a	Corresponding document section
⁹⁰ Sr	7.16 E+07	6.1 and 6.2
⁹⁰ Y	7.16 E+07	6.1
⁹³ Zr	3.63 E+03	6.1
^{93m} Nb	2.69 E+03	6.1
⁹⁹ Tc	3.26 E+04	6.1
¹⁰⁶ Ru	1.04 E+05	6.1
^{113m} Cd	1.69 E+04	6.1
¹²⁵ Sb	2.08 E+05	6.1
¹²⁶ Sn	1.19 E+03	6.1
¹²⁹ I	6.30 E+01	6.1
¹³⁴ Cs	8.89 E+04	6.1
¹³⁷ Cs	4.64 E+07	6.1 and 6.2
^{137m} Ba	4.39 E+07	6.1
¹⁵¹ Sm	2.75 E+06	6.1
¹⁵² Eu	1.48 E+03	6.1
¹⁵⁴ Eu	1.47 E+05	6.1
¹⁵⁵ Eu	1.36 E+05	6.1
²²⁶ Ra	6.31 E-02	6.1
²²⁷ Ac	8.76 E+01	6.1
²²⁸ Ra	7.71 E+01	6.1
²²⁹ Th	1.81 E+00	6.1
²³¹ Pa	1.56 E+02	6.1
²³² Th	2.11 E+00	6.1
²³² U	1.23 E+02	6.1
²³³ U	4.76 E+02	6.1
²³⁴ U	3.46 E+02	6.1
²³⁵ U	1.45 E+01	6.1
²³⁶ U	9.57 E+00	6.1
²³⁷ Np	1.41 E+02	6.1
²³⁸ Pu	2.77 E+03	6.1

*Table ES-2. Summary of Global Best-Basis Inventories for Radioactive Components.
(3 Sheets)*

<i>Radionuclide</i>	<i>Cr^a</i>	<i>Corresponding document section</i>
<i>²³⁸U</i>	<i>3.22 E+02</i>	<i>6.1 and 6.3</i>
<i>²³⁹Pu</i>	<i>3.91 E+04</i>	<i>6.1</i>
<i>²⁴⁰Pu</i>	<i>8.93 E+03</i>	<i>6.1</i>
<i>²⁴¹Am</i>	<i>6.99 E+04</i>	<i>6.1</i>
<i>²⁴¹Pu</i>	<i>2.29 E+05</i>	<i>6.1</i>
<i>²⁴²Pu</i>	<i>1.16 E+00</i>	<i>6.1</i>
<i>²⁴²Cm</i>	<i>7.70 E+01</i>	<i>6.1</i>
<i>²⁴³Am</i>	<i>9.34 E+00</i>	<i>6.1</i>
<i>²⁴³Cm</i>	<i>1.00 E+01</i>	<i>6.1</i>
<i>²⁴⁴Cm</i>	<i>2.42 E+02</i>	<i>6.1</i>

^a*Decayed to January 1, 1994.*

It is recognized that the standard inventory as reported in Tables ES-1 and ES-2 will not satisfy all user's needs. Expansion of the data set to include confidence intervals and other program-specific needs (liquid/solid split, soluble/insoluble split, etc.) is already being planned. Users are referred to the Tank Characterization Database¹ for the most current Standard Inventory data set.

¹*PNL, 1994, TWINS User Guide Tank Waste Characterization Information Network System Version 4.0, PNL-8824-2, Pacific Northwest Laboratory, Richland, Washington.*

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LIST OF TERMS

1C	First decontamination cycle of the bismuth phosphate process
2C	Second decontamination cycle of the bismuth phosphate process
1WW	Concentrated first cycle raffinate from PUREX process
224	Final decontamination/concentration cycle of the bismuth phosphate process
Al-Si	Aluminum-silicon (as in Al-Si bond layer)
ANN	Aluminum Nitrate Nonahydrate - $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$
BiPO_4	Bismuth phosphate
CAW	Current acid waste
CF	Concentration factor
CWZr2	REDOX process coating waste, zirconium cladding
CW	Cladding (coating) waste
D2EHPA	Di(2-ethylhexyl)phosphoric acid - $(\text{C}_8\text{H}_{17}\text{O})_2\text{POOH}$
DKPRO	Computer code for radioactive decay and separations processing
DN	Dilute non-complexed
DOE	U.S. Department of Energy
DQO	Data Quality Objective
DSC	Differential scan calorimetry
DSSF	Double-shell slurry feed
DST	Double-shell tank
DTPA	Diethylene-triamine penta acetate
EB	Evaporator bottoms
EDTA	Ethylenediamine tetra acetic acid
EIS	Environmental Impact Statement
FEIS	Final Environmental Impact Statement
FIC	Food Instrument Company (FIC) Automatic Surface Level Gauge
Fuel Grade	Refers to plutonium material containing $> 6 \text{ wt} \% \text{ }^{240}\text{Pu}$ after irradiation
FY	Fiscal year
GWD	Gigawatt days
HAPO	Hanford Atomic Products Operation
HEDTA	N(2-hydroxyethyl)ethylenediamine tetra acetate

LIST OF TERMS

HDW	Hanford Defined Waste
HEDR	Hanford Environmental Dose Reconstruction Project
HLW	High-level waste
HTCE	Hanford Tank Content Estimate
HSW	High-salt waste
ICP	Inductively coupled plasma spectroscopy
I&E	Internal & external (as in I&E cooled fuel elements)
IDB	Integrated database
ITS	In-tank solidification
LANL	Los Alamos National Laboratory
LAW	Low Activity Waste
LLW	Low-level waste
LWPR	Light Water Pressure Reactor
MCi	Mega curie (1 E+06 Ci)
MT	Metric ton (1 MT = 1,000 kg)
MTHM	Metric ton of heavy metal
MTU	Metric ton of uranium
MUF	Material unaccounted for
MW	Metal waste
MWd	Megawatt days
NCAW	Neutralized current acid waste
NCRW	Neutralized cladding removal waste
NPH	Normal paraffin hydrocarbon
NPR	New Production Reactor (N Reactor)
NTA	Nitilotriacetic acid
ORIGEN	Oak Ridge Isotope Generation (computer code)
ORNL	Oak Ridge National Laboratory
PCW	PUREX process cladding removal waste
P3	Aging waste from PUREX process high-level waste (1983 to 1988)
PF	Partitioning factor
PFP	Plutonium Finishing Plant (Z Plant)

LIST OF TERMS

PHMC	Project Hanford Management Contractor
PL1	PUREX process low-level waste (1956 to 1972)
PL2	PUREX process low-level waste (1983 to 1988)
PN	Partially neutralized
PNNL	Pacific Northwest National Laboratory
ppm	Parts per million
PRF	Plutonium Reclamation Facility
PRTR	Plutonium Recycle Test Reactor
PTA	Phosphotungstic acid - $P_2O_5 \cdot 24WO_3 \cdot 44H_2O$
PUREX	Plutonium-uranium extraction (process or plant)
RCRA	<i>Resource Conservation and Recovery Act of 1976</i>
RCW	Reduction and oxidation process cladding waste
REDOX	Reduction and oxidation (process or plant)
RIBD	Radioisotope buildup and decay
RMC	Remote Mechanical C
RSD	Relative standard deviation
S	Analytically determined inventories
SORWT	Sort on radioactive waste type
SST	Single-shell tank
TBP	Tri-butyl phosphate - $(C_4H_9O)_3PO$
TCD	Tank Characterization Database
TCR	Tank Characterization Report
TGA	Thermo-gravimetric analysis
TH1	Thoria high-level waste or cladding waste (1966 campaign)
TH2	Thoria high-level waste or cladding waste (1970 campaign)
TLM	Tank Layer Model
TOC	Total Organic Carbon
TRAC	Track Radioactive Components
Tri-Party Agreement	<i>Hanford Federal Facility Agreement and Consent Order</i>
TRU	Transuranic
TWRS	Tank Waste Remediation System

LIST OF TERMS

UNH	Uranyl Nitrate Hexahydrate - $UO_2(NO_3)_2 \cdot 6H_2O$
UR	Uranium recovery
UR/TBP	Uranium recovery/tri-butyl phosphate (process or plant)
WESF	Waste Encapsulation Storage Facility
WHC	Westinghouse Hanford Company
WSTRS	Waste Status and Transaction Record Summary

STANDARD INVENTORIES OF CHEMICALS AND RADIONUCLIDES IN HANFORD SITE UNDERGROUND TANK WASTES

1.0 INTRODUCTION

After the primary mission at the Hanford Site changed from plutonium production to environmental restoration, the U.S. Department of Energy (DOE) established the Tank Waste Remediation System (TWRS) in 1991 to safely manage and dispose of the radioactive wastes stored in underground tanks. Key activities for the TWRS include overseeing tank farm operations and identifying, monitoring, and resolving the safety issues associated with those operations and with the tank wastes. Disposal activities involve designing equipment, processes, and facilities for retrieving wastes, and processing waste into a form that is suitable for long-term storage/disposal. Safety analyses, engineering evaluations, risk assessments, and regulatory issues connected with these activities require information about the chemical and physical properties of tank wastes. To support these activities, chemical information usually is required in the form of tank waste component inventories, on either a global (total) or tank-specific basis.

There are multiple sources of tank waste inventory information, often with inconsistent data. A task was initiated in FY 1996 to establish a best-basis standard inventory for chemicals and radionuclides in Hanford Site tank waste. The goal is to resolve differences among the many reported inventory values and to provide a consistent inventory basis for all waste management and disposal activities. This report summarizes the work performed to establish standard tank component inventories on a global basis.

1.1 BACKGROUND

Global tank waste inventory information is presently reported in several sources including the following:

- *Estimated Inventory of Chemicals Added to Underground Waste Tanks, 1944 Through 1975* (Allen 1976)
- *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987)
- *Single-Shell and Double-Shell Tank Waste Inventory Data Package for the Tank Waste Remediation System Environmental Impact Statement* (Golberg and Guberski 1995)
- *Chemical and Radionuclide Inventory for Single and Double-Shell Tanks* (Shelton 1996)

- The *Hanford Tank Content Estimate* (HTCE) documents (Brevick et al. 1997a, 1997b, 1997c, and 1997d)
- The *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 3* (Agnew et al. 1996) and the *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4* (Agnew et al. 1997a), which report predictions of the Hanford Defined Waste (HDW) model developed by Los Alamos National Laboratory (LANL)
- National databases, such as the *Federal Facility Compliance Act Database* and *Integrated Database* (DOE 1995).

Each of the inventory reports listed contains the best inventory knowledge available at the time of publication. For example, inventories reported in the *Single-Shell and Double-Shell Tank Waste Inventory Data Package for the Tank Waste Remediation System Environmental Impact Statement* (Golberg and Guberski 1995) are based on those established by Allen (1976) but also include those chemicals used in process operations after 1975. The inventories reported by Shelton (1996), in support of TWRS process flowsheet modeling work, correspond with DOE (1987) values with two exceptions: (1) TWRS double-shell tank (DST) inventories were revised to reflect sampling data, and (2) TWRS single-shell tank (SST) component inventories for aluminum, chromium, phosphate, and chloride were revised as the result of a recent (1995) evaluation of these inventories. Finally, the HDW model predictions were based on historical data and do not directly correspond to any of the global inventory reports. Not surprisingly, the various global inventory sources provide inconsistent inventory values.

Several attempts have been made over the years to distribute global component inventories among individual waste tanks. The distribution is performed by: (1) estimating component inventories using results from sample analyses, or (2) predicting component inventories based on process knowledge and historical information. The result is that tank-specific inventory sources also often report inconsistent inventory values.

Tank-specific inventories are contained in several sources including the following:

- Agnew et al. (1996 and 1997a), which report predictions of the HDW model developed by LANL
- Tank Characterization Reports (TCRs)
- The HTCE documents (Brevick et al. 1997a, 1997b, 1997c, and 1997d)
- *Radionuclide and Chemical Inventories for the Single-Shell Tanks* (Van Vleet 1993a) and *Radionuclide and Chemical Inventories for the Double-Shell Tanks* (Van Vleet 1993b)

- Multiple electronic databases, e.g., Tank Characterization Database (TCD) (PNL 1994), Braun Database (Forehand 1995), and ICF-Kaiser Hanford Company Database (Brevick et al. 1995).

The methodology used to establish a best-basis inventory on a global or tank-specific basis involves a thorough review of all pertinent information sources to identify errors, biases, inconsistencies, and missing information. The data sources typically include sample analyses, process flowsheets, waste transaction records, reactor fuel data, and essential material records. The reconciliation process used to estimate a best-basis inventory is described in Sections 5.0, 8.0, and Appendix J of this report.

This report presents work performed to establish standard global component inventory estimates. This work includes: global inventory reconciliations for 26 nonradioactive components that account for approximately 99 percent of the total tank waste inventory, global inventory reconciliations for an additional four (4) minor chemical components, and global inventory values for 46 radionuclides. Tank-by-tank best-basis inventories that include the same suite of chemicals and radionuclides for each of the 177 Hanford Site underground storage tanks will be published in the respective TCR. To ensure that inventory values resulting from these evaluations are technically defensible and reproducible, every attempt was made to document the bases, i.e., originating sources of assumptions, data, and background information, that support the inventory values.

1.2 DOCUMENT OVERVIEW

Information in this report is presented as follows:

- Section 1.0 provides background information regarding tank inventories.
- Section 2.0 provides an overview of the Standard Inventory task plan and shows where the global component inventory work fits within this plan.
- Section 3.0 summarizes data requirements that served as the foundation for the global inventory work.
- Section 4.0 introduces the chemical processes that generated waste inventories, the types and amount of fuel processed, and the system inventory losses.
- Section 5.0 presents evaluations and global best-basis inventory values for individual components.
- Section 6.0 presents the methods used to establish global inventories for 46 radionuclides.

- Section 7.0 evaluates the sources of minor chemical components that contribute to the tank waste inventories and presents the global best-basis inventories for four minor components.
- Section 8.0 defines details of the methodology used to determine the tank-by-tank best-basis inventories for each of the 177 Hanford Site underground waste storage tanks.
- Section 9.0 lists the references used in this document.
- Appendix A provides a summary of the users' data needs for chemical and radionuclide inventories.
- Appendix B presents details of fuel processing history at the Hanford Site, including production data and fuel processing rates for the various separations facilities.
- Appendix C defines the flowsheet bases for the bismuth phosphate (BiPO_4) process wastes. This information is the standard basis for use in defining global and tank-by-tank inventories.
- Appendix D defines the flowsheet bases for the Reduction and Oxidation (REDOX) process wastes. This information is the standard basis for use in defining global and tank-by-tank inventories.
- Appendix E provides calculations supporting the global aluminum inventory evaluation (Section 5.1).
- Appendix F provides calculations supporting the global lead inventory evaluation (Section 5.12).
- Appendix G provides sample fuel activity records, supporting data and assumptions referred to in the global radionuclide inventory evaluation (Section 6.0).
- Appendix H defines the basis for corrosion product (iron, nickel, and chromium) inventories.
- Appendix I defines limitations on content of the Standard Inventory.
- Appendix J contains guidance for preparing tank-by-tank inventory evaluations (i.e., best-basis inventories).

2.0 STANDARD INVENTORY TASK OVERVIEW

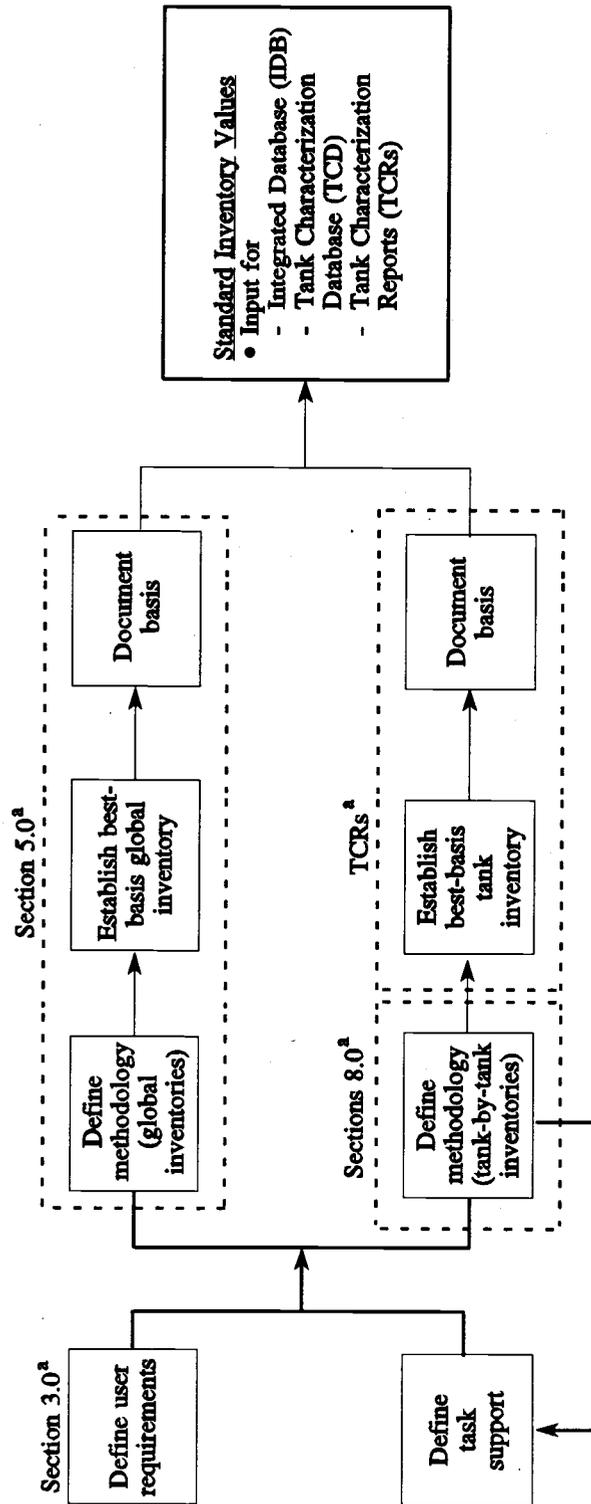
The primary objective of the Standard Inventory task was to establish best-basis waste inventory estimates that will provide consistent data for a multitude of activities related to the safe storage of the tank waste and its eventual treatment and disposal. Additional objectives within the work scope included ensuring that this information is incorporated into the Tank Characterization Database¹ (TCD) (PNL 1994), documented in a published report, and maintained in a structured and controlled manner.

The approach used to implement the Standard Inventory task objectives is defined in the *Work Plan for Defining a Standard Inventory Estimate for Wastes Stored in Hanford Site Underground Tanks* (Hodgson and LeClair 1996) and is schematically depicted in Figure 2.0-1. Specific activities within this plan involved the following:

- Identification of waste components and characteristics, i.e., data requirements, that are important to waste management and disposal activities. The results from this activity are defined in Appendix A and further summarized in Appendix I and Section 3.0.
- Development and documentation of the methodology used to evaluate both global and tank-by-tank inventories. The global and tank-by-tank inventory evaluation methodologies are defined in Sections 5.0 and 8.0, respectively.
- Determination of standard global inventory values for specified waste components by evaluating the bases, i.e., assumptions, data, and background information, that support these values. The results of this effort are documented in Section 5.1 through Section 5.24.
- Determination of standard tank-by-tank waste inventories, in terms of these components, by evaluating available sampling data and historical information. The results of this effort are to be documented in each of the respective Tank Characterization Reports (TCRs).
- Publication of the results in a form that makes the data easily accessible to data users, makes it easy to update the data set, and also allows for archiving of the data and the associated pedigree (usually published as a supporting document). The TCD (PNL 1994) was recognized as an existing resource for providing these capabilities.
- Development of a mechanism for maintenance and update of the Standard Inventory. The results of this effort are discussed later in this section.

¹Internet address is <http://twins.pnl.gov:8001/TCD/main.htm/>.

Figure 2.0-1. Schematic of Task Plan.



TCRs - Tank Characterization Reports
^a Refers to location where applicable documentation can be located.

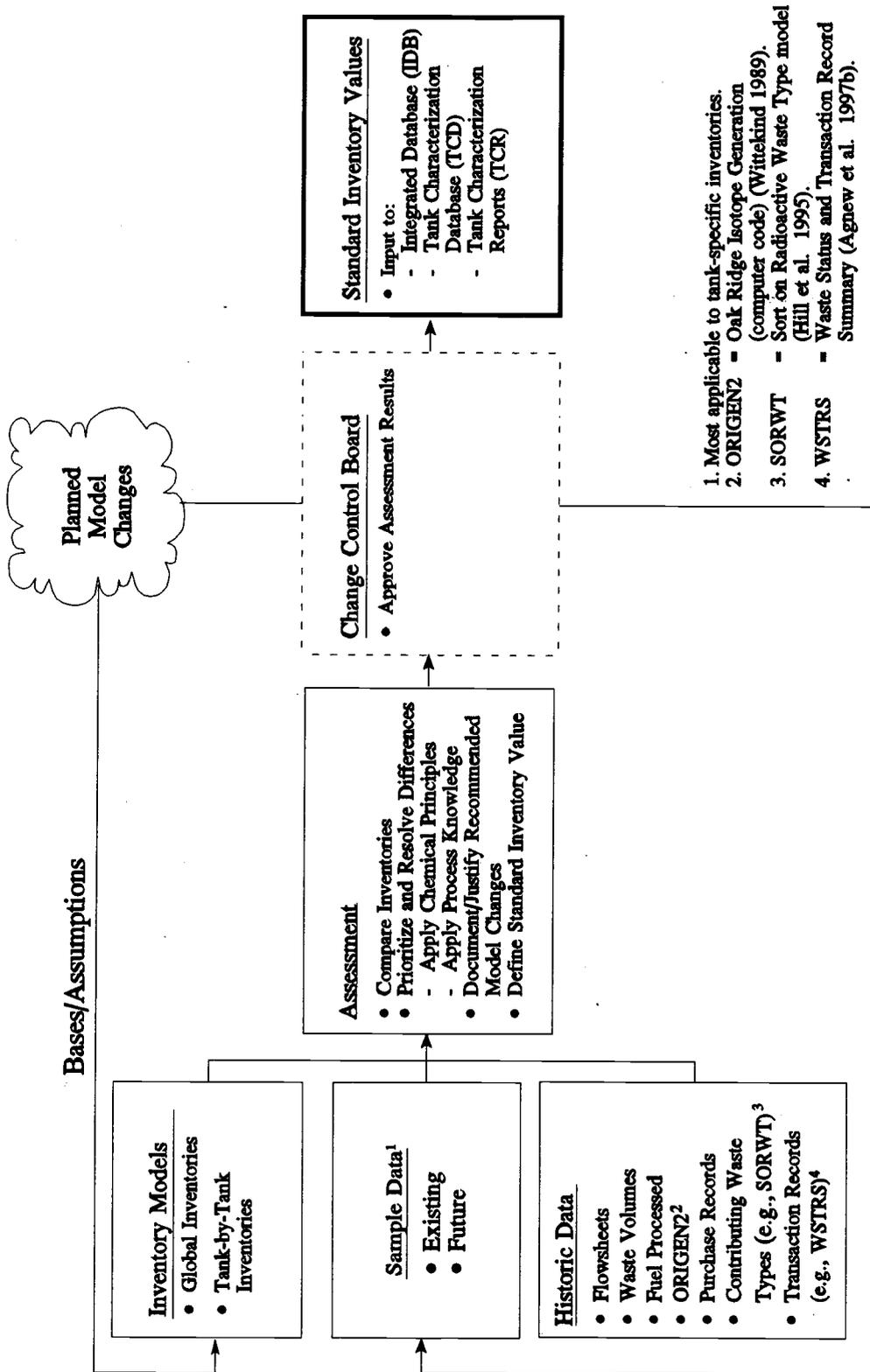
2.1 STANDARD INVENTORY MAINTENANCE

Efforts this fiscal year (1997) will focus on finalizing the initial Standard Inventory baseline that includes global and tank-by-tank inventory values for 26 chemical and 46 radionuclides. Efforts to reconcile the summation of tank inventories to the global inventory, and to improve radionuclide prediction will be pursued in fiscal year (FY) 1998. Maintenance of the Standard Inventory will begin in FY 1998.

For the Standard Inventory values to remain useful, they must be periodically updated and modified to reflect current information available on tank contents. As sample analysis results, modeling data, and other information become available, it must be evaluated and reconciled with existing data in much the same fashion that the initial Standard Inventory values were derived. Revised Standard Inventory values and their associated pedigree (supporting documentation) must then be approved by the change control process and incorporated into the TCD. In instances where the prompt for update is new sample analysis results, the best-basis evaluation will also be documented in the respective TCR. Figure 2.0-2 illustrates the Standard Inventory maintenance process.

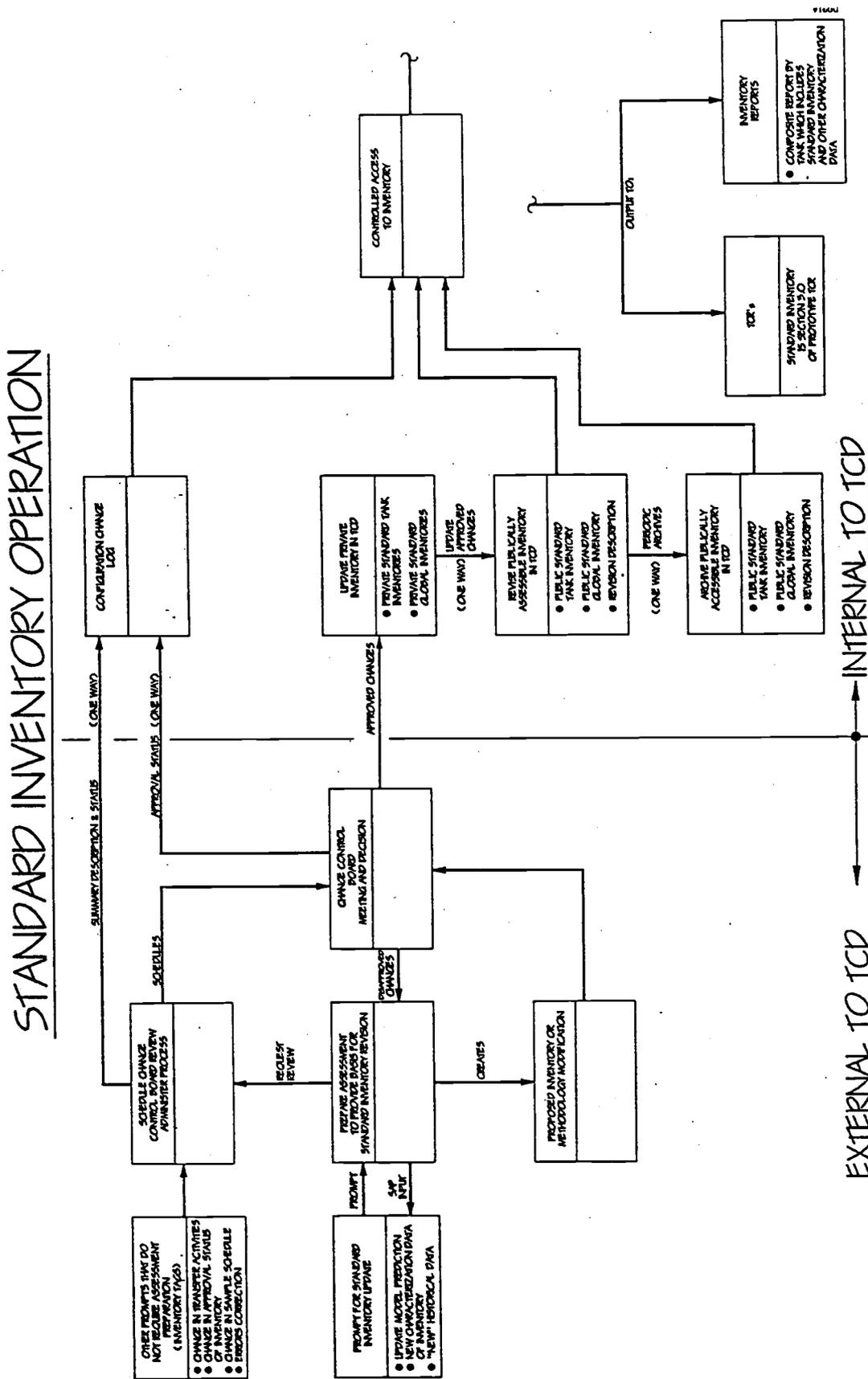
Figure 2.0-3 provides an overview of the Standard Inventory change control process developed during the Standard Inventory User and Policy Workshops conducted in July 1996 (LeClair 1996). This figure serves as a basis for developing the Standard Inventory change control procedures and policies. These procedures and policies will define protocol to be followed for proposing changes to the inventory, approving changes to the inventory, and updating the TCD to report revised Standard Inventory values. Responsibilities, authorities, methodology application, quality assurance practices, and database configuration control will also be addressed by these procedures and policies to be drafted in FY 1997.

Figure 2.0-2. Standard Inventory Maintenance Process.



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Figure 2.0-3. Standard Inventory Change Control Operation.



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3.0 DATA REQUIREMENTS

The primary objective of the Standard Inventory task is to provide reliable waste inventory data that will benefit several safety, waste management, and disposal related activities. Examples of such activities include the following:

- Performance of safety analyses of tank farm operations and of tank wastes
- Performance of risk assessments associated with storage and/or processing of tank wastes
- Design, development, and implementation of waste retrieval, treatment, and disposal systems
- Conduct of performance assessments to evaluate long-term potential doses and risks from waste form disposal systems
- Obtaining regulatory permits.

To ensure that this task encompassed data needs for all the above-listed activities, an initial effort in October 1995 involved reviewing existing Data Quality Objective (DQO) documents and contacting cognizant personnel to identify specific data requirements. The results from this effort are contained in Appendix A, Summary of Data Requirements.

3.1 STANDARD INVENTORY CONTENT LIMITATIONS

The data requirements defined in Appendix A are quite extensive and, since available resources were limited, it was necessary to focus this task on a subset of the Appendix A data requirements. A discussion of the content limitations and the resulting set of chemicals and radionuclides addressed by the Standard Inventory task is provided in Appendix I. This subset of chemicals and radionuclides was reviewed with data users during the Standard Inventory User and Policy Workshop conducted in July 1996 (LeClair 1996).

3.2 EXPANSION OF THE STANDARD INVENTORY CONTENT

As originally conceived, the Standard Inventory would be the inventory data set having most value to the largest number of users. As a second objective, the Standard Inventory must also be updated and modified on a regular basis so that it reflects the most current information available on tank contents. This second objective imposes some limit on the size of the Standard Inventory data set, therefore, it is recognized that it will not meet every need of all users.

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As currently defined, the standard inventories are single values per analyte for the whole of the 177 tanks (global inventory) and for the whole of each tank (tank-by-tank inventory). The need to include some measure of precision (e.g., confidence interval, error bars, uncertainty estimates, etc.) on each of these analyte inventories has already been recognized and is work planned for FY 1998.

Several other user needs have been identified over the course of the Standard Inventory task including reporting of chemical compound inventories, reporting of the soluble and insoluble fractions of the analyte waste inventory by tank, and reporting of the analyte inventories by liquid and solid phase for each tank. Some of these program-specific needs can often be extracted from the data presented in the best-basis documentation or within the sample analytical data reported in the TCR for each tank. As these additional program needs are identified, it is anticipated that the Standard Inventory will be expanded to report the needed data within the TCD, but such data may not be included in the best-basis inventory discussion of each TCR.

4.0 PROCESS-RELATED BASES AFFECTING GLOBAL INVENTORY ESTIMATES

The global waste inventory represents the sum of all chemical and radiochemical components, e.g., metric tons (MT) of sodium, nitrate, aluminum, etc., and curies (Ci) of $^{239/240}\text{Pu}$, ^{137}Cs , ^{90}Sr , etc., presently stored in SSTs and DSTs. Because of inconsistencies among reported component inventory values, a major objective of the present task is to resolve these inconsistencies and to establish standard inventory values for all waste management and disposal activities.

Evaluations performed to resolve differences among reported values involve reviewing the bases, i.e., assumptions, data, background information, etc., that support these values. These bases are highly dependent on the different separations processes that generated the waste inventories as well as the types and amounts of fuels processed. In addition, changes/adjustments to process flowsheets can have major impacts on tank waste inventories. Waste inventories are also impacted by various process losses and other losses/adjustments that resulted from tank leaks, offsite shipments, transfer of supernatants from tank farms to cribs, etc. Figure 4.0-1 provides an overview of the major system components that impact tank inventories.

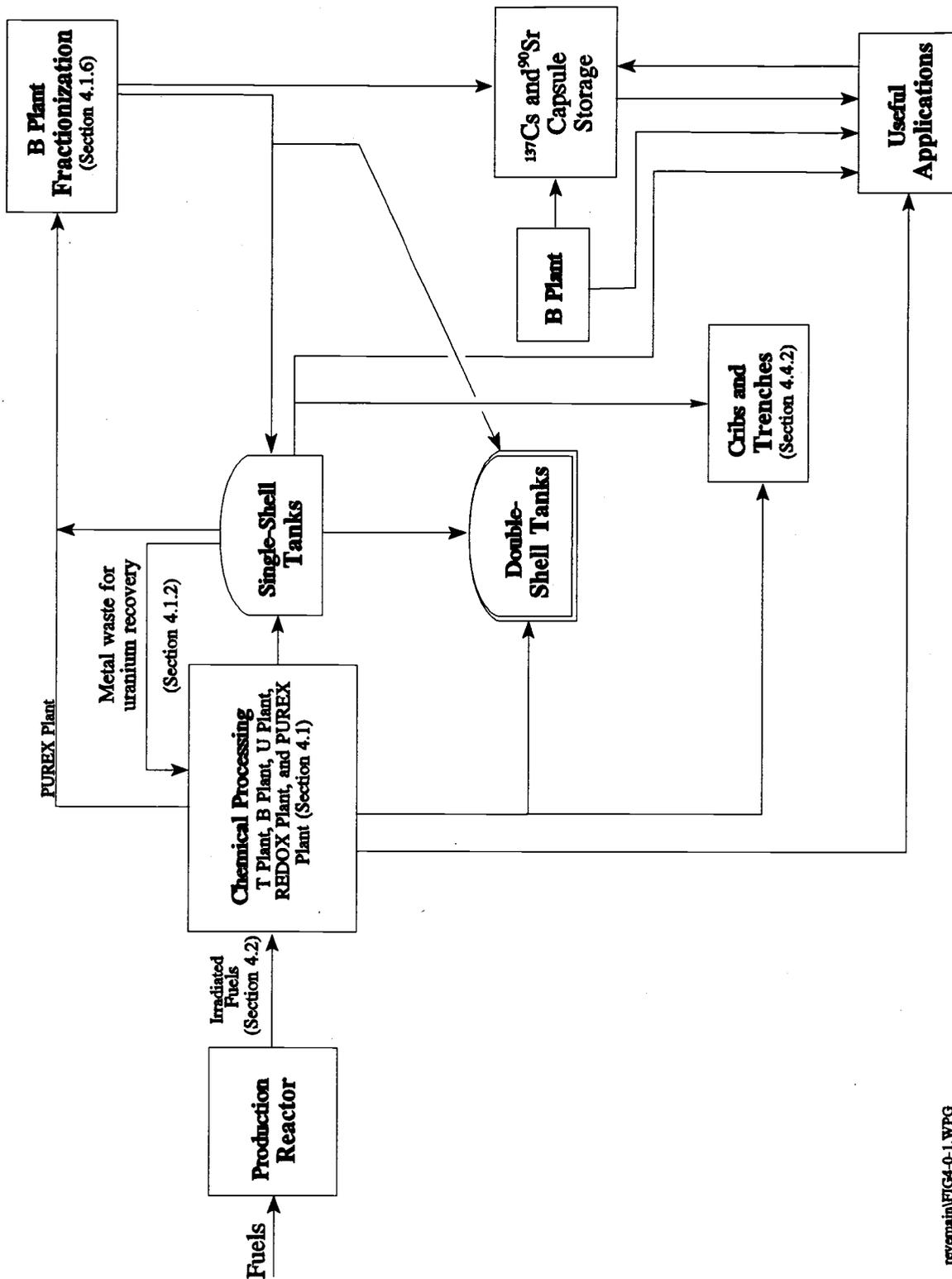
These major system components are discussed further in Sections 4.1 through 4.4. A brief description of the major processes and associated wastes is provided in Section 4.1. An overview of the types and amount of fuel processed is presented in Section 4.2. The major caveats and limitations that arise from using chemical values from flowsheets alone to estimate inventories are addressed in Section 4.3. Section 4.4 summarizes the mechanisms for system losses.

4.1 PROCESS DESCRIPTIONS AND WASTE GENERATION

The majority of wastes stored in underground waste tanks at the Hanford Site originated from processing irradiated uranium fuel and subsequent waste treatment schemes. The primary chemical processes employed to process irradiated fuels were: the BiPO_4 process, the tri-butyl phosphate (TBP) uranium recovery (UR) process, the reduction and oxidation (REDOX) process, the plutonium-uranium extraction (PUREX) process, and the B Plant waste fractionization process. Smaller volumes of wastes originated from research and development programs, laboratory processes, and Plutonium Finishing Plant (PFP) operations, etc.

Waste volumes were reduced in earlier years by decanting selected dilute wastes to the ground and in later years by evaporation/concentration methods. Additional postprocessing of some of the wastes to recover uranium, or to reduce the volume of high-level waste (HLW), resulted in the addition of ferrocyanide and other compounds to the tanks. Following are brief descriptions of these processes.

Figure 4.0-1. Major System Components That Impact Tank Inventories.



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4.1.1 Bismuth Phosphate (BiPO_4) Process

The first production process in the world to separate plutonium from irradiated uranium nuclear reactor fuel was the BiPO_4 process initially operated in the Manhattan Project T and B Plants at the Hanford Site. The process was based on the characteristic that in one valence state (+4), plutonium phosphate (and plutonium fluoride) is essentially insoluble and could be co-precipitated with a BiPO_4 (or lanthanum fluoride) carrier, while a different valence state, plutonium (+6) phosphate (and plutonium fluoride) is very soluble. The plutonium valence could be adjusted with appropriate oxidizing and reducing agents and thus the plutonium could be separated from the bulk uranium and the various fission products. No attempt was made to recover the uranium in this process. This BiPO_4 process operated between 1944 and 1956 in the 221-T and 224-T Buildings (T Plant) and between 1945 and 1952 in the 221-B and 224-B Buildings (B Plant).

As a result of this process, the following waste streams were sent to underground storage tanks for storage or routed through the tanks before being transferred to cribs for ground disposal: coating (cladding) waste (CW) containing the dissolved aluminum cladding, metal waste (MW) containing the uranium and about 90 percent of the fission products, first decontamination cycle (1C) waste containing about 90 percent of the remaining fission products, second decontamination cycle (2C) waste containing essentially all of the remaining fission products, and final plutonium purification/concentration facility (224) wastes that contained low concentration transuranic (TRU) wastes.

The BiPO_4 wastes were generally routed to underground tank "cascades." These were a series (usually three) of connected tanks that overflowed one to another. The wastes were always neutralized to an alkaline pH (>7) to protect the carbon steel tank liner from corrosion, and at this pH the dissolved metal salts in the waste precipitated and settled, primarily as hydroxides or hydrated oxides. Thus, the first tank in the cascade collected the bulk of the solids (including the bulk of the fission products). The cascade settling process, therefore, had the effect of clarifying the supernatant from the chemicals and radionuclides in the inlet slurry, and the top supernatant from the last tank in the cascade was much lower in activity than the inlet waste. The underground waste tanks (nominally 22.9 m [75 ft] in diameter) originally had no provisions for mixing, and solids, once settled, tended to remain in the tanks unless the solids built up to an excessively high level. (The overflow lines between the first and second tanks in many cascades eventually plugged due to solids carryover and crystallization in the lines.) The original T Plant tank farms were 241-T and 241-U Tank Farms, followed by 241-TX and 241-TY Tank Farms. The original B Plant tank farms were 241-B and 241-C Tank Farms, followed by 241-BX and 241-BY Tank Farms.

The CW and 1C wastes were combined in the 1C waste cascades. (When ferrocyanide scavenging of the T Plant 1C waste was started in late 1954, these two wastes were separated since the high pH CW adversely impacted the scavenging.) The MW was later removed from the MW cascades for UR (see Section 4.1.2). The 224 wastes were initially considered "disposed" rather than stored, since they were initially routed to an unlined concrete tank

(361 tank) with the clarified overflow routed to ground by a dry well. Low-activity cell drainage (known as 5-6 waste), e.g., canyon deck flushes, cell flushes, process or utility line leakage, etc., was also routed to this system. Higher activity cell drainage was routed to the 1C cascades. These 224 Building concrete waste tanks became filled with solids and concerns for the possibility of contaminating the site groundwater with plutonium led to replacement of the concrete tank and dry well disposal by routing the 224 wastes to the 200 series (6.1-m [20-ft] diameter carbon steel lined tanks) in 241-T and 241-B Tank Farms by 1946/1947. (The tank overflows were routed to new cribs.) The supernatant from the last tank in the 2C waste cascades was found to be of low enough activity (according to disposal standards of that time) that beginning in 1947 (T Plant) and 1948 (B Plant) the overflow from these cascades was routed to cribs.

By the late 1940's, the 1C waste was the only BiPO_4 waste not being disposed to ground (2C and 224) or scheduled for recovery (MW). Because of cascading, the 1C, 2C, and 224 waste solids, including the great bulk of the fission products and the TRU elements settled and remained in the waste tanks. Atmospheric pressure evaporators (224 T/B) were constructed (1951) in both the 200 West and 200 East areas to concentrate the waste. The condensate from the evaporator was disposed to ground while the concentrate was sent back to the tank farms. Some 1C waste and concentrated 1C waste contained in tanks were also disposed to ground on a specific retention basis to free up badly needed tank storage space in the early 1950's. The 242-B evaporator ran from 1951 to 1954. The 242-T evaporator ran from 1951 to 1955. It was reactivated in 1965 and ran again into 1976.

4.1.2 Tri-Butyl Phosphate Uranium Recovery Process

As noted previously, the BiPO_4 process did not recover the uranium in the irradiated reactor fuel. The uranium, along with approximately 90 percent of the fission products, was routed to the MW tank cascades in the 241-B, 241-C, 241-BX and 241-BY tank farms (for B Plant), and the 241-T, 241-U, and 241-TX farms (for T Plant). The third BiPO_4 plant constructed under the Manhattan Project had not been used, and this facility (U Plant) was modified to perform as a uranium recovery plant during the period 1952 to 1957.

Process vaults containing stainless steel vessels were constructed near each tank farm, and sluice pump pits were added to each MW tank. The tanks containing MW were then sluiced with their own supernatant to the process vaults. There the solids, primarily uranium, were dissolved in nitric acid and the resultant solution/slurry was pumped to U Plant. U Plant contained a feed clarification system (via centrifuges) and a feed adjustment system. The adjusted feed was then routed to one of two parallel solvent extraction lines. Each of these lines contained a feed tank, two mechanically pulsed solvent extraction columns for extracting/scrubbing and stripping the uranium, and a solvent treatment system to clean and recycle the solvent. The recovered and purified uranium nitrate solution was concentrated and then denitrated to UO_3 for offsite shipment.

The U Plant pulse columns were the first production use of this technology for radiochemical separations, and served as a test bed for the later PUREX Plant pulse columns. The solvent extraction lines used an organic phase of TBP in a hydrocarbon diluent as the extractant. One line operated with a TBP concentration of 12.5 vol% and the other utilized a 30 vol% TBP solvent phase to compare the operating characteristics of these two systems. The plutonium valence in the solvent extraction feed stream was adjusted so that any plutonium present would report back to the waste stream for return to the underground storage tanks. This was done because the U Plant was not equipped to recover plutonium, and plutonium limits in the uranium product were very low.

The uranium recovery (UR) waste (also called TBP waste) routed to the storage tanks consisted of the solvent extraction waste (which could be concentrated in U Plant under certain conditions), combined with the solvent wash waste. The solvent cleanup wash was initially a sodium sulfate solution, but was later replaced by a sodium carbonate solution. The wastes were neutralized to a pH greater than 7 before transfer to the tank farms. This UR waste therefore contained all of the components in the MW (but without the carbonate and with only 1 to 2 percent of the uranium). Further changes in the waste stream consisted of addition primarily of nitrate (added as nitric acid), iron (added as a plutonium reductant), sulfate (from the reductant and sulfate wash) and sodium (from sodium carbonate washes and caustic [sodium hydroxide] neutralization). Additional changes to the UR waste stream were affected by ferrocyanide scavenging. (See following section.)

4.1.3 Scavenging Program to Remove Cesium-137 and Strontium-90 From Aqueous Wastes

Ferrocyanide waste scavenging processes were developed at the Hanford Site in the 1950's to provide additional waste storage space while minimizing the construction of additional waste storage tanks. These processes were designed to remove the soluble long-lived fission product ^{137}Cs from liquid wastes. In some cases, where the concentrations of ^{90}Sr in neutralized waste supernatants remained high, supplemental scavenging with calcium was also performed to more fully precipitate the ^{90}Sr . As a result, supernatants could be disposed to the ground, and the volume of waste requiring storage in tanks was reduced.

Three scavenging processes were used to treat the following three different types of wastes: UR waste from U Plant, 1C waste from T Plant, and in-farm UR wastes (wastes already stored in tanks). In general, aqueous waste was pumped to large process tanks where the waste pH was adjusted and the precipitating agents (initially, potassium ferrocyanide and nickel sulfate; later, sodium ferrocyanide and nickel sulfate) were added. If supernatants contained significant ^{90}Sr , calcium nitrate was added to precipitate calcium phosphate. In some campaigns, nonradioactive strontium nitrate was added rather than calcium. The treated waste was stirred to ensure thorough mixing and then pumped to receiver tanks in the tank farms. The precipitate containing the scavenged ^{137}Cs and ^{90}Sr was allowed to settle and the supernatant was discharged to cribs. When layers of the

ferrocyanide sludge built up in the receiver tanks, they were pumped to other tanks for storage so that the settling depth in the receiver tanks remained maximized.

Scavenging of U Plant waste began with a plant test in late 1953 during which the treated waste was routed to tank 241-T-101. Production-scale scavenging began in September 1954. Scavenged UR waste was routed to settling tank(s) in the 241-BY Tank Farm in the 200 East Area (usually tanks 241-BY-106, 241-BY-107, 241-BY-108, or 241-BY-110). The accumulated sludge in the settling tanks was periodically transferred to tank 241-BY-104 or 241-BY-105. The last scavenged waste from U Plant was received in the 241-BY Tank Farm in June 1957.

Routine scavenging of T Plant 1C wastes was started in December 1954 and continued until the plant was shut down in March 1956. Treated 1C waste was routed to tanks 241-TY-101, 241-TY-103, and 241-TY-104. Beginning in May 1955, unscavenged UR waste already stored in 200 East Area tanks was routed to the 244-CR Vault for in-farm scavenging. The scavenged waste was then routed back to other waste storage tanks for settling, sampling, and decanting to cribs. The primary settling tanks for in-farm scavenged waste were tanks 241-C-108, 241-C-109, 241-C-111, and 241-C-112. In-farm scavenging was completed in December 1957.

4.1.4 Reduction Oxidation (REDOX) Process

The REDOX process was chosen to replace the BiPO_4 process in order to lower costs, improve throughput, recover uranium, and improve plutonium recovery to greater than 98 percent. This solvent extraction process operated between 1952 and 1966 in the 202-S Building (S Plant).

Irradiated fuel elements were deacid with caustic and dissolved in nitric acid as was done in the BiPO_4 plants. The dissolver product was then adjusted in acid concentration and plutonium valence for solvent extraction. The extraction solvent was Hexone¹. Aluminum nitrate was added as a source of nitrate ion to drive ("salt") the uranium and plutonium into the solvent phase. Nitric acid could not be used as the nitrate ion source since Hexone reacts violently with nitric acid in even moderate concentrations. The plutonium could then be removed from the solvent by adjusting the valence with a reducing agent, and the uranium could be stripped back into an aqueous stream by adjusting the flow ratio of the aqueous to solvent phases. The solvent extraction operations were performed in packed columns. The REDOX process contained many solvent extraction cycles (i.e., the uranium and/or plutonium was extracted from an aqueous solution into a solvent phase and stripped back into an aqueous solution several times to purify and concentrate these products).

¹Hexone is a tradename of the Union Carbide Company, Danbury, Connecticut, for methyl isobutyl ketone.

Under the original REDOX flowsheet, each of the solvent extraction cycles had a waste stream that was routed to the tank farms for storage (after concentration in evaporators in some cases) and combined with the cladding waste as "salt waste" in the 241-S and 241-SX tank farms. As the REDOX process matured, several of the intermediate cycle wastes were recycled, thereby reducing the plant chemical consumption and increasing the fission product concentration in the wastes. The fission product concentration (and the associated amount of decay heat) eventually became high enough that the wastes "self-boiled." This reduced the waste tank volume required for storage, but necessitated that the waste tank design incorporate features to promote controlled boiling in the tanks. In the mid-1950s, the cladding waste was routed to storage in separate tanks from the solvent extraction wastes, and a route was installed to utilize the 241-U tank farm for storage of some REDOX wastes.

The REDOX Plant processed all the Hanford Site irradiated enriched uranium fuel from 1958 until shutdown in 1966, and was the first plant to process Zircaloy¹-clad fuel using the Zirflex chemical decladding process. Small amounts of other irradiated fuels, e.g., from the Plutonium Recycle Test Reactor (PRTR) and Shippingport reactors, were also processed in REDOX (see Appendix B).

4.1.5 Plutonium-Uranium Extraction (PUREX) Process

The PUREX solvent extraction process operated between 1956 and 1972 in the 202-A Building (PUREX Plant). Operations resumed in 1983, but the facility again shut down in 1990. Aluminum-clad fuels were processed at the PUREX Plant from 1956 to 1972. Some Zircaloy[®]-clad fuels were processed between 1967 and 1972 and only Zircaloy[®] fuels were processed from 1983 to 1989.

The PUREX process campaigns, and subsequent campaigns at B Plant, to remove fission products from PUREX process waste, produced some of the most complicated combination of wastes produced at the Hanford Site. This solvent extraction process involved contacting an organic phase mixture of 30 vol% TBP in normal paraffin hydrocarbon (NPH) with the nitric acid dissolved solution of plutonium and uranium. This process was much more efficient than the REDOX process. The nitric acid used as a salting agent at the PUREX Plant could be partially distilled and reused, unlike the aluminum nitrate at the REDOX Plant. Extraction operations were conducted in mechanically pulsed columns, rather than packed columns as at the REDOX Plant.

The PUREX Plant processed approximately 72 percent of the irradiated fuel processed at the Hanford site (see Appendix B). This included aluminum clad uranium fuel, both natural and enriched, aluminum clad thoria fuel (to recover ²³³U), and Zircaloy[®]-clad N Reactor fuel. The PUREX Plant wastes included both "boiling," i.e., high-level solvent extraction wastes, and "non-boiling" wastes, i.e., the cladding wastes, organic wash wastes

¹Zircaloy and Zircaloy-2 are tradenames of Teledyne Wah Chang, Albany, Oregon.

and cell drainage. The HLW was routed to the 241-A, 241-AX SST farms and later to DST farms. The sodium carbonate organic wash wastes were originally routed to the HLW tanks, but later were routed to non-boiling waste in the 241-C farm for storage, transfer to other tank farms, and subsequent in-farm concentration along with the cladding wastes and cell drainage. All of the thoria wastes produced in the 1966 and 1971 processing campaigns were routed to non-boiling waste storage in 241-C farm. Zirflex decladding wastes were routed to the 241-AW DST farm after process waste additions to the SST farms were discontinued in 1980.

4.1.6 B Plant Waste Fractionization

By the mid to late 1950s, there was a program underway to develop methods to remove the ^{90}Sr and ^{137}Cs from the Hanford HLW for two reasons: (1) the ^{90}Sr was in demand as a relatively long-lived heat source for thermoelectric generators, and the ^{137}Cs as a gamma source; and (2) the first waste storage tank failures (leaks) were seen and it was desirable to make the stored waste less mobile and thus minimize the volume of any future leaks. In this regard, if these two radioisotopes, each with a half-life of approximately 30 years, could be removed from the waste, the remaining waste could be solidified, or at least concentrated and dewatered with minimal concern for the remaining fission product heat in the waste as long as the waste was at least one to two years out of the reactor. Therefore, laboratory development work on processes to recover these two isotopes from the waste was initiated. This work resulted in a strontium removal precipitation process in the PUREX Plant, and the piloting of solvent extraction processes for strontium recovery and purification in the strontium semi-works in the 200 East Area.

The "laid away" BiPO_4 plant in the 200 East Area (B Plant) was reactivated and modified with equipment to "fractionate" the waste, i.e., remove the strontium and cesium and route/return the remaining waste fraction to the waste storage tanks. The B Plant facility and the waste treatment processes are described in detail in the *Waste Management Technical Manual* (Buckingham et al. 1967). B Plant began removal of ^{137}Cs from stored PUREX waste supernatants (PSN) in late 1967 using an ion exchange system. The REDOX waste supernatants (RSN) were processed for ^{137}Cs removal via ion exchange starting in 1970. The 244-AR Vault was constructed with stainless steel interim waste storage tanks so that the waste currently being produced at PUREX could be stored and processed for ^{90}Sr removal in B Plant without having to neutralize and reacidify the wastes. The first current acid waste (CAW) from PUREX was processed for strontium recovery in B Plant in April 1968. A process for the removal of ^{137}Cs from CAW with a phosphotungstic acid precipitation was developed and first used in B Plant in April 1969. The ^{90}Sr in the neutralized and stored PUREX HLW (i.e., those wastes produced before 1968) was present in the settled sludge solids in the 241-A and 241-AX farm SSTs. These sludges were sluiced to the 244-AR vault, washed, and dissolved in nitric acid. The resultant slurry (PUREX Acidified Sludge = PAS) was settled, and the supernatant was transferred to B Plant where the ^{90}Sr was removed via a lead sulfate carrier precipitation process. Processing of the first PAS in B Plant began in late November 1968.

A solvent extraction process was used in B Plant to recover, concentrate and purify the ^{90}Sr and rare earths from the CAW and PAS feeds. This process used Di(2-ethylhexyl)phosphoric acid (D2EHPA) and TBP as the extractant in an NPH diluent. The process was pH sensitive, requiring a buffering agent. The presence of relatively high concentrations of various metals, e.g., Fe and Al, in the waste feed necessitated the use of selected chelating agents to prevent the precipitation and/or extraction of these metals. These chelating agents were water soluble organics such as ethylenediamine tetra acetic acid (EDTA), N(2-hydroxyethyl)ethylenediamine tetra acetate (HEDTA), or citrate. These organics reported to the solvent extraction system wastes, and were the source of the bulk of the organics now found in the Hanford site waste tanks.

The ^{90}Sr , ^{137}Cs and rare earths recovered and stored at B Plant could be used for beneficial purposes in addition to their desired removal to reduce the stored waste heat load. For instance, both ^{90}Sr and ^{137}Cs were shipped to Oak Ridge National Laboratory (ORNL), rare earth fractions were shipped to Pacific Northwest National Laboratory (PNNL), and rhodium-palladium was shipped to Pittsburgh Plate Glass Industries. The bulk of the ^{90}Sr and ^{137}Cs was converted to fluoride and chloride salts respectively, and encapsulated and stored in the Waste Encapsulation and Storage Facility (WESF) constructed at the west end of B Plant.

The B Plant wastes were neutralized and returned to the waste tank farms. The heat load in the waste determined which tank(s) it was routed to. If the heat load was low, e.g., ion exchange wastes (which had been aged before processing) or solvent washes, the waste was routed to tanks that could be used as feed for the In-Tank Solidification (ITS) or Evaporation processes. If the waste had a high-heat load, e.g., the solvent extraction HLW, it was routed to tanks that could store boiling wastes for aging before further processing.

Cesium recovery operations in B Plant were completed in September 1983, and strontium recovery operations completed in February 1985. Waste Fractionation was not performed on the PUREX wastes produced when PUREX was restarted in 1983 to process N Reactor Zircaloy®-clad fuels.

4.1.7 Evaporation of Waste Supernatants and the Formation of Salt Cake

There have been a variety of waste management operations employed at Hanford. There were several efforts to remove fission products from the waste to reduce waste activity. Evaporation was routinely used to reduce overall waste volume. These processes are somewhat related. Each separation process used different solvents and processing chemicals, providing for variations in the evaporated concentrate composition. Additionally, different methods of evaporation were used providing additional variability to their composition. Salt cakes are the byproduct of various waste reduction efforts using evaporation. Salt cakes and evaporator concentrates comprise over 66 percent of the waste volume at Hanford.

One of the early waste management practices in the tank farms was waste segregation. The waste streams from different stages of the separation processes were rarely mixed. Raw waste streams from different separation processes were almost never mixed. The waste streams from these processes usually had two fractions, liquid and solid. The majority of the process wastes generated were liquid, with modest amounts of soluble species dissolved in the solvent stream, carrying the solid fraction as a slurry. The soluble species of note are sodium, aluminum, nitrate, nitrite, hydroxide, sulfate, phosphate, and carbonate.

After the waste stream was initially disposed in the tanks, the entrained solids dropped out of the waste stream and settled on the bottom of the tank. The liquid remained above the solids. Wastes were separated by type and disposed in a particular cascade. Initially, these supernatant wastes from different tanks were not often mixed before evaporation leading to the compositional variability attributable from the different process sources. Over time, to conserve tank space, waste blending was much more prevalent. Therefore, in many cases, no particular analyte, process source, or characteristic can be attributed to the source waste streams

Salt cakes are usually distinguishable from sludges physically and compositionally. Physically salt cakes are lighter in color (white, yellow or light grey is typical) than sludges, have a visually observable crystal structure, are granular in texture, and are usually very soluble in water. Compositionally, they are mostly made up of the sodium salts of nitrate, nitrite, phosphate, carbonate, and hydroxide. Occasionally aluminum salts are observed to be part of a salt cake. Transition metals, such as iron, manganese, and lanthanum, and heavy metals such as lead and uranium, are usually absent from salt cakes. Exceptions have been noted where "dirty" salt cakes have been obtained. In these cases it appears that sludge material had been entrained in the salt cake.

Different evaporation processes also substantially influenced the composition of the salt cakes. There were four different waste volume reduction methods that relied on evaporation: the open-air evaporators, 242-B and 242-T; the in-tank solidification tanks (BY farm); the self-boiling tanks (S, SX, A, and AX Farms), and the vacuum evaporators, 242-S and 242-A. Differences in evaporative processing may be observable and have an impact on eventual retrieval and disposal of the waste.

The earliest salt cakes were formed in 242-B and 242-T evaporators. Their process feeds consisted mostly of bismuth phosphate process supernatants, and later uranium recovery supernatants. These were open-air evaporators that were operated at relatively high temperatures. This may have contributed to the loss of volatile organic components and the formation of carbonate. Their salt cakes may resemble the solid fraction of the wastes that were originally concentrated (e.g. first cycle or uranium recovery waste).

Self-boiling was initially observed in REDOX process waste in S and SX tank farms. Later, it was also observed in PUREX process waste in A and AX tank farms. This characteristic was turned to use as a waste management technique. This process was also open-air evaporation; however, these wastes were much more alkaline than those from the

bismuth phosphate process. This feature of the waste probably contributed to much higher carbonate formation.

The in-tank solidification (ITS) process used an in-tank heater as an open-air evaporator, with a series of tanks connected together in a cooling loop. This promoted salt cake formation in the cooling tanks. However, because of the different equilibrium conditions between the boiling tank and the tanks in the cooling loop, the composition of the salt cake formed in the boiling tanks is probably much different than the composition in the downstream tanks. The ITS process used a separate tank as a feed staging tank. The feed tank was used to collect and blend supernatants from various tanks and processes before introducing them to the evaporator tank. There were two principal ITS heater tanks, tanks 241-BY-102 and 241-BY-112. For a short time, tank 241-BY-101 was used as a prototype tank.

The 242-S and 242-A evaporator/crystallizers were different than the open-air evaporators used in earlier waste volume reduction efforts. They operated under a partial vacuum, reducing the amount of heat needed to boil the supernatants. Operation under a partial vacuum also reduced the interaction with the air, inhibiting carbonate formation. Blending waste feeds is frequently observed in the process records of earlier 242-S evaporator operations. However, more recent 242-A evaporator operations appear to be more segregated, with the contents of a single tank being processed at any particular time. This difference in processing may be observable in sampling data.

4.1.8 Other Processes

A TBP solvent extraction process at the PFP (Z Plant) was used to further purify plutonium product from the processing plants and from plutonium scrap. Plutonium nitrate was converted to plutonium oxide and then smelted to metal in a remote mechanical processing line.

Processing at PFP yielded wastes that contained low concentrations of plutonium and other transuranic elements, but contained high concentrations of metallic nitrates. The plant's waste streams are identified as follows: dilute noncomplexed waste, HLW, high-transuranic sludge, lab waste, transuranic solids, noncomplexed waste, remote mechanical line transuranic solids, and remote mechanical line dilute noncomplexed waste. Before May 1973, waste from PFP was sent to cribs rather than to tank farms.

A relatively small amount of waste was generated at a separations process and equipment development pilot plant (known as the "hot semi-works;" later as the "strontium semi-works"). The plant was retired in 1967. The waste streams that originated from this facility (hot semi-works, strontium semi-works, and fission products wastes) are characteristically high in ⁹⁰Sr content.

4.2 OVERVIEW OF FUEL PROCESSED

Since startup of B Reactor on September 26, 1944, a number of changes occurred in both fuel fabrication and irradiated fuel processing. These changes are noted in this section. A summary of fuel processed at each separations plant is presented first followed by discussions of aluminum and zirconium-clad fuel elements.

4.2.1 Separations Plant Fuel Processing History

Approximately 99,000 metric tons of uranium (MTU) were processed in various separation facilities at the Hanford Site (Appendix B). This value includes the fuel processed in the PUREX Plant between 1983 and 1989 with the Zirflex decladding process. Fuel elements were clad in either aluminum or zirconium to protect the uranium from corrosion. Zirconium-clad fuel was used in N Reactor exclusively, whereas aluminum-clad fuel was used in all the older single-pass reactors. In addition to processing of irradiated uranium, the PUREX Plant was also used to process irradiated thoria (ThO_2). Two thoria processing campaigns were run, one in 1966 and one in 1970. A total of 629 MT of aluminum-clad thoria were processed.

A summary of fuel processed at the various separation plants appears in Table 4.2-1, and the detailed separations plant fuel processing history reconstructed for this report is contained in Appendix B.

Table 4.2-1. Separations Plant Fuel Processing Summary.

Separations plant (operating campaign)	Aluminum-clad fuel		Zirconium-clad fuel
	Natural uranium (MT)	Enriched uranium (MT)	Uranium (MT)
T Plant (1944 to 1956)	5,034	0	0
B Plant (1945 to 1952)	2,766	0	0
REDOX (1952 to 1966)	11,609	7,852	245
PUREX (1956 to 1972)	58,748	7,176	1,572.3
PUREX (1983 to 1989)	0	0	3,890.4
Thoria (PUREX 1966 and 1970)	629 ThO_2	--	--

PUREX = Plutonium-uranium extraction
REDOX = Reduction and oxidation.

4.2.2 Aluminum-Clad Fuel

All of the Hanford Site reactors are single-pass reactors, except for N Reactor. The single-pass reactors used aluminum-clad fuel. Cladding of the fuel was necessary as corrosion studies in the fall of 1942 showed that water would react with the uranium at the reactor operating temperatures (approximately 80 °C) (Burley 1958).

4.2.2.1 Fabrication. Aluminum-clad fuel consisted of a uranium metal core surrounded by an aluminum jacket or can. An aluminum-silicon (Al-Si) alloy braze between the uranium and aluminum components provided for high thermal conduction and a strong metallurgical bond between the components. Fuel fabrication methods changed over the years to reduce costs, and in response to increasingly severe reactor conditions.

Triple Dip Fuel. The first production fuel elements fabricated in quantity were manufactured by the triple dip process. Uranium pieces were first dipped in bronze, then dipped in molten tin to remove excess bronze. The bronze alloy contained 47 wt% copper and 53 wt% tin (Burley 1958). Excess tin was removed by centrifuging the uranium cores before assembling the cores and aluminum alloy jackets in a molten Al-Si alloy bath. The triple dip process introduced tin, silicon, aluminum, and a trace of copper into the waste.

Triple dip fabrication was used from 1944 until March 1954 (Weakley 1958). A basis for estimating when the last triple dip fuel went through the separation facilities has been developed. The approach is based on an assumed fuel fabrication lead time, reported reactor residence times, and reported fuel cooling times. An estimated total of 10,800 MTU of uranium as triple dip aluminum-clad fuel were processed based on an assumed fuel fabrication lead time of 90 days, the reactor residence days (Jaech 1957), and fuel cooling data reported in Roberts et al. (1992). Based on these data and the 90 day assumption, T Plant changed to lead dip fuel about January 1, 1955. The REDOX Plant continued to work off triple dip fuel into the month of July 1955, except for running lead dip fuel during the months of February and March 1955. All of the B Plant production, from start-up in 1945 to shutdown in 1952, was with triple dip fuel. Fuel elements during this time period consisted of solid uranium pieces clad in aluminum with flat aluminum end caps.

Lead Dip Fuel. Lead dipped aluminum-clad fuel was fabricated from March 1954 until the last single-pass reactor shut down in 1971. The lead dip process introduced silicon, aluminum and lead into the fuel processing wastes. The lead dip process left a layer of lead and U-Pb compounds between the uranium core and the Al-Si bond layer. The lead layer was about 0.0025-cm thick (Kahle and Bement 1958). Approximately 82,400 MTU of lead dip aluminum-clad fuel was processed.

Aluminum Introduced by Fuel Fabrication. Aluminum-clad fuel introduced aluminum into fuel reprocessing waste from two sources: aluminum jackets and Al-Si bond layer. Natural uranium slugs were fabricated as 8 Inch and 4 Inch elements. Enriched uranium was fabricated as 6 Inch elements. (The overall lengths of the 4 Inch, 6 Inch, and 8 Inch elements were 11.85 cm, 16.87 cm, and 22.77 cm.¹) Fuel elements were fabricated in both solid, and internal and external (I&E) configurations. (See Fuel Design discussion provided later in this section.) Fuel elements were designated as type "O," for use in the older reactors, type "C" for use in C Reactor, and "K" for use in the KE and KW Reactors. The diameters of the fuel elements varied from 3.658 to 3.744 cm due to differences in design and operation of each generation of reactor. In addition, small changes in dimensions were made due to changes in fuel fabrication and to alleviate problems observed during irradiation.

The aluminum from the fuel jacket material and the Al-Si bond layer was estimated from fuel drawings, records of enriched versus natural uranium processed, and assumptions about solid and I&E fuel. (See Fuel Design discussion provided later in this section.) Information on fuel usage by reactor is not readily available. Since a large fraction of the fuel was irradiated in K Reactor, the dimensions of "K" fuel were assumed for all I&E elements. A total of 3,505 MT of aluminum attributable to the fuel cladding was estimated by these assumptions; about 975 MT from the Al-Si bond layer and 2,530 MT from the jacket components. Uncertainty in the use of 4 Inch elements (discussed in a following section) would increase the aluminum in the fuel cladding from 3,505 MT to 3,530 MT.

4.2.2.2 Fuel Design. Increasing power loads on the reactors required changes in the design and dimensions of fuel elements to improve heat transfer. Changes were also made to prevent "cocking" of elements in the reactor process channels. Special elements were also fabricated for production of special isotopes or specific applications in the reactor.

Solid Fuel Elements. The first fuel elements used at the Hanford Site consisted of a solid uranium core clad in aluminum. The end caps for the first solid fuel elements were flat pieces of aluminum. Later solid elements were fabricated with Tru-line design end caps (see Tru-line elements discussion below). Solid fuel elements were replaced to a small extent between 1954 and 1957 by cored fuel elements, then later to a greater extent by the I&E cooled elements.

Cored Fuel Elements. Hollow or cored fuel elements were fabricated from 1954 until January 1957 (Weakley 1958, DeNeal 1970). The hollow core provided space for expansion of the uranium. Less than 15 percent of the production during this time was cored fuel elements (Weakley 1958).

¹4 Inch, 6 Inch, and 8 Inch fuel elements are named for their approximate length. Although our results are reported in metric units, we have chosen (when referring to source documents) to use other units to preserve the traceability, accuracy, and numerical significance of the original or source work.

Internal and External Elements. I&E elements, had an open center for cooling water. The end caps used the Tru-line profile. I&E elements were first loaded into the reactor central zones in September 1957 and phased in for all reactors over 15 months (DeNeal 1970). Given the reactor residence days reported (Jaech 1957), and the cooling days before processing (Roberts et al. 1992), it is unlikely that any significant quantity of I&E fuel would have been processed before 1959. I&E cores reached 85 percent of the fuel fabrication production in October 1958 (Weakley 1958). Solid fuel elements were deleted from drawings of aluminum-clad fuel in September 1962. This is consistent with an assumption in notes to the Allen (1976) report that only I&E fuel was run in the PUREX Plant after 1964.

Tru-Line Elements. As reactor temperatures increased, fuel failures were observed that were caused by localized losses of cooling from the cocking of fuel elements in the reactor fuel channels. Tru-line elements, used contoured end caps to align fuel elements in the reactor process channels. One end cap of the Tru-line fuel element was slightly raised and the other end cap was slightly recessed (3.5 mm) to obtain self-alignment of the fuel. Use of Tru-line elements in the reactors began in September 1957 (DeNeal 1970). Tru-line fuel elements were fabricated in both solid and I&E designs. Fuel with flat end caps was deleted from drawings of aluminum-clad fuel in 1960 and, thus, is assumed to have been out of production as of that date.

Watermix Fuel Elements. A watermix fuel element consisted of a watermix spool piece and a 6 Inch natural uranium I&E fuel element welded together. The watermix spool was a 5.08-cm long aluminum fabrication designed to mix the reactor cooling water from the internal and external passages (Weakley 1958). DeNeal (1970) states that "The first shipment of watermix elements was received for use in the 17th fuel element position from the downstream end." on August 11, 1958, at the KE and KW reactors. The quantity of watermix fuel elements has not been determined, however, it is thought that watermix fuel elements do not represent a significant percentage of the fuel processed.

Fuel Elements, 4 Inch. Fuel fabrication reports state that the uranium core lengths for standard aluminum-clad fuel elements was nominally 8 Inch for natural uranium fuel elements and 6 Inch for enriched fuel elements. The 4 Inch fuel elements were a special purpose element fabricated to allow continued operation of reactors with graphite growth problems. Swelling of the graphite resulted in bowing of the reactor fuel process tubes and binding of the fuel. This effect was most pronounced in the upper sections of the reactor. Shorter, 4 Inch solid fuel elements were made to allow charging of distorted reactor fuel process tubes (DeNeal 1970).

Swelling of the graphite in the single-pass reactors first became apparent in 1945. The 4 Inch fuel elements were first fabricated in 1947. Conclusive evidence of the mechanism for graphite swelling was established in January 1948. Use of a carbon dioxide atmosphere to purge air from the reactors and minimize graphite swelling was immediately implemented, beginning with D reactor (DeNeal 1970).

The extent of 4 Inch fuel usage has not been established, but is thought to be small since only B, D, and F Reactors were in operation before the mechanism for graphite swelling was established and because 4 Inch fuel elements were only needed in the upper sections of the reactors. All drawings indicate that only natural solid uranium was used in the 4 Inch elements. Weakley (1958) notes that very few 4 Inch natural solid fuel elements were required at the date of his report. The 4 Inch fuel elements were deleted from drawings of aluminum-clad fuel in 1960. Based on gigawatt days (GWDs), the B, D, and F Reactors production before 1960 accounts for only 8 percent of the total (through 1971) single-pass reactor production. Production from 4 Inch fuel elements would have been a fraction of this amount.

The information extracted on the use of 4 Inch elements from fuel fabrication documents is in disagreement with annual reports written by General Electric in 1953 and 1954. These reports imply that 4 Inch elements were the standard element before 1952. If all the elements used before 1952 were 4 Inch instead of 8 Inch elements, the increase in the aluminum cladding estimate would be less than 1 percent.

4.2.3 Zirconium-Clad Fuel Fabrication

N Reactor, which started up in 1964, used only zirconium-clad fuel. Fuel elements for N Reactor consisted of assemblies of two concentric uranium tubes, each clad in a zirconium alloy. The uranium elements were fabricated by coextrusion into Zircaloy-2[®] cladding. Each assembly was made up of an inner and outer element. N Reactor was used to produce both weapons-grade and fuels-grade plutonium. (Fuels-grade plutonium contained more than 6 wt% ²⁴⁰Pu after irradiation.)

There are two basic forms of fuel elements differentiated by the uranium enrichment. Mark IV fuel elements have a pre-irradiation enrichment of 0.947 wt% ²³⁵U in both tubes and an average uranium weight of 22.5 kg. The Mark IV assemblies have an outside diameter of 6.1 cm and lengths of 44.2 cm, 58.9 cm, 62.5 cm, or 66.3 cm. Mark IA fuel elements have a pre-irradiation enrichment of 1.25 wt% ²³⁵U in the outer tube and 0.947 wt% ²³⁵U in the inner tube. Mark IA fuel elements have an outside diameter of 6.1 cm and lengths of 37.8 cm, 49.8 cm, or 53.1 cm. Mark IA elements are commonly referred to as "spike" fuel (Willis 1995).

In addition to production of plutonium, N Reactor was also used to make tritium. The tritium producing elements, known as co-product fuel, consisted of an inner target element of lithium aluminate and an outer uranium element enriched to 2.10 wt% ²³⁵U. Both elements were clad in Zircaloy[®]. The two elements were mechanically separated before the enriched uranium element was sent to the PUREX Plant.

The N Reactor fuel elements were fabricated by a co-extrusion process. Each uranium tube was encased in an inner and outer sleeve of Zircaloy-2[®] with an outer sheath of copper. This assembly was evacuated and sealed to prevent oxidation during preheating and

extrusion. When the tube assembly was extruded under high pressure and elevated temperature, a solid state diffusion bond formed between the uranium core and the Zircaloy-2® cladding.

The extruded fuel was cut into the desired length and a recess was machined into uranium at each end of the fuel section. This was followed by an acid stripping of the copper sheath and an acid etching process to remove residual uranium from the cladding. The tube ends were closed by placing a braze ring, made of Zircaloy-2® and 5 wt% beryllium, and a Zircaloy-2® end cap in the recess in each end of the fuel and induction heating the assembly to brazing temperature (about 1,050 °C) in a vacuum. The junction of the end cap, braze and cladding was then fusion welded to alloy the braze material with the cladding to improve corrosion resistance and provide a hermetic seal.

4.3 CAVEATS AND LIMITATIONS ON ESTIMATING CHEMICAL WASTES FROM FLOWSHEETS

In many cases, the amounts of process chemicals routed to underground tanks and/or disposal sites can be estimated or verified by flowsheet extrapolation. This involves review of the process flowsheet to determine the amount of chemicals used per unit of irradiated fuel production (usually tons uranium or MTU where ton U = 0.9072 MTU). The flowsheet also gives a unit volume for each waste stream. The flowsheet values are then multiplied by the number of production units in the desired time period. The monthly and annual production for the separations plants at the Hanford Site are listed in Appendix B.

4.3.1 Additional Data Needed to Accurately Extrapolate from Flowsheets

Estimates made from the process knowledge extracted from flowsheets can be quite accurate, and in many cases form the basis for estimates of liquid volumes and chemicals discharged to waste sites. However, there are several things that the estimator must keep in mind when preparing/using such estimates.

Flowsheets were have been found only for the major Hanford Site processes, e.g., BiPO₄, UR, REDOX, etc. Other activities were performed for which flowsheets have not been located or have survived, e.g., Du Pont (1946 p. 178) reports that in April 1946, the MW line from T Plant plugged up. A flush of the line with 9,100 kg (20,000 lb) of 10 percent sodium bicarbonate cleared the line. A program of bimonthly flushing of these lines in both 200 areas was initiated. These chemical additions do not show up on any flowsheet.

Other processes may not be directly tied to production figures, e.g., early neptunium purification in the REDOX and PUREX Plants. Neptunium recovery/accumulation proceeded during normal U-Pu processing, but Np purification processing was performed on a campaign basis between U-Pu production campaigns. Fission product recovery in the PUREX headend facilities also proceeded more or less independently of U-Pu operations. Between 1972 and 1983, PUREX conducted semi-monthly HNO_3 flushes.

Flowsheets were written assuming the plant was at equilibrium on hot (produced from irradiated fuel) feed. In actual practice, the plants, at least REDOX and PUREX Plants, usually started up and shutdown on cold feed (recycled uranium). The cold feed startup was to bring the solvent extraction columns to equilibrium with uranium, thereby minimizing product losses and fission product contamination of the uranium and plutonium. The cold feed shutdown was to push (strip) the fission products and plutonium out of the solvent extraction columns, thereby allowing the columns to be emptied and minimizing radiation damage to the solvent.

There were also other reasons to recycle cold feed, such as rework of out of specification uranium or plutonium, to provide a sink for plutonium storage during maintenance of plutonium processing equipment, etc. A major reason for recycling cold uranium during the N Reactor fuel Zirflex processing at the PUREX Plant from 1983 through 1989, was that the dissolving cycle for this fuel was quite slow. The solvent extraction system processing rate could just barely be slowed to match the dissolution rate, so often uranium was recycled to avoid the problems encountered in startup and shutdown of the solvent extraction system.

The net result of the above discussion is that to make an accurate extrapolation of the flowsheet waste streams. The cold feed processed must be known (or estimated) in addition to the production numbers (hot feed processed). The following cold feed recycle estimates are used in this document; they may be changed in later revisions as additional data are located:

Decladding. This always was a batch operation and very little, if any, rework was done. Thus, a cold feed percentage is not appropriate for this operation in any of the processes.

Bismuth Phosphate. Again, all of the BiPO_4 processing was on a batch basis, only the plutonium was recovered, and very little recycle was performed. Page 507 of the *Bismuth Phosphate Manual* (GE 1944) notes that through November 1945, no MW solution from either B or T Plant had been reworked. (These early periods of plant operation usually show high rework.)

Uranium Recovery (U Plant). The TBP UR process utilized solvent extraction. The uranium product was calcined for shipment offsite; thus, there very likely was significant rework of cold uranium. However, data concerning this rework have not yet been located, so no cold feed percentage has been established.

REDOX. This solvent extraction process is assumed to have experienced significant recycle of cold uranium, particularly following plant startup and when the plant was switched from continuous to weekly operation in October 1958. The following percentages of cold feed production (i.e., rework) are inferred from aluminum nitrate nonahydrate (ANN) consumption records, and should be added to the hot feed production.

1952 to 1953, 40 percent cold or rework feed (add to hot feed)

1954 to 1957, 8 percent cold or rework feed (add to hot feed)

1958 to 1966, 44 percent cold or rework feed (add to hot feed).

PUREX. The following values should be used until additional data are located.

For aluminum-clad fuel:

1956 to 1958, 15 percent cold feed (add to hot feed)

1959 to 1961, 10 percent cold feed (add to hot feed)

1962 to 1972, 6 percent cold feed (add to hot feed) (Hobart and Larson 1970).

For N Reactor fuel:

Through 1972, 9 percent cold feed (add to hot feed) (Hobart and Larson 1970)

1983 through 1989, 63 percent cold feed (add to hot feed) (Schofield 1991).

4.3.2 Status of Supplemental Information on Other Processes

To date, records that can be used for estimation of flowsheet deviations have not been identified for strontium semi-works or B Plant Waste Fractionization. In addition a number of smaller operations at the Hanford Site, such as the hot semi-works, and the PFP, have not been included in previous global inventory reviews. Secondary operations and special processing campaigns may have limited documentation.

Process flowsheets for the thoria campaigns in PUREX would not include data on the large volume of chemicals required to flush the process equipment before and after thoria processing. (Flushing of the plant was necessary to minimize cross contamination of the plutonium, thorium, and uranium products.) However, details of the chemicals used to flush the equipment for the thoria campaigns are included in the first campaign reports.

4.4 OTHER BASES AND ASSUMPTIONS THAT IMPACT GLOBAL INVENTORIES

Individual tank waste inventories are also impacted by the inadvertent routing of products (i.e., product losses of uranium, plutonium and neptunium to the waste tanks) along with the chemicals and fission products from the reprocessing, recovery, and purification processes. Another factor is that because of the waste management practices used in tank farms, the waste routed to a particular waste tank often did not remain in the tank, i.e., it subsequently was routed to another destination, either deliberately or inadvertently.

4.4.1 Product Losses

The primary reason for building and operating the Hanford Site was the production of plutonium, with subsequent recovery for use in nuclear weapons. Thus, minimizing plutonium losses during the reprocessing of the irradiated fuel was always an objective. The plutonium losses in the BiPO_4 process ran approximately 2.6 percent total (Schneider 1951) with approximately 0.4 percent losses in the wastes from the Extraction, Second Decontamination and Plutonium Concentration Purification Cycles, and a larger 1.4 percent loss in the First Decontamination Cycle.¹ The subsequent solvent extraction reprocessing flowsheets (REDOX and PUREX) typically had lower plutonium losses to waste in addition to also recovering uranium. Because these were continuous processes, the waste batches with high product losses could be recycled to recover the product values without having to stop normal production and/or develop complicated rework processes. It was also much easier to recycle out-of-specification products, either plutonium or uranium, in order to maintain high product purity. The plutonium losses in the REDOX and PUREX processes ran on the order of 0.5 percent or less.

The BiPO_4 process did not recover uranium. Essentially all of the uranium was routed to the MW tank cascades. The uranium was subsequently recovered via sluicing and acid dissolution (in new stainless steel process vessels) of the MW from these cascade tanks. The dissolved uranium solution was transferred to the U Plant, 221 U, which was the third, unused BiPO_4 plant built during World War II. The U Plant was modified by the installation of new equipment to recover uranium by a TBP solvent extraction UR process. The plutonium valence in the solvent extraction feed stream was adjusted so that any plutonium present reported back to the waste stream returned to underground tanks since the U Plant was not equipped to recover plutonium, and plutonium limits in the uranium product were very low. The U Plant uranium losses ran around 1 percent. The REDOX and PUREX process uranium losses were on the order of 0.2 percent (see Section 6.3).

¹The percent plutonium losses quoted in this section are those downstream of the decladding and dissolution processes and are considered typical. Plutonium losses are discussed further in Section 6.0 and detailed in Appendix G.

Neptunium-237 was a valuable byproduct at the Hanford Site, particularly from the late 1950's to the 1970's. The ^{237}Np was recovered from the irradiated fuel, purified, and further irradiated to produce ^{238}Pu via neutron capture. The half-life of ^{238}Pu made it particularly suitable as a thermoelectric generator. Unrecovered neptunium reported to the HLW just as did the plutonium losses. In the late 1980's, the DOE determined that there no longer was a need for the Hanford Site ^{237}Np , so neptunium recovery was stopped and the onsite recovered ^{237}Np inventory (12.73 kg) was dumped to waste tank 241-AZ-102.

4.4.2 Wastes Discharged to Ground

Several Hanford Site waste streams were discharged to ground for disposal. Most of these were wastewater streams, e.g., cooling water, steam condensate, etc. These streams normally did not contain radioactivity or appreciable chemicals, and even under failure conditions any such inventory was low. However, there were some process waste streams that were disposed to ground in the 1940's and 1950's that contained substantial amounts of chemicals and radionuclides. These streams originated from the BiPO_4 process and included wastes (all supernatants) from the 224 (plutonium concentration/purification) process, the 2C, some of the 1C, and the UR wastes from U Plant (and 1C from T Plant) after scavenging for ^{137}Cs removal. These wastes were discharged to ground either via overflow to a reverse well or crib from a settling tank (the original 224 waste disposal method) or the last tank in a cascade of at least three tanks (for later 224 and 2C wastes). The 1C and UR wastes were decanted to cribs, usually with a floating suction pump to avoid discharging solids to the disposal site. Waite (1991) contains listings of chemicals and radionuclides discharged to the various ground disposal sites. The chemical discharges are estimates based on process flowsheet knowledge for the 224 and 2C wastes; and on process knowledge and samples for the 1C and UR wastes. In general, reported radionuclide discharges were always based on samples. Usually, only the major constituents were estimated; thus, there may be other chemicals discharged to ground that do not show in the listings. The process flowsheet should be consulted if it is suspected a chemical was discharged but no estimate is shown. Waite (1991) is the best source of chemical discharge estimates, at least until the solubility model in the HDW model (Agnew et al. 1996), has been benchmarked.

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5.0 BEST-BASIS GLOBAL INVENTORY EVALUATIONS

Global component inventories for SSTs and DSTs currently serve as waste composition data for TWRS process flowsheet modeling work, safety analyses, and risk assessments. Information used to establish these inventories originated from key historical records, e.g., essential material purchase records, from various chemical flowsheets used in reprocessing of irradiated Hanford Site reactor fuels, and from calculations of radionuclide isotope generation and decay.

The HDW model (Agnew et al. 1996) generates chemical and radiochemical inventory estimates for each of the underground tanks as well as global inventories for SSTs and DSTs. Shelton (1996) also reports tank inventories and global inventories for the Hanford Site underground waste tanks. This TWRS baseline inventory has been used to support the TWRS process flowsheet modeling work. The HDW model global component inventories are often inconsistent with the TWRS global component inventories. Table 5.0-1¹ illustrates these inconsistencies for some chemical components. Note, for example, the large discrepancy in the reported sodium inventory values. The sodium inventory is of particular importance because it influences the volume, and subsequently the cost, of low-level waste (LLW) that will be produced and stored on the Hanford Site.

Because global component inventories are important to numerous users, one of the first objectives of the standard inventory task was to resolve differences among reported global inventory values and to establish best-basis inventory values. The methodology for resolving these differences and providing best-basis values includes evaluating and documenting the bases, i.e., assumptions, data, and background information, that support these values. This methodology is depicted schematically in Figure 5.0-1. The best-basis global chemical inventory values derived as part of this evaluation process are summarized for comparison purposes in the last column of Table 5.0-1. The methodology for developing radionuclide inventory values is discussed in Section 6.1 and the global evaluation for total uranium is presented in Section 6.3.

¹The HDW model inventories in Table 5.0-1 reflect the global values reported in HDW model Rev. 3 (Agnew et al. 1996) rather than the global values from HDW model Rev. 4 (Agnew et al. 1997). This is due to the fact that most of the global inventory evaluations reported in Sections 5.1 through 5.24 were conducted in fiscal year 1996 and used HDW Rev. 3 model as the basis for comparison.

Table 5.0-1. Comparison of Hanford Defined Waste and Tank Waste Remediation System Inventories. (2 Sheets)

Component	Single-shell tanks		Double-shell tanks		Global inventory		
	HDW (MT)	TWRS (MT)	HDW (MT)	TWRS (MT)	HDW (MT)	TWRS (MT)	Best-basis (MT)
Al	5,530	3,390 ^a	2,190	1,570	7,720	4,960 ^a	7,845
Bi	513	262	11.3	2.23	524	264	580
Ca	497	128	122	25.7	619	154	214
Ce	NR	232	NR	2.78	NR	235	8.8
Cl	576	415 ^b	381	272	957	687 ^b	500
CO ₃	2,880	1,660	1,950	1,550	4,830	3,210	4,830
Cr	652	268 ^c	133	84.9	785	353 ^c	785
F	269	812	369	389	638	1,200	1,360
Fe	1,615	631	220	162	1,835	793	1,230
Hg	5.81	NR	1.05	NR	6.86	NR	2.1
K	181	55.3	300	564	481	619	481
La	40.3	1.86	0.137	21.1	40.4	23.0	51
Mn	15.1	121	23.6	28.3	38.7	149	105
Na	25,850	57,300	14,430	11,200	40,300	68,500	54,200
Ni	144	203	38.2	10.9	182	214	111
NO ₂	8,980	6,520	4,580	3,010	13,600	9,530	85,700
NO ₃	29,100	100,000	16,200	7,490	45,300	107,000	
OH _{TOTAL}	16,500	12,700	6,510	4,010	23,000	16,700	23,000
Pb	268	28.3	10.7	6.20	279	34.5	279
PO ₄	3,310	4,730 ^d	715	237	4,030	4,970	6,000
Si	477	458 ^a	136	234	614	692	570
SO ₄	2,000	1,650	1,220	394	3,220	2,040	5,000
Sr	167	36.0	0.0290	0.619	167	36.6	31.3
TOC	908	473	897	669	1,800	1,140	4,000

Table 5.0-1. Comparison of Hanford Defined Waste and Tank Waste Remediation System Inventories. (2 Sheets)

Component	Single-shell tanks		Double-shell tanks		Global inventory		
	HDW (MT)	TWRS (MT)	HDW (MT)	TWRS (MT)	HDW (MT)	TWRS (MT)	Best-basis (MT)
U _{TOTAL}	2,100	1,420	240	37.7	2,340	1,460	965
Zr	37.0	382	195	305	232	687	440

HDW = Hanford Defined Waste model (Agnew et al. 1996)

NR = Not reported

TWRS = Tank Waste Remediation System (Shelton 1996)

^aIncludes aluminum that may be found in cancrinite (2NaAlSiO₄ · 0.52NaNO₃ · 0.68H₂O); total aluminum inventory adjusted from the 1987 Environmental Impact Statement (DOE 1987).

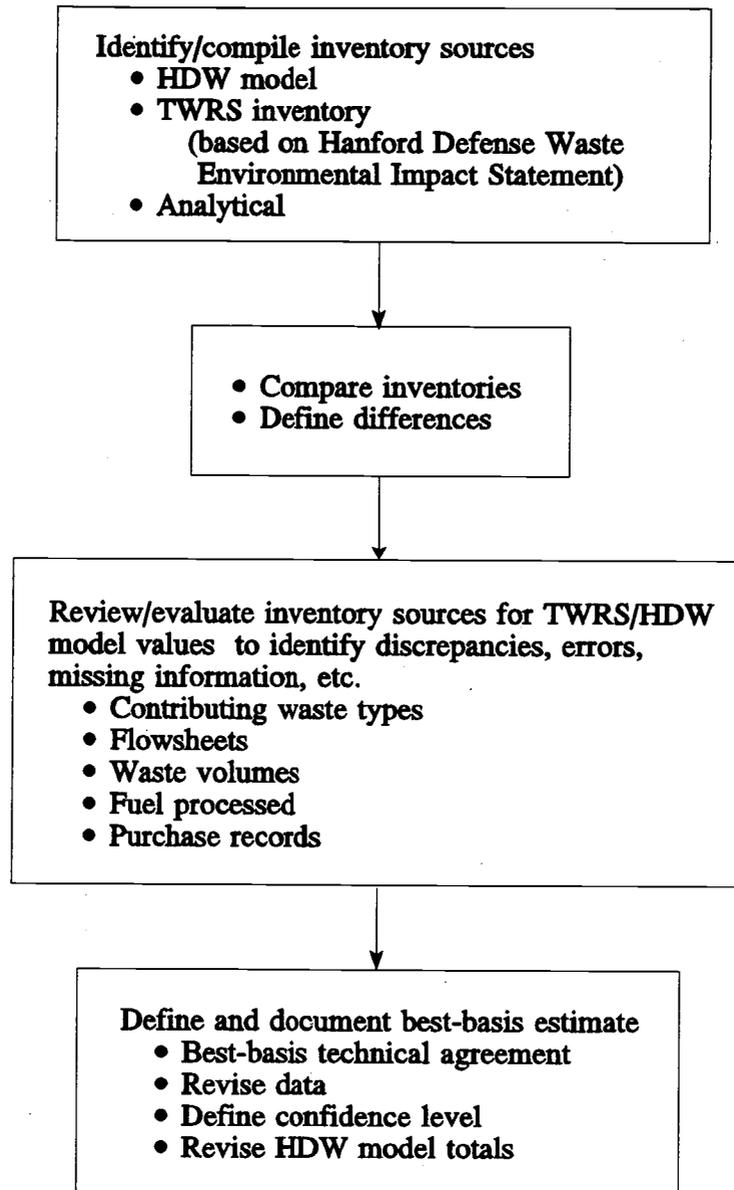
^bSingle-shell tank inventories were adjusted from 4.0E+01 MT reported in the 1987 Environmental Impact Statement (DOE 1987).

^cSingle-shell tank inventories were adjusted from 95.9 MT reported in the 1987 Environmental Impact Statement (DOE 1987).

^dSingle-shell tank inventories were adjusted from 8,740 MT reported in the 1987 Environmental Impact Statement (DOE 1987).

^eIncludes silicon that may be found in cancrinite (2NaAlSiO₄ · 0.52NaNO₃ · 0.68H₂O).

Figure 5.0-1. Methodology for Developing Global Inventory Values for Major Components.



Legend

- HDW - Hanford Defined Waste
- TWRS - Tank Waste Remediation System

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Data quality for the work described in this section was maintained by adhering to the following steps:

1. Critically examine all input data and assumptions used in the TWRS Inventory and HDW model inventory calculations to identify any discrepancies, errors, missing information, etc.
 - Identify waste types that contributed to the majority of the component inventory.
 - Evaluate process stream compositions from chemical flowsheets.
 - Review amount of fuel processed, waste transfers, e.g., crib discharges, purchase records.
 - Review/evaluate tank waste sample analyses.
2. Document all bases and obtain agreement among the majority of technical experts that the revised input data are sound.

Retracing the evolution of data has proved challenging, and every attempt is being made to document original sources of data and information to ensure that inventory values resulting from these evaluations are technically defensible and reproducible. The following Sections 5.1 through 5.24 present global inventory evaluations for 24 chemicals.

Several sections provide tables that total the various mass contributions for a chemical component. At the author's discretion, these totals are often rounded to two or three significant figures. The summation of individual values may not be exactly equal to the rounded value presented.

5.1 ALUMINUM

The primary sources of aluminum (Al), a major constituent in tank wastes, were: (1) the Al cladding on the irradiated fuel (greater than 90 wt% of the fuel processed at the Hanford Site was aluminum-clad), and (2) the Al added as aluminum nitrate nonahydrate (ANN) - $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ as a salting agent in the REDOX solvent extraction process. Smaller sources of aluminum were the aluminum canisters used to contain the early New Production Reactor (NPR) (N Reactor) fuels processed at the REDOX Plant in 1965 and 1966, ANN salting agent for the Plutonium Finishing Plant (PFP) solvent extraction system, and aluminum added as ANN to complex fluoride ion, thereby reducing the corrosion of the stainless steel process vessels and piping. The PUREX Plant used ANN for this purpose during thorium fuel processing and zirconium-clad fuel decladding (Zirflex process), and all plants used ANN when fluoride ion was used in flushes.

The total mass of Al in the tank waste is important since the amount is so large that it will greatly impact the volume of the final waste glass products. The only larger mass metallic component is sodium (see Table 5.0-1). Aluminum is relatively widespread throughout the waste tanks. Aluminum from the bismuth phosphate process decladding waste was combined with the first decontamination cycle (1C) wastes. Solids from this waste stream, including aluminum solids, preferentially settled in the first tank of the 1C cascades. When the 1C supernatants were later concentrated, more solids crystallized out in the evaporator bottoms tanks. Aluminum concentrations are high in almost all the REDOX process waste tanks (primarily S, SX, and U farm tanks), both from the ANN additions and the fact that the REDOX Plant cladding wastes and concentrated solvent extraction wastes were not initially segregated in the waste tanks; wastes were segregated after 1954. The PUREX process aluminum cladding wastes were always routed to the C farm tanks; however, the waste supernatants were later concentrated in the In-Tank Solidification (ITS) systems (bottoms in BY farm) and the 242-T Evaporator (bottoms in the TX farm). Eventually the supernatants were further concentrated in the vacuum evaporator-crystallizers, 242-S for 200 West Area and 242-A for 200 East Area.

5.1.1 Identification of Aluminum Inventory Values

The reported global aluminum inventory values are 7,720 MT (HDW model, Agnew et al. 1996) and 4,960 MT (TWRS, Shelton 1996) (see Table 5.0-1). A separate estimate is provided below to verify these values. The estimate details are shown in Table 5.1-1.

Table 5.1-1. Estimated Metric Tons Aluminum in Tank Waste.

Plant	Fuel type	MTU ^a	kg Al/MTU	MT Al
Bismuth phosphate	Solid natural	7,800	32.2	~251
REDOX	Solid natural	10,718	32.2	~345
	Solid enriched	966	35.1	~34
	I&E natural	890	41.7	~37
	I&E enriched	6,964	47.1	~328
	NPR Al canisters	225	15.3	~3
Total REDOX Process Cladding Wastes				~747
REDOX Process Salt Wastes (from ANN salting agent) ^b				~4,210
Total all REDOX Process Wastes				~4,957
PUREX	Solid natural	15,683	32.2	~505
	I&E natural	43,065	41.7	~1,796
	I&E enriched	7,176	47.1	~338
	NPR (ANN to complex fluoride ion) ^b	5,465	15.1	~83
	NPR (fabrication impurity) ^b	5,465	--	~4
	Thoria processing ^b	Decladding, ANN complexing of fluoride ion		~43
Total all PUREX Process Wastes				~2,769
Plutonium Finishing Plant Waste ^b				~88
Total all Aluminum to Waste				~8,065
Aluminum to Ground ^b				~220
Total Aluminum in Tank Waste				~7,845

ANN = Aluminum Nitrate Nonahydrate - $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$

I&E = Internal and External (Cooled)

NPR = New Production Reactor (N Reactor)

PUREX = Plutonium-uranium extraction

REDOX = Reduction and oxidation

^aFrom Appendix B

^bSee text.

5.1.2 Aluminum Inventory Evaluation

5.1.2.1 Aluminum Decladding Wastes. The amount of aluminum depended on the design of the fuel element. Values ranged from approximately 31.8 kg Al/MTU, for the early solid natural uranium elements processed in the bismuth phosphate plants (GE 1944) (32.2 kg Al/MTU was used in Table 5.1-1 for the solid natural uranium fuel to reflect the later increase in cladding wall thickness [Weakley 1958]), to approximately 47.1 kg Al/MTU for the I&E cooled annular enriched uranium (0.947 wt% ^{235}U) fuel elements later processed at the REDOX and PUREX Plants. The bulk of the fuel was natural uranium I&E elements processed at the PUREX Plant in the 1960's. Matheison and Nicholson (1968) reported these elements typically contained approximately 41.7 kg Al/MTU.

5.1.2.2 REDOX Process Salt Wastes. The REDOX process used ANN as a salting agent for solvent extraction with Hexone. The REDOX process flowsheet documents show this usage as approximately 129.2 kg Al/MTU in 1955 (Merrill and Stevenson 1955), and it was even higher when the REDOX Plant started up in 1952; and down to approximately 75.9 kg Al/MTU in 1965 (Isaacson 1965). The value shown in Table 5.1-1 for total MT Al is taken from a source that reported annual ANN usage per ton of uranium processed (Jenkins and Foster 1978). These values are considerably higher than the flowsheet values quoted earlier and reflect the fact that the REDOX Plant recycled a substantial amount of cold uranium in addition to "hot" feed (see Section 4.3.1).

5.1.2.3 Additions for New Production Reactor (Zirflex) Processing. Table 5.1-1 shows entries for the aluminum NPR canisters dissolved at the REDOX Plant, for the ANN added at the PUREX Plant for complexing of fluoride ion and for aluminum contained in the NPR uranium fuel alloy (see Section 7.0).

5.1.2.4 Additions for PUREX Thoria Processing. The PUREX Plant conducted major thoria target processing campaigns in 1966 and 1970 to produce ^{233}U . A total of 629 MT of thoria were processed (see Appendix B). The thoria target elements were clad in aluminum, thus an aluminum decladding waste was produced. Both campaigns also used fluoride ion (in acidic solutions) in precampaign flushes to cleanout the process vessels and lines, and in the thoria dissolution to speed dissolving rates. The fluoride ion then was complexed with ANN in downstream vessels to minimize corrosion. The actual amount of ANN used for fluoride complexing in the 1966 campaign was listed in Isochem (1967). The flowsheet values for the thoria target aluminum cladding are given in Jackson and Walser (1977). The data from these two documents were used to calculate that the thoria campaigns contributed a total of 43 MT of aluminum to the tank waste. (See Appendix E, for calculation details.)

5.1.2.5 Additions for Plutonium Finishing Plant Processing. The PFP contained a Plutonium Reclamation Facility (PRF) that used a carbon tetrachloride-TBP solvent extraction flowsheet to recover plutonium from dissolved scrap and other rework streams. This solvent extraction process, like the REDOX process, used ANN as a salting agent. The waste from this process, along with other PFP liquid wastes, was originally discharged to ground. However, in June 1973, the PFP liquid waste was routed to the underground

storage tanks. The estimated aluminum content of the tanked PFP wastes is shown in Table 5.1-1. The details of this estimate are given in Appendix E.

5.1.2.6 Aluminum in Wastes Discharged to Ground. Waite (1991) gives estimated values for chemicals discharged to the soil. Table E-1 in Waite's document lists $6.69 \text{ E}+05$ kg of sodium aluminate (220 MT of aluminum) disposed. This value is subtracted in Table 5.1-1. The number seems somewhat high since it represents approximately 88 percent of the total aluminum in the bismuth phosphate process cladding wastes. However, the potential error appears to be small compared to the total aluminum added.

5.1.3 Best-Basis Estimate for Aluminum Inventory Value

The total aluminum estimate shown in Table 5.1-1 (7,845 MT) compares quite favorably with the HDW model (Agnew et al. 1996) estimate of 7,718 MT (the difference is approximately 1.6 percent). However, inspection of the details of the HDW estimate show major differences in the sources (and disposal) of aluminum as compared to Table 5.1-1. Table E-1 in Appendix E lists the process sources of aluminum for both the HDW and current (Table 5.1-1) estimates. It can be seen that while there are several areas of good agreement (e.g., REDOX and PUREX Plant cladding wastes, PUREX Plant ThO_2 , PRF) there are also areas that differ substantially, e.g., the BiPO_4 cladding waste, the REDOX Plant salt (ANN) wastes, the B Plant LLW, and the aluminum disposed to ground. These discrepancies have been pointed out to the HDW model authors and preliminary feedback is that they believe that the values developed in this analysis are more reasonable. Thus, the values shown in Table 5.1-1 should be accepted as the best-basis inventory for aluminum.

5.2 BISMUTH

The first fuel reprocessing scheme used at the Hanford Site to separate plutonium from the bulk of dissolved nuclear reactor fuel was based on the characteristic of plutonium, in its lower oxidation states, to coprecipitate with bismuth phosphate. Application of the bismuth phosphate process resulted in three bismuth-containing waste streams being sent to SSTs for storage or routed through SSTs before being transferred to cribs. These three streams, 1C, 2C, and 224, are the source terms for the in-tank bismuth inventory.

Bismuth is important to disposal operations because (1) as a major component in some washed sludges, it can potentially impact glass volume and quantity, and (2) it can interfere with transuranic element extraction if some of the more extensive separation processes are employed.

5.2.1 Identification and Comparison of Bismuth Inventory Values

Inventory values reported for bismuth are summarized in Table 5.2-1. The value reported in the *Hanford Defense Waste-Environmental Impact Statement* (DOE 1987) and used in TWRS engineering evaluations appears to originate from the value derived by Allen (1976). As a result, these three inventory values are essentially equivalent in terms of two significant figures, i.e., 260 MT bismuth. The predicted HDW model value of 524 MT is twice as large. Table 5.2-2 provides a comparison of the Allen and HDW model inventory values by waste stream. The bismuth concentration, the volume of waste assumed transferred to/through storage tanks, and the resulting bismuth inventory are provided in this table for each waste stream.

Table 5.2-1. Reported Bismuth Inventory Values (MT Bismuth).

Reference	Inventory
Estimated Inventory (Allen 1976)	259
<i>Hanford Defense Waste Environmental Impact Statement</i> (DOE 1987)	261
Tank Waste Remediation System (Shelton 1996)	264
Hanford Defined Waste model (Agnew et al. 1996)	524

Table 5.2-2. Comparison Between Allen^a and Hanford Defined Waste Model Bismuth Inventories by Waste Stream.

Stream	Allen ^a			Hanford Defined Waste Model ^b		
	Conc (M)	Volume (kgal)	Inventory (MT)	Conc (M)	Volume (kgal)	Inventory (MT)
1C	0.01	29,400	233	0.014	28,298	313
2C	0.006	5,530	26	0.01 ^c 0.0053 ^d	8,962 ^c 22,727 ^d	71 ^c 95 ^d
224				0.0062	8,300	41
Total			259			520 ^e

1C = First decontamination cycle of the bismuth phosphate process

2C = Second decontamination cycle of the bismuth phosphate process

224 = Final decontamination/concentration cycle of the bismuth phosphate process

^aAllen (1976)

^bAgnew et al. (1996)

^cAgnew et al. (1996) refers to the second cycle bismuth phosphate process operated between 1944 and 1949 as 2C1

^dAgnew et al. (1996) refers to the second cycle bismuth phosphate process operated between 1950 and 1956 as 2C2

^eDifference between 520 MT calculated and 524 MT reported may be due to rounding.

5.2.2 Bismuth Inventory Evaluation

In Table 5.2-1, the difference between the Allen (1976) and the HDW model global bismuth inventories stems mainly from the differences in the 2C and 224 waste volumes assumed transferred to/through the tanks. Unlike the HDW model, Allen appears to have assumed that the 2C waste from T Plant and the 224 waste were immediately cribbed instead of cascaded through tanks before cribbing. The question is: which one of these inventories, if either, is correct? The following evaluation was performed to help answer this question. The bismuth inventory was examined from two different bases: (1) from a process history basis, similar to that used by Allen and for the HDW model, and (2) from an analyzed waste sample basis.

The bismuth phosphate process flowsheet (Schneider 1951) provided bismuth concentration information for each of the process waste streams. These concentrations were multiplied by the total waste stream volumes to obtain a process history inventory estimate, based on the assumptions that all wastes were cascaded through tanks and that all bismuth precipitated in these tanks before cribbing the aqueous wastes. Analytical data provided bismuth concentration information for in-tank wastes. These data were assumed to be representative of the sludges formed by the three process waste streams, i.e., 1C, 2C, and 224. Concentrations were multiplied by the measured density and the total volume assumed for each process sludge to obtain an analytical-based inventory estimate. The results are summarized in Table 5.2-3.

Table 5.2-3. Bismuth Inventories Projected from Historical Process Information and from Analytical Data for the Bismuth Phosphate Process Waste Types.

Stream	Process ^a			Analytical			
	Conc (M)	Volume (kgal)	Inventory (MT)	Tank waste, density (g/mL)	Conc ^b $\mu\text{g/g}_{\text{sludge}}$	Solids volume ^c (kgal)	Inventory (MT)
1C	0.012	30,350	288	T-104, 1.15	18,900	3,300	271
2C	0.0063	28,980	144	B-110, 1.35	20,100	1,500	149
224	0.0056	33,100	147	B-201, 1.25	103,000	300	147
Total			579				567

^aSchneider (1951)

^bColton (1995)

^cLaFemina (1995).

A second waste sample-based bismuth inventory was derived using bismuth inventories for specific waste types (Table 5.2-4). Waste types called out in Table 5.2-4 were identified by Hill and Simpson (1994) with the Sort On Radioactive Waste Type (SORWT) model; inventory information for each waste type was obtained from Colton (1995).

The global bismuth inventories listed in Tables 5.2-3 and 5.2-4 support the global inventory predicted by the HDW model (Table 5.2-2), although differences are apparent among the waste-specific inventories. Most noticeable is the difference between the inventory for 224 waste predicted by the model, 41 MT (Table 5.2-2), and the inventories for 224 waste projected from waste sample data, 147 MT and 135+ MT (Tables 5.2-3 and 5.2-4). The sample-based bismuth inventories for 224 waste correspond with the process inventory that is based on bismuth precipitating from 125,000 kL (33,100 kgal) of 224 waste, while the model-based value reflects bismuth precipitating from only 31,000 kL (8,300 kgal) waste.

Table 5.2-4. Bismuth Inventory Projected from Sample Analysis Data for Sort On Radioactive Waste Type Groups.

SORWT Group No. (waste type)	Bi Inventory (MT) (Colton 1995)
XXV.i(1C)	36.3
IX(1C-CW)	24.0
XI(1C-TBP)	67.4
VIII(1C-EB)	118
XXV.I(TBP-1Cf)	34.7
XII(TBPf-1C)	30.4
Total 1C-containing wastes	311
XVI(2C-5,6)	50.8
XV(2C-224)	96.0
VI(224)	135
Total 2C- and 224-containing wastes	282
Total	593

1C = First decontamination cycle of the bismuth phosphate process

2C = Second decontamination cycle of the bismuth phosphate process

224 = Final decontamination/concentration cycle of the bismuth phosphate process

CW = Cladding waste

EB = Evaporator bottoms

TBP = Tri-butyl phosphate waste

SORWT = Sort On Radioactive Waste Type from Hill and Simpson (1994).

5.2.3 Best-Basis Estimate for Bismuth Inventory Value

Based on evaluations discussed in this report, the best-basis estimate for bismuth inventory is 580 MT for the following reasons:

- This inventory value reflects the estimated bismuth inventory used in the bismuth phosphate process. This value was picked instead of the inventory value reported by HDW model because of the uncertainty in the volume of 224 waste assumed for the model. The deviation of ± 60 MT from the best-basis estimate inventory value encompasses the value currently predicted by the HDW model and the values projected from analyzed waste samples.
- The inventory value reported by Allen (1976) that supports the *Hanford Defense Waste-Environmental Impact Statement* (DOE 1987) and TWRS inventory values is incomplete. This value does not include bismuth from the 2C and 224 wastes that were cascaded through the tanks before being transferred to cribs.

5.3 CALCIUM

Only two Hanford Site processes added calcium as a flowsheet material. Calcium was added to scavenge ^{90}Sr from tank waste supernatants, and also it was used during plutonium metal production. Calcium is ubiquitous in Hanford Site sludges, however, suggesting trace contamination from water and essential materials as potential sources. Typically, process operations used deionized water, but calcium could also come from large amounts of untreated water used to flush transfer lines. Many of the essential materials used at the Hanford Site had specifications (Palmer 1978) for maximum calcium content ranging from 0.0007 wt% (hydrazine solution) to 0.025 wt% (cadmium nitrate), but the specification for the bulk chemicals NaOH and HNO_3 was 0.002 wt%, which would account for almost all of the calcium from chemical additions.

A few nonroutine additions of calcium, such as cement and diatomaceous earth, are described in the following discussion. Some cribbed waste passed through beds of CaCO_3 as a final treatment before discharge to the ground, but none of this calcium should be found in underground tanks. We have not been able to substantiate or quantify other anecdotal leads such as the use of sacrificial grout linings in process cells.

The extremely low solubility of compounds like CaCO_3 and $\text{Ca}_3(\text{PO}_4)_2$ limits the ability to remove calcium using proposed sludge washing methods. The presence of calcium in Hanford Site HLW sludges can potentially impact sludge vitrification processes. Once calcium is in the melter, under certain conditions a floating $\text{Ca}_3(\text{PO}_4)_2$ phase can form which adversely affects melting rate.

5.3.1 Identification and Comparison of Calcium Inventory Values

Revisions of the HDW model have reflected calcium inventories ranging from 1,120 MT (Agnew 1995) to the current estimate of 619 MT (Agnew et al. 1996). The revised HDW tank inventory (Agnew et al. 1996) distributes 497 MT to SSTs and 122 MT to DSTs, with 121 MT lost to the cribs and tank leaks.

By comparison, the TWRS Inventory (Shelton 1996) accounts for 154 MT of calcium in the tanks distributed 128 MT to SSTs and 26 MT to DSTs (see Table 5.3-1).

Table 5.3-1. Current Reported Inventory Values for Calcium.

	All tanks (MT)	Single-shell tanks (MT)	Double-shell tanks (MT)
Hanford Defined Waste model (Agnew et al. 1996)	619	497	122
Tank Waste Remediation System Inventory (Shelton 1996)	154	128	26

5.3.2 Calcium Inventory Evaluation

The table of "chemicals added" in Appendix B of Agnew et al. (1996) suggests that a base concentration of 0.018M calcium, apparently extrapolated from analytical data, was assigned to most waste streams. Some streams are as high as 0.02M to account for known process additions of calcium. This base concentration seems unusually high in view of the known solubility and infrequent use of calcium. Agnew et al. (1996) acknowledged that their estimate cannot account for the source of a large fraction of the calcium. An important observation about this approach is the impact of an erroneous base concentration. Only a small fraction is attributed to deliberate process additions while 90 to 99 percent of the calcium is accounted for in the base concentration. The HDW model supernatants typically contain 0.009M calcium, and control the split between supernatants and sludges. For comparison, untreated Columbia River water is 0.00075M calcium.

The Sections 5.3.2.1 through 5.3.2.5 attempt to account for calcium by evaluating the potential sources, referred to as the source-based evaluation.

5.3.2.1 Fuel Fabrication. The calcium content of fuel is not known, although judging by the minor element evaluation in Section 5.15, calcium is not recognized as a fuel constituent of any significance. It is reasonable to assume that no calcium comes from fuel.

5.3.2.2 Chemical Process Operations. Chemical processing introduced calcium to the waste through intentional routine additions, however, such additions of calcium were limited to two chemical processes: in-plant/in-farm scavenging of ⁹⁰Sr, and slag and crucible processing for plutonium metal production at Z Plant. The *Inventory of Chemicals Used at*

Hanford Site Production Plants and Support Operations (1944 to 1980) (Klem 1990) substantiates that calcium was not used for the Precipitation Separations Process (i.e., bismuth phosphate) at T Plant (1944 to 1956) or B Plant (1945 to 1952), but was used during Uranium Recovery (In-Plant Scavenging) at U Plant (1952 to 1958) and for In-Farm Scavenging. The amount used for in-plant scavenging is documented; Stedwell (1957) reported that in-plant scavenging used 34,000 kg (75,000 lb) of CaCO_3 for the treatment of 121,000 m^3 (32,000,000 gal) of work. This amounts to 13.6 MT of calcium. Sloat (1955) points out that CaCO_3 was converted to $\text{Ca}(\text{NO}_3)_2$ with nitric acid before using in the scavenging process.

In-farm scavenging is summarized in Jeppson (1993). According to Appendix A of that document, the actual volume processed was 45,800 m^3 (12,100,000 gal) and the total calcium consumption was approximately 20 MT.

The Semiworks Pilot Plant (1955 to 1967) lists $\text{Ca}(\text{NO}_3)_2$ as an essential material, and B Plant process control reports from very early waste fractionization operations indicates CaCO_3 was consumed, probably for preparing test simulants, but the amount used was undoubtedly small.

The second source of process calcium was from slag and crucible waste at Z Plant. A rough estimate of calcium consumption is 20 MT which left the plant in the PRF CAW stream. Virtually all of the slag and crucible waste was discharged to cribs before Z Plant was connected to the tank farm in May 1973. A very small percentage of Z Plant calcium resides in the tanks and is not included in the tank waste inventory.

The REDOX Plant (1951 to 1967) used no calcium. There is no evidence of calcium use at the PUREX Plant (1955 to 1972). Radionuclide removal from PUREX process wastes has not been investigated as a possible source of PUREX process calcium as of this writing.

5.3.2.3 Process Chemical Impurities. The major process chemical with potential for calcium impurities is NaOH. The sodium reconciliation in Section 5.20 identifies 54,200 MT sodium in the tanks, with an additional 23,000 MT routed to the cribs, corresponding to 134,300 MT NaOH or 268,600 MT of 50 wt% caustic soda, assuming all sodium came from caustic (not a true assumption but reasonable for this purpose) and that all calcium in the caustic remained in the tanks. As noted previously, the essential material specification for caustic was 0.002 wt% calcium. The common industry specification for commercial grade caustic soda is 0.006 wt% CaO or 0.004 wt% calcium. Applying the more conservative industry specification, essential material impurities account for only 10 MT calcium.

Agnew et al. (1996) acknowledges that most of the water in Hanford Site waste was deionized before use, but nevertheless attributes much of the calcium in the HDW inventory to exaggerated water hardness. Using the Agnew et al. (1996) estimate of 1,640,000 m^3 (433,000,000 gal) net water volume, the untreated water hardness of 0.00075M calcium, and

assuming that no calcium was lost through cribbing, the maximum contribution from water hardness is 50 MT calcium.

5.3.2.4 Non-routine Tank Farm Additions. In 1966 tank farm operations added 57 MT of Portland cement to 241-BY-105. At 64 wt% CaO (Flinn and Trojan 1975), this contributed 26 MT calcium. Between 1970 and 1972, tank farm operations added 354 MT of diatomaceous earth to the tanks. At 0.9 wt% CaO (Buckingham and Metz 1974), this added 2.3 MT calcium to the tanks.

5.3.2.5 Blowsand. Section 5.19 estimates the amount of blowsand accumulation in the tanks from all sources at 25 MT. With Hanford soil a nominal 6 wt% CaO (Voogd 1993), the contribution of calcium from blowsand is 1 MT.

5.3.2.6 Summary of Source-Based Evaluation. The source-based approach described in Section 5.3.2.1 through 5.3.2.5 accounts for 123 MT of calcium. As noted above, there are other potential sources for which we currently have no estimating basis, so the source-based estimate could increase. By comparison, the TWRS Inventory is 154 MT.

Table 5.3-2. Summary of Calcium Sources.

Sources	Estimated Source (MT)
Chemical Process Operations	33.6
Chemical Impurities	10
Water Hardness	50
Non-Routine Additions	28.3
Blowsand	1
Total	123

5.3.2.7 Analytical Basis. A recent projection of calcium in SST sludge by Colton 1995, shown in Table 5.3-3, places the SST calcium at 172 MT. Colton's estimate is based on sludge analytical data and waste volumes. The low value for calcium in TBP sludge seems peculiar since scavenging of TBP raffinates is the only known intentional process addition of calcium. Through a personal communication, Colton acknowledged that no sludge data were available for most of the TBP wastes, and that no projection could be made without a basis. Earlier in Section 5.3.2.2, however, the calcium usage for scavenging was estimated at approximately 34 MT calcium. Substituting that value for Colton's 18 MT of TBP calcium, we arrive at a revised estimate in Table 5.3-3.

Table 5.3-3. Calcium in Single-Shell Tanks Estimated From Sludge Samples.

Process	Colton estimate (MT)	Revised estimate (MT)
BiPO ₄	42	42
TBP	18	34
REDOX	37	37
PUREX	75	75
Total	172	188

PUREX = Plutonium-Uranium Extraction

REDOX = Reduction and oxidation

TBP = Tri-butyl phosphate.

The presence of calcium in PUREX sludge samples was the basis for a projection of 30 MT by Allen (1976), although no calcium was actually used in the PUREX process. Since Colton had a better sample base from which to project, Colton's number may be more reasonable. Colton's revised sludge-based estimate is more than the TWRS estimate of 128 MT in SSTs, but less than half the HDW model prediction of 497 MT in SSTs. In the future, it would be worthwhile to investigate in more detail how the HDW model (Agnew et al. 1996) arrived at the calcium inventory of the defined waste streams.

5.3.3 Best-Basis Estimate for Calcium Inventory

Both the source-based evaluation outlined above and an independent estimate based on sludge analyses suggest that the HDW model estimate is too high. At the same time, there is evidence that the source-based evaluation may be too low to account for SST calcium.

The sample-based estimate of 188 MT of calcium in SSTs, which is somewhat more conservative than the TWRS inventory and the source-based inventory, is considered the best-basis estimate. There is no reason at this time to suspect the sample-based TWRS estimate of 26 MT in DSTs. This brings the best-basis inventory of calcium to 214 MT (Table 5.3-4).

Table 5.3-4. Best-Basis Estimate of Calcium Inventory in Hanford Site Underground Tanks.

Tanks	MT
Double-shell tanks	26
Single-shell tanks	188
Total	214

5.4 CARBONATE

The following were the primary sources of carbonate (total inorganic carbon), CO_3 , in tank wastes:

1. Absorption of atmospheric carbon dioxide, CO_2 , from ventilation air and airlift circulator air passing through the underground waste storage tanks.
2. Solvent treatment from PUREX and B Plant solvent extraction processes. The U Recovery Process potentially used carbonate for solvent treatment also.
3. The B Plant cesium ion exchange system used ammonium carbonate for scrub and elution streams.
4. Flushing of tank farm pipelines in the 1940's with sodium bicarbonate.

Carbonate exists as an anion. Multivalent metal salts of carbonate are generally insoluble. Monovalent metal (sodium and potassium) salts of carbonate are generally soluble in the alkaline waste supernatant. Multiple passes through the tank waste evaporator process can ultimately result in carbonate salt cakes. In the Hanford wastes, greater than 90 percent of the carbonate is expected to be water and/or caustic wash soluble.

5.4.1 Identification of Carbonate Inventory Values

The reported global carbonate inventory values are 4,830 MT (HDW model, Agnew et al. 1996), 3,210 MT (TWRS, Shelton 1996), and 4,830 MT by this report.

The methodology and analysis to obtain a total global carbonate inventory of 4,830 MT carbonate is contained in Section 5.4.2.

5.4.2 Carbonate Inventory Evaluation

Total carbonate added to tank wastes as process chemicals in PUREX and B Plant was 1,130 MT CO_3 . This carbonate was contained in 1,560 MT Na_2CO_3 used for organic treatment (Section 5.20, Sodium).

The HDW model (Agnew et al. 1996) estimates a total CO_3 inventory of 4,830 MT. The HDW model does not include absorption of CO_2 into tank wastes or degradation of organic carbon as contributions to carbonate.

The largest amount of carbonate in tank wastes is produced by absorption of atmospheric CO_2 from ventilation and sparge air in the underground waste storage tanks. The absorption of CO_2 into tank wastes simultaneously reacts with two free NaOH molecules

to form one Na_2CO_3 and one H_2O molecules. Over time the free NaOH is converted to Na_2CO_3 and the waste pH decreases. Tank farm operations has periodically added NaOH to selected tanks to raise the pH again for corrosion protection.

Modeling of CO_2 absorption into tank wastes would be difficult and the results unreliable. Future assessments of global TOC can use the summation of individual tank inventories when they are available. This study uses the total carbonate inventory estimated by the HDW model (Agnew et al. 1996) and calculates 3,700 MT CO_3 from CO_2 absorption by difference.

DuPont (1946, p. 178) reported that in April 1946, the metal waste line from T Plant plugged up. A flush of the line with 9,100 kg (20,00 lb) of 10 percent sodium bicarbonate cleared the line. A program of bimonthly flushing of these lines in both 200 areas was initiated. Subsequent Uranium Recovery (UR) operations released the metal waste carbonate content to the atmosphere as CO_2 when the metal waste was acidified. This study assumes no carbonate in UR wastes that originated from BiPO_4 metal waste.

5.4.3 Best-Basis Estimate for Carbonate Inventory

Table 5.4-1 provides the results from this evaluation for carbonate, together with comparable estimates from other sources. The global best-basis estimate for Hanford Site tank waste is about 4,830 MT of carbonate. This estimate is the HDW model estimate of 4,830 MT carbonate (Agnew et al. 1996). The current value of 3,210 MT carbonate in the TWRS reference chemical inventory (Shelton 1996) is based on 1987 EIS (DOE 1987) estimates for tank wastes, corrected for known additions to the waste tanks.

Table 5.4-1. Estimated Amount of Carbonate in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
PUREX organic wash	714	-	NA
B Plant organic wash	413	-	NA
CO_2 absorption	3,700	0	NA
Total	4,830	4,830	3,210

HDW = Hanford Defined Waste
 NA = Not attributed to individual waste streams
 PUREX = Plutonium-uranium extraction
 TWRS = Tank Waste Remediation System.

The current Hanford Site tank waste inventory estimate of 4,830 MT carbonate has the highest uncertainty in the amount of CO₂ absorbed in the tank wastes. Sample analyses and carbonate inventory of individual tanks in the future may show that total carbonate inventory is greater than 4,830 MT.

5.5 CERIUM

Cerium added to the waste tanks is primarily from two sources: (1) reactor fission products and (2) ceric ammonium nitrate added during the first cycle of the bismuth phosphate process. Other minor sources include laboratory wastes and engineering development testing.

5.5.1 Identification of Cerium Values

Earlier estimates of cerium have been based on the bismuth phosphate process flowsheet and waste discharge records or estimates of fission products. Table 5.5-1 compares some of the reported values.

Table 5.5-1. Reported Cerium Inventories.

Reference	Inventory (MT)	Comment
Tank Waste Remediation System Baseline (Shelton 1996)	235	EIS values for SSTs. Sample results for DSTs.
Hanford Defined Waste model (Agnew et al. 1996)	Cerium not included	
<i>Hanford Defense Waste Final Environmental Impact Statement</i> (DOE 1987)	230	
TRAC (Jungfleisch 1984)	4	Reports as 2.86 E+04 gram moles. Through 1980 only.
Allen (1976)	244 or 333	Reports inventory of Ce and Ce(OH) ₃ at same mass value. Through 1972 only.

DST = Double-shell tank
EIS = Environmental Impact Statement
SST = Single-shell tank
TRAC = Track radioactive components.

5.5.2 Cerium Inventory Evaluation

5.5.2.1 Chemical Additions. With the exception of TRAC, all previous estimates of cerium inventory of the underground storage tanks are traceable to an estimate made in 1976 (Allen 1976). This document indicated a total cerium inventory of "730,000 pounds" (331 MT) based on the 1951 bismuth phosphate process flowsheet (Schneider 1951) and the waste volumes from a production data notebook (Roberts et al. 1992).

A review of the hand-written calculations for this document (Allen 1976) shows an exponent error for the waste molarity. The molarity used was high by a factor of 100. The corrected inventory would be 3.3 MT of cerium.

The Allen (1976) report contains an additional error in that both Ce^{+3} ion and the $Ce(OH)_3$ compound are reported at the identical "730,000 pounds" (331 MT). The *Hanford Defense Waste Disposal Alternatives* document (RHO 1985a) reported the cerium as 230 MT based on 331 MT of $Ce(OH)_3$, which in turn was used for the TWRS baseline. There are slight variations in the reported values, likely the result of rounding and using different conversion factors. Also, a small quantity (approximately 3 MT) was added to the TWRS baseline (Shelton 1996) to account for the sample results from DSTs.

A spreadsheet also based on the 1951 bismuth phosphate process flowsheet (Appendix C) estimates the cerium added to the underground storage tanks at 3.1 MT using the flowsheet chemical additions and waste volumes.

Minor amounts of cerium were added to the wastes tanks through laboratory wastes (Klem 1990). The only known plant usage outside of bismuth phosphate processing was a one-time process test at the REDOX Plant which contributed less than 50 kgs of cerium.

5.5.2.2 Cerium Fission Product. Fission product cerium is actually the prime contributor to the tank waste inventory. TRAC estimated this inventory at 4 MT through 1980.

Re-estimation of the cerium fission product inventory yields a slightly higher value. Cerium inputs are estimated at 5.03 MT for fuel reprocessed up to 1972 (RIBD) and 0.37 MT for post-1983 PUREX Plant operation (ORIGEN2), for a total of 5.4 MT of cerium. An independent ORIGEN2 calculation reported in Section 7.2 estimates the total fission product cerium production at 5.71 MT.

5.5.3 Best-Basis Estimate for Cerium Inventory

The best-basis global cerium inventory is 8.8 MT from all sources. This small quantity is unlikely to have a significant impact on future tank waste treatment operations.

5.6 CHLORIDE

Most of the chloride in Hanford Site wastes was introduced as an impurity in sodium hydroxide. Chloride has been identified as a component of concern with respect to formulation of both high-level and LLW glasses. Chloride salts have limited retention in glass and can contribute to the formation of secondary molten salt phases during melting operations. Because of its high volatility, chloride can also impact offgas system design.

Due to the high solubility of the chloride anion, the bulk of the chloride in Hanford Site tank wastes is found in the supernatant and interstitial liquids within the tank solids.

Chloride is widespread throughout the waste tanks. Chloride is a monovalent halogen anion and essentially all chloride salts are soluble in acidic and alkaline solutions. In the Hanford Site wastes, approximately 99 percent of the total chloride is expected to be water soluble. The insoluble one percent of chloride is found in the sludges as complex compounds. Soluble chloride in the tank wastes is associated with alkaline soluble sodium chloride (NaCl).

The tank wastes were processed for solids removal (i.e., settling) and/or volume reduction after they were routed to the tanks. The volume reduction processes included scavenging, evaporation, and crystallization processes. These processes redistributed the sodium and associated chloride within the storage tank complex:

1. The scavenging process treated supernatants containing sodium chloride for cesium and strontium removal and discharged the treated supernatants to cribs for disposal of the decontaminated sodium salt solutions and associated chloride.
2. The evaporation processes using the 242-B and 242-T evaporators removed water to reduce tank waste volume, resulting in increased sodium and chloride concentrations and transfer to different storage tanks.
3. The later crystallization processes evaporated additional water from the supernatants to crystallize sodium nitrate salts and further reduce tank waste volume. This crystallization preferentially crystallizes sodium nitrate and provides a decontamination from the associated chloride in the salt crystals. The saturated mother liquor from this crystallization in the tanks is drained from the salt crystals by the process of "salt well pumping" and transferred to the DSTs to become Double-Shell Slurry Feed (DSSF). The residual SST interstitial liquor and DSSF contains the remaining uncrystallized sodium nitrate and the other soluble sodium salts including sodium chloride.

As a result, the SSTs currently contain approximately 29 percent of the total waste chloride in interstitial solutions with high sodium concentrations, eight percent associated with SST sludges, and less than 1 percent in the sodium nitrate crystals. The DSTs contain

the remaining approximately 63 percent of the total waste tank inventory of chloride as predominately supernatants.

5.6.1 Identification of Chloride Inventory Values

The reported global chloride inventory values are 957 MT (HDW model, Agnew et al. 1996), 687 MT (TWRS, Shelton 1996), and 500 MT by this report.

The methodology and analysis to obtain a total global chloride inventory of 500 MT chloride is contained in the following sections.

5.6.2 Chloride Inventory Evaluation

The Hanford Site tank sodium inventory determined by this report is based on analytical data for chloride and sodium and the estimated tank waste inventory of sodium. The available TCR inventories for tanks where both the sodium and chloride inventories are based on chemical analysis are summarized in Tables 5.6-1 and 5.6-2 for DSTs and SSTs, respectively.

Table 5.6-1. Double-Shell Tank Characterization Report Inventories for Sodium and Chloride.

Tank	Sodium, MT	Chloride, MT	Tank	Sodium, MT	Chloride, MT
241-AN-102	562	14.7	241-AW-103 (NCRW)	270	0.078
241-AP-102	426	12.1	241-AW-105 (NCRW)	175	6.56
241-AP-105	519	7.3	241-AZ-101 (NCAW)	17	0.027
241-AP-106	24	0.24	241-AZ-102 (NCAW)	27	0.061
241-AW-101	906	20.4	241-SY-102 (PFP)	86.2	1.3
241-AW-104	26	0.71	Total	575	8.0
241-AY-102	5	1.2	Chloride to sodium mass ratio = 0.0140		
241-SY-101	1,390	56			
241-SY-103	656	17.3			
Total	4,514	130			
Chloride to sodium mass ratio = 0.0289					

NCAW = Neutralized Current Acid Waste
 NCRW = Neutralized Cladding Removal Waste
 PFP = Plutonium Finishing Plant.

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Table 5.6-2. Single-Shell Tank Characterization Report Inventories for Sodium and Chloride.

Tank	Sodium, MT	Chloride, MT	Tank	Sodium, MT	Chloride, MT
241-AX-102	28	0.47	241-C-110	86	1.12
241-B-101	80	0.84	241-C-201	0.4	0.01
241-B-102	23	0.04	241-C-203	1	0.001
241-B-103	50	0.47	241-S-104	219	5.9
241-B-104	153	1.01	241-SX-113	0.4	0.01
241-B-110	327	1.55	241-T-102	4	0.02
241-B-111	102	1.09	241-T-104	150	1.7
241-B-112	24	0.23	241-T-105	34	0.15
241-B-201	5	0.23	241-T-106	6	0.03
241-BX-101	18	0.10	241-T-107	134	0.56
241-BX-103	24	0.03	241-T-108	30	0.22
241-BX-107	192	2.14	241-T-111	80	0.98
241-BY-108	171	1.68	241-TY-104	33	0.21
241-C-107	128	0.61	241-TY-106	9	0.13
241-C-108	33	0.25	241-U-202	1	0.01
241-C-109	25	0.21	Total	2,171	22.0

Chloride to sodium mass ratio = 0.0102.

The DST wastes are segregated into two categories for this evaluation. The NCRW and NCAW wastes produced in the 1983 to 1989 period and the PFP wastes were effectively segregated into five tanks (251-AW-103, 241-AW-105, 241-AZ-101, 241-AZ-102, and 241-SY-102). The remaining DSTs have waste inventories derived from the previous processing of SST wastes (fission product recovery, waste evaporation, crystallization, and saltwell pumping). The chloride to sodium ratio in the DSTs that do not contain NCRW, NCAW, or PFP is representative of the ratio that can be expected in the SST interstitial liquids. These DST supernatants include Dilute Noncomplexed (DN) and DSSF that are the products of evaporation, crystallization, and saltwell pumping in the SSTs.

The available tank characterization results for SSTs are for tanks containing sludges. SST tanks containing salt cake and interstitial liquids can be estimated by using the following:

1. Using the DST Cl/Na ratio for the non NCRW/NCAW/PFP TCR results as the Cl/Na ratio for the interstitial liquid fraction.
2. Assuming a chloride decontamination of 100 from the SST sludge Cl/Na ratio for crystallization of sodium nitrate results in a Cl/Na ratio of 0.0001 for the sodium nitrate crystal fraction.

The sodium distribution in the DSTs and SSTs for salt cake, sludges, and interstitial liquids is estimated in Section 5.20. Combination of the sodium waste type inventories with the comparable Cl/Na ratio provides a total chloride and distribution for the tank wastes as shown in Table 5.6-3.

Table 5.6-3. Distribution of Chloride in Hanford Site Tank Wastes.

Waste	Sodium, MT	Cl/Na mass ratio	Chloride, MT
DST, NCRW/NCAW/PFP	575	0.014	8
DST, non-NCRW/NCAW/PFP	10,600	0.029	310
Subtotal DST	11,200	0.0284	318
SST interstitial liquid	4,900	0.029	142
SST sludge	3,800	0.0102	39
SST salt crystals	34,300	0.0001	3
Subtotal SST	43,000	0.0042	181
Total	54,200	0.0092	500

DST = Double-shell tank

NCAW = Neutralized current acid waste

NCRW = Neutralized cladding removal waste

PFP = Plutonium Finishing Plant

SST = Single-shell tank.

The HDW model chloride inventory estimate of 957 MT assumes that sodium hydroxide purchased at the Hanford Site contained a chloride impurity of 1.0 wt% (Agnew et al. 1996). This assumption accounts for the bulk of the HDW model chloride inventory - the rest coming from minor additions of chloride in the UR process. While it is true that commercial grade caustic typically contained sodium chloride upwards to 1.0 wt% (chloride to 0.61 wt%), there is no indication that caustic purchased by the Hanford Site actually contained this level of impurity. The specification for chloride in sodium hydroxide at the Hanford Site (Palmer 1978) was < 0.9 wt% for chloride but this still doesn't address the average amount of chloride impurity actually delivered to the facilities.

A 50 wt% sodium hydroxide solution contains about 0.29 g Na/g solution. The chloride/sodium mass ratio that corresponds to 1.0 wt% chloride contamination is 0.034 (0.01 g Cl/0.29 g Na).

The mass ratio of total chloride to total sodium in the HDW model is 0.024, (957 MT Cl/40,280 MT Na), which is 160 percent higher than the 0.0092 estimate by this evaluation (500 MT Cl/54,200 MT Na).

5.6.3 Best-Basis Estimate for Chloride Inventory Value

Table 5.6-4 provides the results from this evaluation for chloride, together with comparable estimates from other sources. The best-basis global estimate for Hanford Site tank waste is about 500 MT of chloride. This estimate is significantly lower than the HDW model estimate of 957 MT chloride (Agnew et al. 1996) and the current value of 687 MT in the TWRS reference chemical inventory (Shelton 1996).

Table 5.6-4. Estimated Amount of Chloride In Hanford Site Tank Wastes, MT.

Waste	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
DSTs	318	360	272
SSTs	181	597	415
Total	500	957	687

DST = Double-shell tank

HDW = Hanford Defined Waste

SST = Single-shell tank

TWRS = Tank Waste Remediation System.

The present estimate is based primarily on reported inventories based on analytical results in available Tank Characterization Reports and the estimated sodium inventory from this document (Section 5.20). The analytical based TCR inventories for DST sludges and supernatants in this analysis represent 32 percent of the projected SST interstitial liquid and DST sludge and supernatant inventories. The analytical based Tank Characterization Report inventories for SST sludges in this analysis represent 57 percent of the projected SST sludge inventories.

The HDW model is based on flowsheet caustic consumption and an assumed maximum chloride impurity level in the caustic, and waste transfer records from Hanford Site processing plants. The TWRS inventory is based on the 1987 EIS (DOE 1987) estimate of the Hanford Site tank inventory in 1983 plus additions to the tank system after 1983.

5.7 CHROMIUM

Chromium in the Hanford Site tank waste came from three generic sources: (1) chemical additives used in both the bismuth phosphate and REDOX separations processes, (2) corrosion of stainless steel piping, tube bundles and vessels of the separations plants, and (3) chromic acid used as a decontamination agent in various plants (mainly T Plant). Essentially no chromium was introduced from fuel element impurities or fission products.

In the bismuth phosphate processes, chromium was added in the form of $\text{Na}_2\text{Cr}_2\text{O}_7$ for the purpose of controlling the oxidation state of plutonium. In the REDOX process, chromium was added as $\text{Na}_2\text{Cr}_2\text{O}_7$ for adjusting plutonium to the hexavalent state, but chromium nitrate was also added during early years of plant operation to effect the complete reduction and precipitation of manganese in the headend ruthenium oxidation/scavenging precipitation step.

The addition of excess dichromate to act as a holding oxidant meant that both Cr^{+3} and Cr^{+6} compounds were present in the alkaline waste leaving the plant. Therefore, chromium can be expected to exist in tank waste as both soluble and insoluble compounds. The Cr^{+3} fraction should be found in waste sludges while the Cr^{+6} compounds will likely have accumulated with supernatant solutions. Chromium becomes an important constituent in disposal efforts because of its limited solubility in borosilicate glass, and Cr^{+6} has regulatory implications.

5.7.1 Identification and Comparison of Chromium Inventory Values

The global inventory for chromium as projected by the HDW model (785 MT) is significantly greater than the value estimated for the TWRS inventory (353 MT). In addition, the 1974 estimate of chromium consumption in separations plants (Allen 1976), presumably the original source for the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987), is significantly larger than values that were reported in the TWRS inventory (718 MT chromium as estimated by Allen versus 353 MT chromium as reported in the TWRS inventory).

5.7.2 Chromium Inventory Evaluation

To resolve this significant disparity in chromium inventory values, calculations were performed to reevaluate in greater detail the amount of chromium consumed in the REDOX fuels separation plant during its operating lifetime, 1952 through 1966. This reevaluation begins with the REDOX process (see Table 5.7-1) since other separations or waste treatment processes did not add significant quantities of chromium in comparison to that added in the REDOX process. Six REDOX process flowsheet documents plus monthly reports were inspected to identify dates when changes were made in individual stream flow rates or

chromium concentrations. Monthly chromium consumption factors (gmol/MTU) varied as much as a factor of 1.8, from highest consumption in the mid 1950's to lowest consumption in the mid 1960's. These consumption data were then combined with a file of historical uranium throughput by month to generate a total consumption value over the life of the REDOX Plant. This value was then increased by an estimated 15 percent in 1952 through 1957 and 44 percent in 1958 through 1966 to allow for flowsheet inefficiencies and periods of rework. A range of results is shown in Table 5.7-1.

Table 5.7-1. Total Chromium Additions in REDOX Plant Operations 1952 to 1966 (MT of Chromium).

Uncertainty in Tank H4 chromium addition, 1952-1959	Uncertainty in timing of flowsheet changes	
	Minimizing assumptions	Maximizing assumptions
Low: (398 gmol/MTU)	703	749
High: (596 gmol/MTU)	796	895

REDOX = Reduction and oxidation.

Table 5.7-1 gives the results of several sensitivity calculations to account for uncertainties in the timing of process changes and to account for a period of frequent flowsheet modifications related to the headend ruthenium oxidation step. (In the 1952 to 1959 time period, the headend chromium addition was frequently modified; surviving flowsheet documents do not record these variations. This situation is unfortunate because of all the REDOX process chemical addition streams, the headend chromium addition was the largest source of chromium in the entire process.) The result of these uncertainties is that total chromium consumption in the REDOX Plant could have ranged from 703 MT to 895 MT.

The global chromium inventory not only includes REDOX process consumption (i.e., 703 to 895 MT) but also must account for consumption in B and T Plant operations as well as stainless steel corrosion in all four separations operations. B and T Plant operations have previously been estimated to have consumed 19 MT of chromium (Allen 1976). The contribution to global chromium from all corrosion sources (68 MT) can be estimated from a similar analysis done for corrosion of iron and corrosion of nickel (Sections 5.10 and 5.15, respectively). By this current reestimate, the global chromium inventory (the sum of REDOX process, B and T Plant consumption, and stainless steel corrosion sources) is in the range of 790 to 980 MT.

5.7.3 Best-Basis Estimate for Chromium Inventory Value

The reestimate of chromium consumption and corrosion sources, presented above, leads to the conclusion that total tank inventory values could not possibly be as low as reported in the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) (268 MT) or in the TWRS inventory (353 MT). That the TWRS data source could be in error may be explained as follows:

- Inspection of the paper trail leading to the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987), and subsequently to the TWRS inventory, suggests that Allen's original estimate of Cr^{+3} ($3.8 \text{ E}+06$ gmol [equivalent to 198 MT of chromium]) was corrupted by a factor of 0.5 in early engineering flowsheets supporting publication of ERDA 77-44 (ERDA 1977).
- Allen's original estimate of hexavalent chromium ($1.0 \text{ E}+07$ gmol [equivalent to 520 MT of chromium]) was inadvertently dropped from the listing of waste chemicals in the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) support document (RHO 1985a).

In summary, it appears that the HDW estimate of total chromium inventory (785 MT) is corroborated by the current reestimate range (790 to 980 MT). Since the HDW estimate is essentially equal to the lower end of the reestimate range, it is recommended that the HDW value of 785 MT be adopted as the best-basis inventory for chromium.

5.8 FLUORIDE

Fluoride in various chemical forms, e.g., KF, NH_4F , HF, and $(\text{NH}_4)_2\text{SiF}_6$, was routinely used in all Hanford Site fuel reprocessing plants and also in the Hanford Site PFP. Compounds of fluoride were added in process operations for several specific purposes. For example, in the PUREX Plant operation large amounts of NH_4F were used to prepare the $\text{NH}_4\text{F-NH}_4\text{NO}_3$ solution used to dissolve Zircaloy® cladding; KF was used as a catalyst in nitric acid dissolution of ThO_2 in the two PUREX Plant thorium campaigns. The REDOX Plant also used an $\text{NH}_4\text{F-NH}_4\text{NO}_3$ solution to dissolve cladding in processing relatively small amounts of Zircaloy®-clad fuel. Ammonium fluorosilicate [$(\text{NH}_4)_2\text{SiF}_6$] was added in both the first and second decontamination cycles of the bismuth phosphate process to increase decontamination of plutonium from fission product zirconium and niobium.

Also, in the bismuth phosphate process solutions of HF were added in the product concentration cycle to provide fluoride ion for precipitation of LaF_3 as a carrier for plutonium. And, in PFP so-called "B Acid" (11 M HNO_3 - 0.2 M HF) was routinely used to dissolve PuO_2 and other plutonium-containing materials.

Fluoride in the Hanford Site's SSTs and DSTs is likely distributed between solid (sludge and salt cake) and liquid (supernatant and interstitial liquid) phases. Knowledge of

the inventory, both global and tank-by-tank, of fluoride is important for at least two reasons. The amounts of waste, either high-level (i.e., sludge, or low-level, i.e., pretreated salt cake) which can be satisfactorily accommodated in candidate glass forms may be limited by the fluoride content of the waste. At vitrification temperatures substantial amounts of fluoride, as HF or other volatile species, may evolve from glass melters. Design of suitable melter offgas systems requires knowledge of the inventory of fluoride-bearing gases to be handled.

5.8.1 Identification and Comparison of Fluoride Inventory Values

Table 5.8-1 compares fluoride inventory values from two data sources. The HDW model Rev. 3 (Agnew et al. 1996) predicts the 177 Hanford Site tanks to contain only about one-half of the fluoride inventory presently carried in the TWRS database. Both the HDW model (Rev. 3) and the TWRS data base agree very well on the amount of fluoride in the DSTs, 369 versus 389 MT. However, the HDW model (Rev. 3) predicts the SSTs to contain only about one-quarter as much fluoride as stated in the TWRS database. The inventory of fluoride in the 28 DSTs is based upon analyses of samples taken from each of the tanks and is, therefore, considered highly reliable.

Table 5.8-1. Currently Reported Inventories of Fluoride in Hanford Site Tanks.

Tanks	Fluoride (MT)	
	Hanford Defined Waste model (Rev. 3) data ^a	Tank Waste Remediation System database ^b
Single-shell tanks	269	812
Double-shell tanks	369	389
All	638	1200

^aAgnew et al. (1996)

^bShelton (1996).

5.8.2 Fluoride Inventory Evaluation

The following sections summarize the major sources of fluoride in tank wastes.

5.8.2.1 Fluoride from Bismuth Phosphate Process Operation.

$(\text{NH}_4)_2\text{SiF}_6$. Appendix C of this report provides detailed chemical flowsheet data for the bismuth phosphate process which was operated at the Hanford Site from 1944 through 1956. Data in Table C1-2 (Appendix C) show that significant amounts of $(\text{NH}_4)_2\text{SiF}_6$ were routinely added in both the first and second decontamination cycles of the bismuth phosphate

process. The function of the added ammonium fluorosilicate was to provide increased decontamination of plutonium from fission product niobium and zirconium which accompanied plutonium in the original product recovery cycle.

Table 5.8-2 lists the fluoride content of the $(\text{NH}_4)_2\text{SiF}_6$ used in the bismuth phosphate process. The behavior of the fluorosilicate anion when the decontamination cycle waste solutions were made alkaline is not known exactly. Very possibly, in alkaline media, the fluorosilicate species decomposed to fluoride ion and one or more silicon species.

Table 5.8-2. Sources of Fluoride in Hanford Underground Tanks.

Plant or process	Plant operation	Form of added fluoride	MT of fluoride
Bismuth Phosphate	Addition of $(\text{NH}_4)_2\text{SiF}_6$	$(\text{NH}_4)_2\text{SiF}_6$	694.00
	Addition of HF	HF	593.00
REDOX	Reprocess N reactor fuels	NH_4F	21.20
	0.947 wt% ^{235}U fuel	NH_4F	4.86
	spike fuel		
	Reprocess Shippingport reactor fuel	NH_4F	14.00
PUREX	Reprocess PRTR fuels	NH_4F	1.84
	Al-Pu alloy fuels	NH_4F	1.84
	$\text{PuO}_2\text{-UO}_2$ fuel		
PUREX	Reprocess N Reactor fuels	NH_4F	54.40
	0.947 wt% ^{235}U fuel	NH_4F	409.00
	1967 to 1972		
	1983 to 1989		
	spike fuel	NH_4F	108.60
	1967 to 1972	NH_4F	75.80
	1983 to 1989	NH_4F	58.50
	coproduct fuel		
Reprocess irradiated thoria	KF	2.45	
PUREX	Reprocess PRTR fuels	NH_4F	4.55
	Al-Pu alloy fuels	NH_4F	6.31
	$\text{PuO}_2\text{-UO}_2$ fuel		
Plutonium Finishing Plant	PuO_2 dissolution and off gas scrubbing	CaF_2	86.50
TOTAL			2,127.00

PRTR = Plutonium Recycle Test Reactor

PUREX = Plutonium-Uranium Extraction

REDOX = Reduction and Oxidation.

Hydrofluoric Acid. For lanthanum fluoride precipitation, data in Table C1-2 indicate that in the bismuth phosphate process 80 g of HF were added for each MT of uranium processed. A total of 7800 MT of uranium were processed through the B and T Plants. Hence, 593 MT of fluoride ion were consumed (as HF) during the time the bismuth phosphate process was operated in the B and T Plants. This quantity of fluoride ion was used in precipitation of LaF_3 , a carrier for plutonium, in the product concentration cycle.

5.8.2.2 Fluoride from REDOX Process Operations.

Processing of N Reactor Fuels. For plutonium production purposes the N Reactor used Zircaloy®-clad uranium metal. Two basic types of fuel elements were used: uranium metal uniformly enriched with ^{235}U to contain (preirradiation) 0.947 wt% ^{235}U and so-called "spike" elements which were fabricated to contain uranium metal enriched to 0.947 wt% ^{235}U and uranium metal enriched to 1.25 wt% ^{235}U . Also irradiated in the N Reactor were so-called "coproduct" fuel elements. The latter consisted of an outer shell of Zircaloy®-clad uranium metal containing 2.1 wt% ^{235}U surrounding an inner cylinder of lithium metal. The co-product fuel elements were used to demonstrate the capability of N Reactor for simultaneously producing plutonium and tritium.

In its last few years of operation the REDOX Plant reprocessed a small quantity of irradiated Zircaloy®-clad N Reactor uranium metal fuel. Data in Appendix B show that a total of 228 MT of uranium were reprocessed including 186.7 MT of uranium containing, before irradiation, 0.947 wt% ^{235}U , 37.4 MT of uranium in spike N Reactor fuel elements (0.947 wt% ^{235}U /1.25 wt% ^{235}U , and 4.0 MT of uranium containing, before irradiation, 1.25wt% ^{235}U .

- **Reprocessing of 0.947 wt% ^{235}U Fuel.** In 1965 and 1966 186.7 MT of Zircaloy®-clad N Reactor fuel containing, before irradiation, 0.947 wt% ^{235}U were declad and dissolved. The chemical flowsheet prepared by Hanson (1966) called for the use of 1.02 MT of NH_4F (0.53 MT of fluoride) for decladding 4.67 MT of uranium. Decladding of 186.7 MT of N Reactor fuel required 21.2 MT of fluoride added as NH_4F .
- **Reprocessing of 1.25 wt% ^{235}U Fuel.** Hanson (1966) prepared chemical flowsheets for decladding and dissolving both spike (0.947 wt% and 1.25 wt% ^{235}U) and 1.25 wt% ^{235}U enriched N Reactor fuel. (It is convenient to address together fluoride consumption for dissolution of these latter two types of fuel.) The Hanson flowsheet involved use of 2,370 lb of NH_4F to remove cladding from 4.68 MT of uranium contained in the spike fuel elements. For processing of 37.4 MT of uranium in the spike fuel 1.89E+04 lb of NH_4F (equivalent to 4.41 MT of fluoride) were required. An additional 0.45 MT of fluoride as NH_4F was used to declad 4.0 MT of N Reactor fuel containing, before irradiation, 1.25 wt% ^{235}U .

Reprocessing of Plutonium Recycle Test Reactor Fuels.

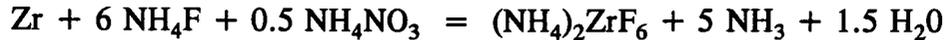
- **Reprocessing of Al-Pu Alloy Fuels.** In its initial operation the PRTR employed Zircaloy®-clad Al-Pu alloy fuel elements. Campaigns to declad, dissolve, and recover plutonium from irradiated Al-Pu alloy fuel were carried out in the Hanford Site REDOX Plant in February and March 1963 (Hanson 1962, Jenkins and Foster 1978). Zircaloy® cladding was removed according to flowsheet conditions defined by Hanson (1962). For each dissolver charge, 1,975 lb of NH_4F (equivalent to 0.46 MT of fluoride) were consumed. Historical records (HW-76848 1963, HW-84845 1963) show that a total of four charges of the PRTR Al-Pu alloy fuel were processed. A total of 1.84 MT of fluoride, as NH_4F , were used to process the four dissolver charges.
- **Reprocessing of PRTR $\text{PuO}_2\text{-UO}_2$ Fuels.** Chemical flowsheets for decladding and dissolution of PRTR 0.48 wt% $\text{PuO}_2\text{-UO}_2$ fuels in the REDOX Plant were drawn up by Hanson (1965). These flowsheets were followed to process four dissolver charges (McDonald 1965) of the $\text{PuO}_2\text{-UO}_2$ fuel. For each charge 1,976 lb of NH_4F (equivalent to 0.46 MT of fluoride) were used to remove the Zircaloy® cladding. The total decladding and dissolution campaigns consumed 1.84 MT of fluoride as NH_4F .

Shippingport Reactor Fuel Processing. Shortly before it ceased operations in 1967 the Hanford Site REDOX Plant reprocessed highly irradiated Zircaloy®-clad UO_2 fuel from the commercial Shippingport nuclear reactor. Shippingport reactor fuel containing 13 MT of uranium was declad in six dissolver charges (McDonald 1966, 1967), each charge contained 5,900 lb of UO_2 and 2,200 lb of Zircaloy® cladding. The Zircaloy® cladding of the irradiated Shippingport reactor fuel was heavily oxidized from long exposure to pressurized water at high temperatures. To ensure satisfactory undercutting of the oxide layer and complete dissolution of the unoxidized Zircaloy® cladding the chemical flowsheet (Davis 1966) used for each dissolver charge called for five separate Zirflex process cycles. Three of the Zirflex process cycles each used 0.847 MT of NH_4F (equivalent to 0.437 MT of fluoride) while the other two cycles each used 1.0 MT of NH_4F (equivalent to 0.5 MT of fluoride). For each dissolver charge of Shippingport reactor fuel the decladding cycles consumed 2.34 MT of fluoride. A total of 14 MT of fluoride, as NH_4F , were used in the REDOX Plant Shippingport reactor fuel campaigns.

5.8.2.3 Fluoride from PUREX Process Operations.

Processing of N Reactor Fuels. From 1967 to 1972 the PUREX Plant reprocessed irradiated N Reactor 0.947 wt% ^{235}U , spike, and coproduct fuel. In addition, from 1983 through 1988 the PUREX Plant continued to reprocess N Reactor 0.947 wt% ^{235}U and spike fuel elements. Table B-5 in Appendix B lists the metric tons of uranium contained in the various types of N Reactor fuel.

The initial step in all N Reactor fuel reprocessing operations in both the PUREX and REDOX Plants was to dissolve the Zircaloy® cladding according to the chemical reaction:



Chemical flowsheets referred to later provide details of the quantity of NH_4F used in PUREX Plant operations to declad the various types of N Reactor fuel.

- **Reprocessing of 0.947 wt% ^{235}U Fuel.** From 1967 through 1972 the PUREX Plant reprocessed 413.2 MT of uranium in which the ^{235}U content, before irradiation, was 0.947 wt% and 0.71 MT of uranium in which the ^{235}U content, before irradiation, ranged from 0.72 to 0.8 wt%. These fuels were reprocessed according to flowsheet conditions stated by Davis (1967) which called for the use of 7.6 moles of NH_4F for each mole of zirconium present. Information in Davis (1967) indicates that the 413.9 MT of uranium was associated with $3.76\text{E}+05$ moles of zirconium, allowing for the fact that Zircaloy-2® contains only about 98 wt% zirconium. For this amount of zirconium 106 MT of NH_4F (equivalent to 54.4 MT of fluoride) were used in the decladding step.

Additional reprocessing of N Reactor fuel containing, before irradiation, 0.947 wt% ^{235}U continued in the PUREX Plant from 1983 through 1988. Data in Appendix B indicate 3,299 MT of uranium were reprocessed in this period. Dissolution of Zircaloy® cladding was performed according to the flowsheet conditions stated in the *PUREX Chemical Flowsheet for Processing N Reactor Fuels*, ARH-2362 (ARHCO 1972). According to these flowsheet conditions, 1.13 MT of fluoride (1,400 gal of 11.2 M NH_4F) were used to remove cladding from 9.11 MT of uranium. Overall, then, 409 MT of fluoride were consumed in reprocessing 3,299 MT of uranium.

- **Reprocessing of Spike Fuel.** A total of 794.2 MT of uranium contained in N Reactor spike fuel elements was reprocessed in the PUREX Plant from 1967 through 1972. Flowsheet conditions stated by Davis (1967) indicate 1.24 MT of NH_4F (equivalent to 0.64 MT of fluoride) were used to dissolve the cladding from each charge of 4.68 MT of spike fuel uranium. Overall, 108.6 MT of fluoride (as NH_4F) were consumed in removing the Zircaloy® cladding from 794.2 MT of N Reactor spike Zircaloy®-clad fuel.

Additional reprocessing of N Reactor spike fuel continued in the PUREX Plant from 1983 through 1988. Data in Appendix B indicate spike fuel containing 591.5 MT of uranium were reprocessed in this latter period. Flowsheet conditions for applying the Zirflex process to declad spike fuel are presented in ARH 2362 (ARHCO 1972). According to these flowsheet conditions, 1.16 MT of NH_4F (equivalent to 0.60 MT of fluoride) were used to remove cladding from each dissolver charge of spike fuel containing 4.68 MT of uranium. In

reprocessing fuel containing 591.5 MT of uranium, a total of 147.6 MT of NH_4F , equivalent to 75.8 MT of fluoride, were used.

- **Reprocessing of N Reactor Coproduct Fuel.** Campaigns were conducted during 1968 and 1969 in the PUREX Plant to reprocess N Reactor coproduct fuel containing 359.3 MT of uranium (Appendix B). Flowsheet conditions stated by Austin and Nicholson (1967) indicated 1.57 MT of NH_4F (equivalent to 0.81 MT of fluoride) were used to remove cladding from coproduct fuel containing 6.14 MT of uranium. A total of 113.4 MT of NH_4F (equivalent to 58.5 MT of fluoride) were consumed in reprocessing of 359.3 MT of uranium in coproduct fuel.

Reprocessing of Thoria Fuels. In two separate campaigns (1966 and 1970) irradiated thorium oxide targets were reprocessed in the Hanford Site PUREX Plant to recover valuable ^{233}U . The thoria fuel was dissolved in nitric acid solutions containing added KF to accelerate the dissolution rate. Section 5.18, Table 5.18-1, indicates that a total of 5.03 MT of potassium, as KF, was used in the two thoria campaigns. The latter amount of KF contains 2.45 MT of fluoride.

Reprocessing of Plutonium Recycle Test Reactor Fuels.

- **Reprocessing of Al-U and Al-Pu Fuels.** As noted in Section 5.14.2.1, the Hanford Site PUREX Plant reprocessed small amounts of various unirradiated Al-U and Al-Pu alloys including Zircaloy[®]-clad PRTR fuel assemblies and/or rods, LWPR fuel elements, thin Phoenix fuel plates and small disks, and miscellaneous aluminum alloy fuel samples. Ten dissolver charges were required to declad and dissolve the entire load of Al-U and Al-Pu alloys. Five of the dissolver charges contained fuel clad in Zircaloy[®]. The chemical flowsheet for processing these latter charges (Mathieson and Nicholson 1972a) required use of 3,910 lb of NH_4F (equivalent to 0.91 MT of fluoride) to declad each charge. A total of 4.55 MT of fluoride, as NH_4F , was used to declad all the Zircaloy[®]-clad alloy fuel in the 1972 PUREX Plant campaign.
- **Reprocessing of 0.5 and 1.0 wt% $\text{PuO}_2\text{-UO}_2$ Fuel.** The Hanford Site PUREX Plant reprocessed Zircaloy[®]-clad irradiated PRTR $\text{PuO}_2\text{-UO}_2$ fuel in both 1969 and 1970. Flowsheets following these reprocessing operations were prepared by Mathieson and Nicholson (1969) and Watrous (1969b). In 1969 two dissolver charges of 0.5 wt% $\text{PuO}_2\text{-UO}_2$ fuel were declad using 3,390 lb of NH_4F (equivalent to 0.79 MT of fluoride) for each charge. In 1969 also one charge of 1.0 wt% $\text{PuO}_2\text{-UO}_2$ fuel was reprocessed again using 3,390 lb of NH_4F (0.79 MT of fluoride) for the decladding step. Thus, a total of 2.37 MT of fluoride, as NH_4F , were used in reprocessing PRTR $\text{PuO}_2\text{-UO}_2$ fuel in 1969.

In 1970 the PUREX Plant continued reprocessing irradiated PRTR PuO₂-UO₂ fuel. Six separate charges of such fuel were de-clad by the Zirflex process. The first charge required 1.53 MT of NH₄F (equivalent to 0.79 MT of fluoride) for the de-cladding step. The remaining five charges each required 0.63 MT of fluoride, as NH₄F, to dissolve the Zircaloy® cladding. Thus, a total of 2.52 MT of fluoride, as NH₄F, was consumed in the 1970 PUREX Plant campaign to reprocess PRTR PuO₂-UO₂ fuel. And, a total of 6.31 MT of fluoride, as NH₄F, was used in both the 1969 and 1970 PUREX PRTR fuel reprocessing campaigns.

5.8.2.4 Fluoride from PFP Operations. During its long period of operation the PFP generated substantial quantities of acidic liquid waste containing fluoride ion. As indicated earlier, fluoride ion was added routinely to the nitric acid used to dissolve plutonium oxide and other plutonium -bearing solids. The resulting dissolver solution constituted feed to a TBP solvent extraction process used to recover and purify plutonium. The aqueous raffinate from the TBP process was combined with other PFP liquid wastes, some of which also contained fluoride ion.

Beginning in early 1973 the composite aqueous acidic waste stream from the PFP was made alkaline and routed to various underground tanks. Starting in 1982 and continuing through 1989 neutralized PFP aqueous waste was routed exclusively to tank 241-SY-102. A recent TCR (Winters 1995) for this latter tank states that 6.27E+06 L (6.27E+03 m³) of neutralized PFP waste were introduced into tank 241-SY-102. Overall, according to a recent estimate by Bergquist (1996) about 1 E+07 L (1 E+04 m³) of PFP waste containing fluoride were discharged to Hanford Site tanks from 1973 through 1989.

Higley of Westinghouse Hanford Company (1996) stated, during a telephone conversation, that the PFP liquid waste came from three different operations: Plutonium Reclamation Facility (PRF), Remote Mechanical C (RMC) Line, and the Analytical Laboratory and Plutonium Process Support Laboratory. The PRF waste included waste generated by TBP solvent extraction operations, process distillate, steam condensate, and laboratory waste. Nominally, the flowrate of the solvent extraction waste known as high-salt waste (HSW) was 235 L/h. The fluoride ion content of the HSW varied between 0.02 and 0.2M, Higley indicated that the nominal fluoride ion content of the HSW was 0.17M. During a PRF campaign, process distillate, steam condensate, and laboratory wastes were generated at a rate of 80 L/h. These low-salt wastes, typically, did not contain fluoride ion.

Wastes generated during a RMC Line campaign included spent scrubber solution, steam condensate, process distillate, and laboratory waste. Spent scrubber solution containing 3.0M fluoride was generated at a rate of 25 L/h. The average flowrate of low salt waste and laboratory waste during a RMC Line campaign was 90 L/h.

A recent communication from Bergquist (1996), who reviewed available historical records, states that the PFP routed 86.5 MT of fluoride to the Hanford Site tanks.

5.8.2.5 Fluoride Discharged to the Ground. Section 4.4.2 of this report describes the various types of process wastes that were disposed of to the ground in the 1940s and 1950s. All these streams originated from bismuth phosphate process operation. Waite (1991) contains listings of chemicals and radionuclides discharged to the various ground disposal sites. Data reported by Waite indicate 772 MT of fluoride were discharged to the ground.

5.8.3 Best-Basis Estimate for Fluoride Inventory Value

Data summarized in Table 5.8-2 indicate as much as 2,127 MT of fluoride were introduced into the Hanford Site underground tanks. Of this amount, 772 MT were disposed of to the ground. The best-basis inventory for fluoride in the SSTs and DSTs is 1,360 MT. This value is in reasonable agreement with the 1,200 MT carried in the TWRS database (Table 5.8-1).

5.9 HYDROXIDE

The following were the primary sources of hydroxide, OH, in tank wastes:

1. Sodium hydroxide used to dissolve aluminum cladding from irradiated fuel in the bismuth phosphate, REDOX, and PUREX fuel reprocessing processes.
2. Sodium hydroxide used to neutralize acidic wastes prior to discharge to the underground carbon steel storage tanks from the bismuth phosphate, uranium recovery, REDOX, PUREX, and B Plant fission product recovery processes.

Hydroxide exists as an anion. Monovalent metal (sodium and potassium) salts of hydroxide are generally soluble in the alkaline waste supernatant. Multivalent metal salts of hydroxide are generally insoluble. The exceptions are hydroxide complexes of aluminum and chromium that can exist as anions, aluminate ($\text{Al}(\text{OH})_4^{-1}$) and chromite ($\text{Cr}(\text{OH})_4^{-1}$), respectively. In the Hanford wastes, the fraction of soluble hydroxide is a function of waste neutralization and storage conditions that affect the split of aluminum between the insoluble aluminum hydroxide ($\text{Al}(\text{OH})_3$) and the soluble sodium aluminate ($\text{NaAl}(\text{OH})_4$).

5.9.1 Identification of Hydroxide Inventory Values

The reported global hydroxide inventory values are 23,000 MT (HDW model, Agnew et al. 1996), 16,700 MT (TWRS), and 23,000 MT by this report.

The methodology and analysis to obtain a best-basis global hydroxide inventory of 23,000 MT hydroxide is contained in Section 5.9.2.

5.9.2 Hydroxide Inventory Evaluation

The hydroxide inventory is determined by performing a global ionic charge balance. The global ion balance is shown in Table 5.9-1. The charge balance is performed on the component inventories determined by this document using the same basic methodology in the HDW model. The silicon inventory of 570 MT is converted to the equivalent 1,540 MT silicate, SiO_3^{2-} , to correspond to the HDW model methodology. The total organic carbon ionic charge of -0.33 per carbon atom is determined from the total organics defined in Section 5.23.

Table 5.9-1. Global Ion Balance. (2 Sheets)

Component	Molecular weight	Inventory, MT	Ion charge	Million equivalents	
				Positive	Negative
Al	26.98	7,850	+3	873	
Bi	208.99	580	+3	8.3	
Ca	40.08	214	+2	10.7	
Ce	140.13	8.5	+3	0.2	
Cl	35.457	500	-1		14.1
CO_3	60.011	4,830	-2		161
Cr	52.01	785	+3, +6	71.4 ^a	
F	19.00	1,360	-1		71.6
Fe	55.85	1,230	+3	66.1	
Hg	200.61	2.04	+2	0.02	
K	39.10	481	+1	12.3	
La	138.92	51	+3	1.1	
Mn	54.94	105	+4	7.6	
Na	22.991	54,200	+1	2,360	
Ni	58.71	110	+2	3.7	
NO_3/NO_2	60.41 ^b	85,700	-1		1,420
OH_{TOTAL}	17.008	23,000	-1		1,350
Pb	207.21	279	+2	2.7	
PO_4	94.975	6,000	-3		189
SiO_3	76.09	1,540	-2		40.6
SO_4	96.066	5,000	-2		104
Sr	87.63	31.3	+2	0.7	

Table 5.9-1. Global Ion Balance. (2 Sheets)

Component	Molecular weight	Inventory, MT	Ion charge	Million equivalents	
				Positive	Negative
U _{TOTAL}	238.07	965	+6	24.2	
Zr	91.22	430	+4	18.9	
TOC	12.011	4,000	-0.33		110
Total	-	-	-	3,460	3,460

^aAssumes 40 percent Cr⁺³, 60 percent Cr⁺⁶

^bAssumes 90 percent NO₃, 10 percent NO₂.

The chromium in tank wastes is a mixture of +3 and +6 valent chromium. The chromium is assumed to be 60 percent Cr⁺⁶ based on the global evaluation of chromium solubility (Colton 1996). Chromium +6 is water soluble and Cr⁺³ is not.

The hydroxide ion equivalents are determined by the difference between total positive equivalents and total negative equivalents not including hydroxide. The resulting 1,350 million equivalents of hydroxide is equal to 23,000 MT of hydroxide.

As stated by the HDW model (Agnew et al. 1996), aluminum is counted as the +3 cation, not as aluminate. The hydroxide value reported includes the hydroxide complexed to the aluminum as aluminate. Also, the hydroxide assumed precipitated with multivalent metal ions is partially converted to hydrated oxides and oxides. Therefore, the free hydroxide in supernatants is more than an order of magnitude lower than the hydroxide value reported in Table 5.9-1. The free hydroxide in all the tank wastes is less than 100 million charge equivalents or gmoles hydroxide. The free hydroxide and amount of aluminum present as aluminum hydroxide or sodium aluminate is a function of total CO₂ absorption and historical tank farm evaporator operation (see Section 5.4, Carbonate).

5.9.3 Best-Basis Estimate for Hydroxide Inventory

Table 5.9-2 provides the results from this evaluation for hydroxide, together with comparable estimates from other sources. The best-basis global estimate for Hanford Site tank waste is 23,000 MT of hydroxide. The HDW model estimate is 23,000 MT hydroxide (Agnew et al. 1996). The value of 16,700 MT hydroxide in the TWRS reference chemical inventory (Shelton 1996) is based on 1987 EIS (DOE 1987) estimates for tank wastes, corrected for known additions to the waste tanks.

Table 5.9-2. Estimated Amount of Hydroxide^a in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
Total	23,000	23,000	16,700

HDW = Hanford Defined Waste

TWRS = Tank Waste Remediation System

^aDoes not include equivalent oxide content in phosphate, sulfate, and silicate anions and assumes no aluminate or chromite anions.

5.10 IRON

Iron (Fe) in Hanford Site tank waste is generated from two sources:

1. Iron containing chemicals added in fuel reprocessing operations, e.g., ferrous sulfamate, ferrous ammonium sulfate, and ferrous nitrate.
2. Corrosion of stainless-steel piping and equipment during fuel reprocessing operations and waste evaporator campaigns, and in-tank corrosion of the mild steel waste storage tanks.

Iron in the alkaline tank waste is a trivalent cation. The ferric salt is insoluble in alkaline solutions. In the Hanford Site wastes, it is expected that greater than 99 percent of the total Fe will report to the HLW.

5.10.1 Identification of Iron Inventory Values

The reported global Fe inventory values are 1,835 MT (HDW model, Agnew et al. 1996), 793 MT (TWRS, Shelton 1996), and 1,230 MT (best-basis) estimated by this report.

The methodology and analysis to obtain a total global Fe inventory of 1,230 MT Fe is contained in the following sections.

5.10.2 Iron Inventory Evaluation

The Hanford Site tank Fe inventory is estimated by computing the total amount of Fe added to each of the separations processes and adding the amounts of Fe in the wastes from corrosion. Sources of information include the following:

1. Process flowsheets (Schneider 1951, GE 1951b, Crawley 1960, Isaacson 1965, Boldt 1966, and Schofield 1991).
2. Partial chemical usage summaries (McDonald 1968, Allen 1976, McDonald and Hogan 1977, and Hogan 1992).
3. Tank transaction records and estimates from the HDW model (Agnew et al. 1995a, Agnew et al. 1996).
4. Crib discharges (Waite 1991).
5. Corrosion analysis contained in Appendix H of this document.

These sources are used to develop Fe inventory estimates for each of the chemical separations processes.

5.10.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process operated at T Plant from December 1944 through 1956 and at B Plant from April 1945 through 1952. The Hanford Site production records indicate that 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the bismuth phosphate processing is developed in Appendix C based on the Schneider (1991) flowsheet. The Appendix H analysis provides an estimated bismuth phosphate processing corrosion input to the wastes of 32 MT Fe. Comparable HDW process and corrosion Fe values are obtained from the waste volume out and iron concentrations of the HDW. The HDW Fe concentrations used are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

The Fe usage in the bismuth phosphate processing is shown in Table 5.10-1 for the Schneider (1991) flowsheet and for the HDW model (Agnew et al. 1996).

Table 5.10-1. Amount of Iron in Bismuth Phosphate Process Wastes, MT.

Waste stream	Present estimate (Schneider 1991)	HDW model (Agnew et al. 1996)
Metal waste (MW)	0	122
First cycle and coating waste (1C/CW)	201	277
Second cycle (2C)	186	214
224 waste	0	28
Corrosion	32 ^a	^b
Total	419	640

HDW = Hanford Defined Waste

^aAppendix H

^bCorrosion contribution included in the streams above.

Metal Waste. The first steps in the bismuth phosphate process dissolved the uranium metal slugs and extracted the plutonium by carrier precipitation with bismuth phosphate. Sulfuric acid was added to the solution to complex the uranyl ion (UO_2^{+2}) and avoid uranyl phosphate precipitation. This resulted in the metal waste stream. The metal waste stream contained the bulk of the uranium and approximately 90 percent of the long-lived fission products (e.g. ^{137}Cs and ^{90}Sr) and was sent to the SSTs for storage.

The stored metal waste containing the uranium from bismuth phosphate processing was later retrieved as the feed for the UR process.

Cribbed Bismuth Phosphate Process Wastes. Portions of the other bismuth phosphate process waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The total Fe discharged to cribs and specific retention trenches was 1 MT Fe in 259,000 m³ (68.5 Mgal) (Waite 1991).

The HDW model estimates 20 MT Fe was cribbed from the bismuth phosphate process in 173,000 m³ (45.8 Mgal) (Agnew et al. 1996). The HDW estimate for bismuth phosphate process wastes is obtained from the crib volume and the supernatant Fe concentration of Agnew et al. (1996). The Fe concentrations are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

Summary for Bismuth Phosphate Process Wastes. The residual MW, 1C, 2C, and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.10-2. This report estimates the residual bismuth phosphate process tank wastes contain 418 MT of Fe versus the 488 MT estimated by the HDW model.

Table 5.10-2. Estimated Amount of Iron in Bismuth Phosphate Process Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
Bismuth phosphate process wastes produced	419	640
Bismuth phosphate process wastes cribbed	1	20
Bismuth phosphate process wastes sent to uranium recovery	0	132
Bismuth phosphate process wastes in single-shell tanks/double-shell tanks	418	488

The principal differences in the present estimate and the HDW model are the input chemicals in the flowsheets assumed and the HDW assumed global presence of 0.016M corrosion Fe in the bismuth phosphate process wastes.

5.10.2.2 Uranium Recovery Wastes. The UR process began in 1952 to extract uranium from the bismuth phosphate process MW stored in 241-B, 241-BX, 241-BY, 241-C, 241-T, 241-TX, and 241-U Tank Farms.

Uranium Recovery Chemicals Added. The design basis flowsheet for the UR process is provided in the *Uranium Recovery Technical Manual* (GE 1951a). According to values in the UR flowsheet, a scrub stream injects 0.05M of $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ and 0.1M of NH_3SO_3 at 50 flows into the "RA" solvent extraction column to treat the MW feed that contains 0.26M of uranium at 104 flows. In essence, the UR flowsheet requires 0.05 gmol of $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ to treat 0.52 gmol of uranium. On this basis, 170 MT of Fe would have been needed to process 7,500 MTU.

From process records, Allen (1976) estimated the amount of $\text{Na}_4\text{Fe}(\text{CN})_6$ and $\text{K}_4\text{Fe}(\text{CN})_6$ used for in-plant and in-tank scavenging of UR wastes. Altogether, 396 MT of $\text{Na}_4\text{Fe}(\text{CN})_6$ and 60 MT of $\text{K}_4\text{Fe}(\text{CN})_6$ were reportedly used. This is equivalent to adding 81.9 MT of Fe to UR waste from scavenging (36 MT from in-tank and 45.9 MT from in-process scavenging). A ferrocyanide scavenging flowsheet analysis (Borsheim and Simpson 1991) projected total iron use of 39 MT Fe. The actual use from records is indicated as twice the flowsheet value.

With the 170 MT of Fe added as ferrous ammonium sulfate and 82 MT of Fe added as ferrocyanide, the total amount of Fe added in UR was 252 MT.

The Appendix H analysis provides an estimated UR corrosion input to the tank wastes of 45 MT Fe.

Cribbed Uranium Recovery Wastes. The HDW model analysis of transfer records from the UR operation indicates that 62.5 vol% of the total UR waste streams containing 3 wt% of the Fe (13 MT Fe) were cribbed.

Waite (1991) states that 155,000 m³ (41 Mgal) of scavenged UR wastes containing 22 MT of Fe was discharged to cribs and specific retention trenches. The 22 MT of cribbed Fe represents 7 percent of the projected Fe content in the UR wastes.

Summary for Uranium Recovery Process Wastes. Table 5.10-3 provides a summary iron material balance for the UR process for this analysis and the HDW model.

Table 5.10-3. Estimated Amount of Iron in Tank Wastes from the Uranium Recovery Process, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
Bismuth phosphate process wastes sent to UR	0	132
UR chemicals added	252	291
UR corrosion	45 ^a	-
Total UR wastes produced	297	423
UR wastes cribbed	(22)	(13)
UR wastes in single-shell tanks/double-shell tanks	275	410

UR = Uranium Recovery
^aAppendix H.

5.10.2.3 REDOX Process Wastes. According to the site production records, 19,706 MTU were processed through the REDOX process from 1952 through 1966 (Appendix B). Chemical purchase records can also be used to generate an estimate of net iron consumption (McDonald 1968). According to these records, 30,000 kg (66,000 lb) of $\text{Fe}(\text{NH}_2\text{SO}_3)_2$ were consumed in the REDOX Plant from 1965 through 1966. This is equivalent to 6.75 MT of Fe produced from oxidation of the sulfamate. The site production records indicate 2,604 MTU were processed through the REDOX Plant from 1965 to 1966 (Appendix B). This results in a recorded 2.59 kg process iron in REDOX process wastes per MTU processed.

From 1952 to 1966, a total of 19,706 MTU were processed through the REDOX Plant (Appendix B). Based on these figures and the assumption of 2.59 kg process Fe/MTU, it is calculated that 50 MT of Fe was introduced by the REDOX process from ferrous sulfamate. This Fe content is 125 percent of the value predicted by the REDOX process Flowsheets 5 through 9.

Because the REDOX process product streams were frequently reworked, the plant experienced frequent startups and shutdowns (five day a week operation for Flowsheets 4 and 5), and specialty processing campaigns for neptunium and nonstandard fuel (performed during the periods represented by Flowsheets 7, 8, and 9), the REDOX process flowsheets cannot be used to accurately account for all of the chemicals added to the process. The flowsheet provides chemical usage at equilibrium operation. The nonstandard operations result in chemical use higher than predicted by flowsheets. Therefore, the reported usage of Fe being 125 percent of the flowsheet values for Flowsheets 5 through 9 is consistent (Section 4.3.1).

The Appendix H analysis provides an estimated REDOX process corrosion input to the tank wastes of 8.5 MT Fe.

Summary for REDOX Process Wastes. Table 5.10-4 provides a summary iron material balance for the REDOX process for this analysis and the HDW model. The HDW Fe values are obtained from the waste volume out and iron concentrations of the HDW.

Table 5.10-4. Estimated Amount of Iron in Tank Wastes from the REDOX Process, MT.

Stream	Present estimate	HDW model (Agnew et al. 1996)
Neutralized Solvent Extraction Waste	50	372
Cladding Waste	0	16
Corrosion	8.5 ^a	_ ^b
REDOX process wastes in single-shell tanks/double-shell tanks	58	388

HDW = Hanford Defined Waste

REDOX = Reduction and oxidation

^aAppendix H

^bCorrosion contribution included in the streams above.

5.10.2.4 PUREX Process Wastes. The PUREX process came on line in January 1956. The PUREX Plant processing can be broken down into three main periods: 1983 through 1989, 1965 through 1982, and 1956 through 1964.

1983 through 1989. The period from 1983 through 1989 processed Zircaloy®-clad fuel exclusively. The total iron use by the PUREX Plant in the 1983 through 1989 period is summarized in Table 5.10-5 for this analysis and the HDW model.

Essential material usage records can be used to define the chemical consumption for the PUREX Plant from 1983 to 1989 (Hogan 1992, and personal communication with J. J. Hogan). J. J. Hogan, personal communication, stated that 103 MT (227,800 lb) of ferrous sulfamate was used at the PUREX Plant from 1983 through 1991. This material would result in 23 MT of iron in the PUREX Plant tank wastes.

Table 5.10-5. Estimated Amount of Iron in 1983 through 1989 PUREX Plant Tank Wastes, MT.

Tank	Present estimate	HDW model (Agnew et al. 1996)
CWZr2	-	47
P3	-	28
PL2	-	97
Essential Material Records	23 ^a	-
Corrosion	36 ^b	- ^c
Total	59	172

CWZr2 = Coating waste from REDOX process, zirconium cladding

HDW = Hanford Defined Waste

P3 = Aging waste from PUREX process high-level waste (1983 to 1988)

PL2 = PUREX process low-level waste (1983 and 1988)

PUREX = Plutonium-uranium extraction process

^aJ. J. Hogan (personal communication)

^bAppendix H

^cCorrosion contribution included in the streams above.

The basis for the PUREX Plant 1983 through 1989 wastes in Agnew et al. (1996) is not defined. The HDW iron values are obtained from the waste volume out and iron concentrations of the HDW.

1965 through 1982. The site production records indicate 25,179 MTU and 629 MT of thoria were processed in the PUREX Plant from 1965 to 1972 (Appendix B). The PUREX Plant was maintained in standby from 1973 through 1982. Monthly essential material usage records can be used to define the chemical consumption for the PUREX Plant from 1965 through 1982 (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.10-6.

Table 5.10-6 shows that the total process Fe used by PUREX Plant processing from 1965 through 1982 is 106 MT process iron.

Table 5.10-6. Iron Use in PUREX Process from 1965 through 1982.

Year	MT processed			Chemical use, MT
	Aluminum-clad U	Zirconium-clad U	ThO ₂	Fe(NH ₂ SO ₃) ₂
1965 ^a	5,385		5	90.7
1966 ^a	3,806		197	80.8
1967 ^a	4,459	123		69
1968 ^b	3,393	427		84.6
1969 ^b	2,568	228		63.2
1970 ^b	555	92	427	33.6
1971 ^b	2,988	287		32.7
1972 ^b	552	416		17.9
1973-1982 ^b	0	0	0	0
Total	23,706	1,572	629	472
Subtot Fe, MT	-	-	-	106 ^c

PUREX = Plutonium-uranium extraction

^aMcDonald (1968)

^bMcDonald and Hogan (1977)

^cTotal PUREX process iron from 1965 through 1982.

1956 through 1964. The third period covers the PUREX production process from 1956 through 1964. During this period, 42,320 MTU were processed in the PUREX Plant (Appendix B). From 1956 through 1964, only aluminum-clad natural uranium fuel was processed through the PUREX Plant.

Examination of the PUREX Plant processing history in Table 5.10-6 shows that 1965 and 1967 processed predominately aluminum-clad fuel. The average chemical usage in 1965 and 1967 was 16 kg of ferrous sulfamate/MTU containing 3.61 kg of process Fe per MTU. A 1968 PUREX aluminum-clad flowsheet (Matheison and Nicholson, 1968) gave iron use at 1.91 kg of process Fe per MTU.

The resulting estimated chemical use in PUREX Plant from 1956 through 1964 is shown in Table 5.10-7. The total PUREX process iron generation from 1956 through 1964 is estimated at 153 MT iron based on process iron use of 16 kg ferrous sulfamate or 3.61 kg of process Fe per MTU.

Table 5.10-7. Iron Use in PUREX Process from 1956 through 1964.

Year	MTU processed	Estimated chemical use, MT	
		Fe(NH ₂ SO ₃) ₂ , 16 kg/MTU	Fe, 3.61 kg/MTU
1956	2,286	36.6	8.2
1957	3,989	63.8	14.4
1958	4,707	75.3	17.0
1959	4,635	74.2	16.7
1960	5,599	89.6	20.2
1961	5,438	87	19.6
1962	5,483	87.7	19.8
1963	5,024	80.4	18.1
1964	5,157	82.5	18.6
Total	42,318	677	153

Total estimated PUREX process iron from 1956 through 1964 = 153 MT.

Summary for PUREX Process Wastes. Iron use by the PUREX Plant is summarized in Table 5.10-8. This analysis gives a total PUREX Plant iron generation of 455 MT iron based on the following:

1. Extrapolation of reported 1965 and 1967 usage for years 1956 through 1964 (153 MT Fe, 34 percent).
2. Reported 1965 through 1982 usage (106 MT Fe, 23 percent).
3. Reported 1983 through 1989 usage (23 MT Fe, 5 percent).
4. Estimated corrosion 1956 through 1989 usage (178 MT Fe, 38 percent).

The process wastes of PUREX Plant, B Plant, and tank farms wastes from 1956 through 1982 are combined in Table 5.10-8 for comparison purposes. The individual process contributions in the HDW model (Agnew et al. 1996) were not developed by this report. The 1956 through 1983 value of 397 MT Fe is determined by the difference of the total Fe (1,835 MT Fe, Agnew et al. 1996) and the sum of other wastes discussed in this section (1,438 MT Fe).

Table 5.10-8. Estimated Amount of Iron in PUREX Process Wastes, MT.

Years	Estimated iron content, MT Fe			
	Present estimate			HDW model (Agnew et al. 1996)
	Fe(NH ₂ SO ₃) ₂ chemical use	Corrosion	Total	
1956 - 1964	153	-	-	-
1964 - 1982	106	-	-	-
Subtotal, 1956 - 1982	259	142	403	397
1983 - 1989	23	36	59	172
Total	282	178	462	569

HDW = Hanford Defined Waste.

5.10.2.5 B Plant Process Wastes. No Fe containing chemical additions were identified for B Plant and tank farms from 1965 through 1982 from monthly essential material usage records (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Chemical corrosion in B Plant is included in the PUREX Plant corrosion developed by Appendix H.

5.10.2.6 Other Cribbed Wastes. Agnew et al. (1996) defines portions of waste streams other than the bismuth phosphate and UR process wastes discussed above as having been cribbed. These streams are identified in Agnew et al. (1996) as P1, CWP1, OWW1, DW, T1-SltCk, and WTR. The fraction of the streams cribbed is given in Appendix D, pages D-49 and D-53 (Agnew et al. 1996). The total iron in the streams is given by the volume and the concentration. The summed products of these three values give a total 32 MT calculated amount of Fe cribbed.

This report's analysis is based on the reported cribbed iron given by Waite (1991). The cribbed iron content in bismuth phosphate and UR process wastes given in the previous sections includes all Fe cribbed in the common crib and specific retention sites. Therefore, the other cribbed wastes identified in Agnew et al. (1996) are included in the bismuth phosphate or UR process values identified by Waite (1991) and no separate value is identified by this report's analysis.

5.10.2.7 N Reactor Decontamination Wastes. Agnew et al. (1996) defined the residual amount of N Reactor decontamination wastes in the DSTs after a single vault of phosphate/sulfate waste grout was produced. The N Reactor decontamination waste stream, (N) is defined in Appendixes B and D pages B-4, B-20, and D-53 (Agnew et al. 1996) with composition attributed to Lucas (1989). The resulting residual N Reactor Decontamination Hanford Site tank inventory is 12 MT Fe.

5.10.3 Best-Basis Estimate for Iron Inventory Value

Table 5.10-9 provides the results from this evaluation for iron, together with comparable estimates from other sources. The best-basis global estimate for Hanford Site tank waste is 1,230 MT of Fe. This estimate is significantly lower than the HDW model estimate of 1,835 MT Fe (Agnew et al. 1996) and significantly higher than the current value of 793 MT in the TWRS reference chemical inventory (Shelton 1996).

Table 5.10-9. Estimated Amount of Iron in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
Bismuth phosphate process wastes in SSTs/DSTs	418	488	NA
UR wastes in SSTs/DSTs	275	410	NA
REDOX	58	388	NA
PUREX and B Plant (1956-82)	403	397	NA
Removals from cribbed wastes	-	(32)	NA
N Reactor decontamination	12	12	NA
PUREX (1983-89)	59	172	NA
Total	1,230	1,835	793

DST = Double-shell tanks

HDW = Hanford Defined Waste

NA = Not attributed to individual waste streams

PUREX = Plutonium-Uranium Extraction

REDOX = Reduction and Oxidation.

SST = Single-shell tank

TWRS = Tank Waste Remediation System

UR = Uranium recovery.

An independent sample-based projection of the SST Fe inventory was developed by Colton (1997). Colton developed a projected SST inventory of 1,244 MT Fe based on 41 SST samples classified by waste type. The DST sample based iron inventory is 162 MT Fe (Shelton 1996). Thus, the total SST/DST inventory based on available sample data is 1,406 MT Fe.

The present estimate is based on reported chemical consumption for REDOX, PUREX, and B Plants, reported crib discharges for bismuth phosphate and UR processes, and flowsheet values of chemical consumption for the bismuth phosphate and UR processes. The HDW model is based on flowsheet waste compositions and waste transfer records from Hanford Site processing plants. The TWRS inventory is based on the 1986 EIS estimate of Hanford Site tank inventory in 1983 plus additions to the tank system after 1983.

The process wastes of PUREX Plant, B Plant, and tank farms wastes from 1956 through 1982 are combined in Table 5.10-9 for comparison purposes. The individual process contributions in the HDW model (Agnew et al. 1996) were not developed by this report. The 1956 through 1983 value of 397 MT Fe is determined by the difference of the total Fe (1,835 MT Fe, Agnew et al. 1996) and the sum of the individual streams discussed in previous sections (1,438 MT Fe).

The current Hanford Site tank waste inventory estimate of 1,230 MT Fe has the highest uncertainty in the residual wastes produced by the bismuth phosphate and UR processes. These wastes represent 57 percent of the estimated total. Both the current estimate and the HDW model are based on flowsheets for additions and estimated crib discharges for the bismuth phosphate and UR processes.

Corrosion in REDOX, PUREX and B Plants, and in N Reactor represent 24 percent of the estimated total. These corrosion values were developed by extrapolation of available analytical data (Appendix H).

Reported use of Fe precursor chemicals in REDOX and PUREX Plants represent 19 percent of the estimated total. This represents significantly lower uncertainty than the previous two categories of analysis.

Additional tank sampling and analysis of sludges will significantly reduce the uncertainties associated with Fe inventories.

5.11 LANTHANUM

Lanthanum added to the waste tanks was primarily from four sources:

(1) lanthanum nitrate added during 224 Building plutonium purification in the bismuth phosphate process, (2) reactor fission products, (3) strontium scavenging at B Plant in 1983 and 1984, and (4) PUREX process transuranic removal from cladding waste after March 1985. Minor amounts were also used at various Hanford Site analytical laboratories.

5.11.1 Identification of Lanthanum Values

Earlier estimates of lanthanum were based on the bismuth phosphate process flowsheet or technical manual, waste volume transfer records, or estimates of fission products. Table 5.11-1 compares some of the reported values.

Table 5.11-1. Reported Lanthanum Inventories.

Reference	Inventory (MT)	Comment
Tank Waste Remediation System Baseline (Shelton 1996)	23	TRAC for SSTs Samples for DSTs
Hanford Defined Waste model (Agnew et al. 1996)	40.4	
<i>Hanford Defense Waste Final Environmental Impact Statement</i> (DOE 1987)	NA	
TRAC (Jungfleisch 1984)	1.8	Reports as 1.33 E+04 gram moles. Through 1980 only.
Allen (1976)	NA	

DST = Double-shell tank

SST = Single-shell tank

TRAC = Track Radioactive Components.

The TWRS baseline combines SST inventories based on the TRAC (Jungfleisch 1984) estimate with the DST inventories based on waste samples. TRAC did not address the 224 Building wastes and includes only lanthanum from fission products. The DST inventory of 21.4 MT is much higher than would be predicted from the PUREX and B Plant process inputs since 1983 (see following discussion). The lanthanum inventory for Tank 241-AY-101 is stated to be 16 MT, which is unlikely given the lack of an identifiable chemical source.

The HDW model (Agnew et al. 1996) cites an abbreviated operating period for the bismuth phosphate process 224 Building operation (1952 to 1956) and an extremely low waste volume (27 percent of that predicted from the Schneider [1951] flowsheet [see Appendix C]). Lanthanum waste concentrations taken from Lucas (1989), which for 224 Building wastes are based on the bismuth phosphate process technical manual, are only 37 percent of the Schneider (1951) flowsheet. Yet the predicted lanthanum inventory for the 10 tanks receiving the 224 Building waste is 60 percent of that predicted from the Schneider (1951) flowsheet compositions and waste volumes. The HDW model may have a mathematic flaw in this area, because the global inventory is reasonable. Additionally, up to 25 percent of the 224 Building lanthanum was routed to Tanks 241-B-361 and 241-T-361 and are not included in the tank waste inventory (see the following discussion).

5.11.2 Lanthanum Inventory Evaluation

5.11.2.1 Bismuth Phosphate Plant Chemical Additions. A spreadsheet, also based on the 1951 bismuth phosphate process flowsheet (Appendix C), predicts 60 MT lanthanum added to the underground storage tanks from flowsheet chemical additions and flowsheet waste volumes. The Schneider (1951) flowsheet document was issued after 7 years of operation and is felt to be the best data source available. The lanthanum usage was 2.7 times the 1944 technical manual value (GE 1944), but there are indications in the flowsheet document that lanthanum usage rate had increased even further by the time the Schneider (1951) flowsheet was issued.

The waste from the 244 T and B Buildings was originally routed to tanks 241-T-361 and 241-B-361, respectively, and in turn overflowed to dry wells or cribs. Lanthanum would have been present as an insoluble chemical, and most should have settled out in the tanks.

Tank 241-T-361 was active from 1944 to 1947 and Tank 241-B-361 was active from 1945 to 1947 (Freeman-Pollard 1994). These concrete tanks are not part of the DST or SST systems. A very rough estimate based on the MTU processed during those years indicates that up to 25 percent of the lanthanum (15 of the 60 MT) could remain in these two tanks or the associated dry wells/cribs.

5.11.2.2 B Plant Strontium Recovery Usage. B Plant used a rare earth co-precipitation process during 1983 and 1984 to scavenge ⁹⁰Sr from strontium purification wastes (Joyce 1984). Both sulfate precipitation and caustic strike purification wastes were treated to reduce waste losses. Small quantities of a 2.5M rare earth nitrate solution (75 percent lanthanum nitrate/25 percent neodymium nitrate) were added to each waste batch and precipitated as a sulfate.

Compilation of engineering work instructions and procedure data sheets provides the best current estimate of the lanthanum added. Approximately 10,250 kg (22,600 lb) of the 2.5M solution (density 1.57 g/ml) were added. The total lanthanum addition was approximately 1.7 MT.

5.11.2.3 PUREX Process Decladding Waste Usage. The PUREX process also used a rare earth precipitation process to reduce the TRU levels in the Zirflex decladding wastes from March 6, 1985, until plant shutdown. The rare earth nitrate mixture used at the PUREX Plant was predominately a 1.675M lanthanum nitrate and 0.8325M neodymium nitrate, although the B Plant rare earth nitrate mixture was used for the process test (Walser 1985). Approximately 28.4 L (7.5 gal) were added to each batch of decladding waste (WHC 1990), and a second addition was usually made before centrifuging the second portion of the decladding waste batch after July 1987 (Herting et al. 1988). Additionally, until September 1986, actual usage of rare earth nitrate was only 60 percent of that which was specified in

the procedure (Herting et al. 1988). The estimated rare earth nitrate usage is given in Table 5.11-2.

Table 5.11-2. Addition of Rare Earth Nitrate to the PUREX Process Decladding Wastes.

Time period	Number of dissolver charges	Estimated rare earth nitrate usage
3/85 (process test)	10	22 L/charge
8/85 - 8/86	133	17 L/charge
3/87-5/87	4	0
7/87 - 12/89	64	57 L/charge

PUREX = Plutonium-uranium extraction.

A rough calculation indicates 220 L of 1.875M lanthanum and 5,910 L of 1.68M lanthanum were used. The total lanthanum added to the decladding wastes is approximately 1.4 MT. This is somewhat less than the 2.5 MT found in the Aging Waste tanks even after accounting for fission product lanthanum and the initial sludge heel in the tank. It is, however, apparent that the PUREX process usage was relatively small compared the bismuth phosphate process usage.

5.11.2.4 Lanthanum Fission Product. Fission product lanthanum contributes a small amount to the tank waste inventory. TRAC (Jungfleisch 1984) estimated this inventory at 1.84 MT through 1980. Re-estimation of the lanthanum fission product inventory results in a slightly higher value. Lanthanum inputs are estimated at 2.6 MT for fuel reprocessed through 1972 (RIBD) and 0.19 MT for the post-1983 PUREX Plant operation (ORIGEN2), for a total of 2.8 MT of lanthanum. An independent ORIGEN2 calculation, reported in Section 7.2, estimates a total lanthanum fission product production of 2.98 MT.

5.11.2.5 Minor Lanthanum Contributions. Chemicals containing lanthanum were used at various Hanford Site laboratories (Klem 1990), but the quantities included in laboratory waste transfers would be expected to be negligible.

5.11.3 Best-Basis Estimate for Lanthanum Inventory

The best-basis global lanthanum inventory is 51 MT from all sources. The largest uncertainty in this value results from the differences between the 1944 bismuth phosphate process technical manual (GE 1944) and the Schneider (1951) flowsheet, and in the solubility of the lanthanum compounds. Any future improvement to this estimate will require tank specific data.

5.12 LEAD

The aluminum-clad uranium metal fuel used at the Hanford Site contained significant amounts, e.g., >200 MT, of lead. The cladding and Al-Si bond in such fuels also contained smaller amounts of lead. Very small amounts of lead were also present in the Zircaloy®-clad fuel reprocessed at the Hanford Site.

Additional amounts of lead were introduced into the Hanford Site underground tanks from a lead sulfate carrier precipitation process used at the Hanford Site PUREX Plant and B Plant to separate and concentrate ⁹⁰Sr from PUREX process HLW in the 1960's.

From its known chemistry, lead introduced into many Hanford Site SSTs is expected to have concentrated in the sludge phase in these tanks. Selection, development, and testing of candidate glass formulations for vitrification of Hanford Site tank sludges require satisfactory knowledge of the total (global) and tank-by-tank amounts of lead present in the tank sludges.

5.12.1 Identification and Comparison of Lead Inventory Data

The HDW model, Rev. 3 (Agnew et al. 1996) predicts the Hanford Site tank waste to contain 279 MT of lead. The current TWRS baseline reflects the presence of 34.5 MT of lead in both the DSTs and SSTs. As discussed more fully later, it appears likely that the present TWRS baseline does not account for the large amount of lead introduced into tank wastes from decladding and dissolution of aluminum-clad fuels.

5.12.2 Lead Inventory Evaluation

The following sections summarize the major sources of lead in tank wastes.

5.12.2.1 Lead From Fuel Fabrication. Section 7.0 of this report provides a detailed and comprehensive account of the various elements, including lead, which were introduced into the Hanford Site reactor fuel elements during the fuel fabrication process. Such constituents eventually reported to various process waste streams and to the underground tanks.

From the discussion in Section 7.0, specifically Table 7.1-1, the amounts of lead introduced during fabrication of aluminum-clad fuel were as follows:¹

Intermetallic layer	213.00 MT
Al-Si bond	1.93 MT
AL jacket	0.25 MT
Total	220.00 MT

Also, as indicated in Section 7.0, only very little lead was introduced into Zircaloy®-clad fuels. Data in Section 7.0 indicate that no more than 0.04 MT of lead was present in all the Zircaloy®-clad fuels processed at the Hanford Site.

5.12.2.2 Lead From Strontium-90 Recovery Operations.

PUREX Plant. In the 1960's there was great incentive for the Hanford Site to supply megacurie amounts of purified ⁹⁰Sr for use in the United States Defense Programs. To meet this need, Hanford Site scientists developed a Lead Sulfate Carrier Precipitation process (Bray and Van Tuyl 1961) for recovery of ⁹⁰Sr from concentrated PUREX Plant first-cycle waste (1WW solution). This process involved the following:

- Add a mixture of 2M Na₂SO₄ and 1.7M tartaric acid to the 1WW solution
- Add 50 wt% NaOH solution to adjust the pH in the range of 1.5 to 2.5
- Digest at 80 °C for an hour
- Centrifuge
- Further treat the lead sulfate precipitate to obtain a nitric acid solution of ⁹⁰Sr free of sulfate, lead, and most other metal impurities.

The PbSO₄ precipitation process was applied in 1960 and 1961 to obtain about a megacurie of ⁹⁰Sr "crude concentrate." In plant-scale operation, 340 L of 0.5M Pb(NO₃)₂ (35 kg of lead) were added to each 3,800 L batch of concentrated PUREX process HLW (1WW solution). The average ⁹⁰Sr concentration in the 1WW solution in 1960 and 1961 was about 6.34 Ci/L (Van Tuyl 1958, Appendix F of this report). Thus, assuming an overall recovery of 55 percent (Bray and Van Tuyl 1961), 76 batches (3,800 L each) of 1WW

¹The "less than" signs used in Section 7.0 for amounts of lead introduced during the fuel fabrication process are deliberately omitted in this section. This practice allows statements of the maximum global lead inventory. Development of glass formulations which will satisfactorily accommodate the maximum global lead inventory is desirable to ensure engineering conservatism.

solution would have to have been treated with 2,650 kg (2.65 MT) of lead to recover one megacurie of ^{90}Sr (calculational details are shown in Appendix F of this report).

Additional use of the PbSO_4 precipitation process was made in the Hanford Site PUREX Plant during the period of 1961 through 1964 to recover another 8 MCi of ^{90}Sr . As a result of further laboratory development efforts and of the initial plant-scale experience, overall ^{90}Sr recovery was increased from 55 to 90 percent. It was also possible to decrease lead nitrate usage from 35 kg of lead per 3,800 L batch of waste feed to 35 kg per 5,700 L batch of PUREX process acid waste. The net result was that 277 batches (5,700 L each batch) were treated with 8,640 kg (8.64 MT) of lead to recover an additional 8 MCi of ^{90}Sr . (Calculational details are shown in Appendix F of this report.)

Overall, during the period of 1960 through 1964, 11.3 MT of lead were used in the PUREX Plant to recover 9 MCi of ^{90}Sr .

B Plant. In the mid to late 1960's further use of the PbSO_4 carrier precipitation process was made in the Hanford Site B Plant to provide "strontium crude" for follow-on isolation and purification of its ^{90}Sr content. Allen (1976) reported that during the period of 1965 through 1970, operation of the PbSO_4 carrier precipitation process in both the PUREX and B Plants consumed 16.1 MT of lead nitrate, equivalent to 10 MT of lead. There is presently some uncertainty about the use of $\text{Pb}(\text{NO}_3)_2$ in B Plant past 1972. Further evaluation is necessary to determine if additional lead was used.

5.12.3 Best-Basis Estimate for Lead Inventory Value

Data provided to date in Section 5.12.2 accounts for 241.3 MT of lead:

- From fuel fabrication - 220 MT
- From PbSO_4 precipitation process - 21.3 MT. Further evaluations are ongoing to determine if additional lead was used at B Plant for ^{90}Sr recovery.

Therefore, the best-basis estimate for lead in Hanford Site waste tanks is the HDW value of 279 MT.

5.13 MANGANESE

Large amounts of manganese were introduced into the Hanford Site waste tanks as the result of the extensive use of KMnO_4 in the bismuth phosphate, REDOX, and PUREX processes (see Section 5.18). Additional small quantities of manganese were introduced into aluminum- and Zircaloy®-clad fuels during the fuel fabrication process (see Section 7.0).

Manganese can exist in aqueous solution in several oxidation states, but under the alkaline conditions prevailing in Hanford Site tanks, it is expected that some, perhaps most, of the Mn (VII) (from KMnO_4) and Mn(0) (from fuels) both converted to Mn(IV) and precipitated as $\text{MnO}_2 \cdot \text{XH}_2\text{O}$. Selection, development, and testing of candidate glass formulations for vitrification of the Hanford Site tank sludges require satisfactory knowledge of the total (global) manganese content in Hanford tank waste.

5.13.1 Identification and Comparison of Manganese Inventory Data

The HDW model, Rev. 3 (Agnew et al. 1996) predicts the Hanford Site waste tanks to contain only 38.7 MT of manganese. The current TWRS inventory (Shelton 1996) reflects the presence of 149 MT of manganese distributed between the SSTs (121 MT) and DSTs (28 MT).

5.13.2 Manganese Inventory Evaluation

The following sections summarize the evaluation of flowsheet data and analytical information to arrive at a provisional best-basis tank inventory.

5.13.2.1 Use in Fuel Reprocessing Plants. Manganese (VII), in the form of KMnO_4 , was routinely used in the bismuth phosphate, REDOX, and PUREX Plants. In the latter plant KMnO_4 was used as a constituent of the aqueous solvent wash solution. In the REDOX Plant KMnO_4 was used from plant startup in 1952 until September 1959 to oxidize fission product ruthenium to volatile RuO_4 in a head-end oxidation process. And, in the bismuth phosphate process, KMnO_4 was added to oxidize Pu(IV) to Pu(VI).

Historical records (see Section 5.18) indicate that manganese was always added as KMnO_4 in the various separations plants. Table 5.13-1, adopted from data in Table 5.18-1, provides an estimate of the amounts of manganese added in the bismuth phosphate, REDOX, and PUREX Plants. The historical data show that a total of 101.8 MT of manganese were added during operation of the fuels reprocessing plants.

As noted in Section 5.18.2.3, large volumes in liquid wastes from the bismuth phosphate process, after prior treatment in the metal recovery plant, were routed to cribs. Thus, much of the soluble potassium from KMnO_4 , added in the bismuth phosphate process, eventually reported to one or more cribs. However, there is no reason to believe, or any evidence to show, that the manganese fraction of the added KMnO_4 was cribbed. Rather, it is believed that the manganese became part of the sludge fraction of the tank waste and is still present in the SST and DST wastes. The fraction manganese precipitated in the HDW model (Agnew et al. 1996, Appendix B) appears to be conservatively low based on the known chemistry of manganese in alkaline solutions.

Table 5.13-1. Estimates of Manganese Added to Hanford Waste Tanks from Separations Plant Operations.^a

Plant	Chemical added	Flowsheet usage Kg Mn (MTU)	Uranium processed (MTU)	Total manganese ^b (MT)
Bismuth phosphate	KMnO ₄	4.09	7.80E+03	31.9
REDOX	KMnO ₄ (High)	3.85	1.13E+04	50.0
	KMnO ₄ (Low)	2.82	1.13E+04	36.7
PUREX (U-Al)	KMnO ₄	0.32	6.11E+04	21.0
PUREX (U-Zr, through 1972)	KMnO ₄	0.45	1.34E+03	0.656
PUREX (U-Zr, 1983 through 1989)	KMnO ₄	0.45	3.89E+03	2.86
PUREX (Th)	KMnO ₄	--		2.02
Total manganese added to tank waste				101.8

PUREX = Plutonium-uranium extraction

REDOX = Reduction and oxidation

^aAdapted from Table 5.18-1; references provided in Table 5.18-1

^bTakes into account average ratios listed in Table 5.18-1.

5.13.2.2 From Fuels Fabrication. Section 7.0 of this report provides a detailed account of the various elements, including manganese, that were introduced into the Hanford Site reactor fuel elements during the fuel fabrication process. Such constituents eventually reported to various process waste streams and to the underground tanks. From the discussion in Section 7.0 the maximum amounts of manganese which were introduced during fabrication of aluminum-clad fuel were as follows:

Uranium Core	2.33 MT
Al-Si Bond Layer	0.29 MT
Al jacket	0.76 MT
Total	3.4 MT

Also, as indicated in Section 7.0, only very little manganese was introduced into Zircaloy®-clad fuels. Data in Section 7.0 indicate that no more than 0.16 MT of manganese was present in all the Zircaloy®-clad fuels processed at the Hanford Site.

5.13.3 Best-Basis Estimate for Manganese Inventory Value

Data provided in Sections 5.13.2.1 and 5.13.2.2 account for 105 MT of manganese:

From process use of KMnO_4 :	101.8 MT
From fuel fabrication:	3.6 MT

As noted earlier, the HDW model predicts only 38.7 MT of manganese to be present in the Hanford Site tanks. The current TWRS inventory value of 149 MT of manganese, although in better agreement with the 105 MT value determined in this report, appears to be too high. Thus, the best-basis global manganese inventory value is 105 MT.

5.14 MERCURY

Mercury was introduced into Hanford Site underground tanks from two principal sources:

- Use of $\text{Hg}(\text{NO}_3)_2$ in the PUREX and REDOX Plants to catalyze nitric acid dissolution of certain unirradiated Plutonium Recycle Test Reactor (PRTR) aluminum-plutonium alloys and also certain other reactor aluminum alloy fuels (Matheison and Nicholson 1972a, Hanson 1962).
- Use of $\text{Hg}(\text{NO}_3)_2$ in the BiPO_4 process (B and T Plants), REDOX process, and PUREX process to suppress volatilization of radioiodine (^{129}I and ^{131}I) during nitric acid dissolution of short-cooled (less than 90 days) irradiated uranium metal fuel.

From known mercury chemistry, most of the $\text{Hg}(\text{II})$ in the acidic BiPO_4 , REDOX, and PUREX process wastes precipitated as $\text{HgO} \cdot x\text{H}_2\text{O}$ when the wastes were made alkaline. However, no matter where it is present in the existing tank wastes, i.e., sludge and/or salt cake, mercury will present special problems during high temperature vitrification of both the HLW and LLW fractions. Mercury is easily reduced to the metallic state. Metallic mercury so produced may interfere with operation of joule-heated melters. However, more likely, at the high temperatures prevailing in vitrification operations, mercury will readily volatilize and seriously complicate offgas handling equipment and procedures. Knowledge of the amount and location of mercury in the Hanford Site tanks is, thus, essential.

Since mercury is also classified as a toxic metal (*Resource Conservation and Recovery Act of 1976 [RCRA]*), disposition of any mercury condensed from melter offgases presents additional problems.

5.14.1 Identification and Comparison of Mercury Inventory Values

The HDW model, Rev. 3 (Agnew et al. 1996) predicts the Hanford Site tanks to contain 6.86 MT of mercury. Of this total, 5.81 MT are predicted to be in the SSTs and 1.05 MT in the DSTs. Information in Agnew et al. (1996) indicates the basis for the 6.86 MT inventory value is information originally reported by Jungfleisch (1984).

Allen (1976) estimated that 5,000 moles of $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$ (1.0 MT of mercury) were added to the underground tanks during the period of 1944 through 1975. Allen's estimate is known to be low since it does not account for usage of Hg(II) in the BiPO_4 process (B and T Plants). Also, it does not appear to take into account use of mercuric nitrate in the REDOX and PUREX Plant for dissolution of aluminum-alloy fuels.

The TWRS database presently does not include data for the global inventory of mercury. However, the data package (RHO 1985a) assembled for preparation of the *Hanford Defense Waste Environmental Impact Statement* (DOE 1987) indicated the presence of 900 kg (0.9 MT) of mercury in the SSTs. Recent analyses of samples taken from the DSTs indicate that these tanks contain a total of 0.14 MT of mercury. Table 5.14-1 compares mercury data from several sources.

Table 5.14-1. Currently Reported Inventories of Mercury in Hanford Site Tanks.

Tanks	HDW model (Rev. 3) data ^a	Environmental Impact Statement and analytical data
Double-shell tanks	1.05 MT	0.14 MT ^b
Single-shell tanks	5.81 MT	0.9 MT ^c
All	6.86 MT	1.0 MT

HDW = Hanford Defined Waste

^aAgnew et al. (1996)

^bFrom analyses of samples from the double-shell tanks

^cRHO (1985a).

5.14.2 Mercury Inventory Evaluation

The following sections summarize the major sources of mercury in tank waste.

5.14.2.1 Sources of Mercury Introduced Into Tanks.

Dissolution of Al-U and Al-Pu Alloy Fuels. In 1963 the REDOX Plant processed four dissolver charges containing PRTR Al-Pu alloy. The REDOX Plant used the flowsheet from Hanson (1962) to process these charges. Each charge involved use of 0.00462 MT of mercury (as $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$). Dissolution of the four charges of Al-Pu alloy involved use of 0.018 MT of mercury.

In 1972, the Hanford Site PUREX plant reprocessed small amounts of various unirradiated Al-U and Al-Pu alloys including Zircaloy®-clad PRTR fuel assemblies and loose rods, Light Water Pressure Reactor (LWPR) fuel elements, thin Phoenix reactor fuel plates and small disks, and miscellaneous aluminum-alloy fuel samples. Chemical flowsheets for these dissolution campaigns were prepared by Matheison and Nicholson (1972a).

According to the Matheison and Nicholson (1972a) flowsheets, ten separate Al-U and Al-Pu alloy charges were dissolved in nitric acid using added $\text{Hg}(\text{NO}_3)_2$ to catalyze the dissolution. Five of the charges each used 20 kg (44 lb) of $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$, two charges each used 22 lb of $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$ while the final three charges each used 6.8 kg (15 lb) of $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$. Hence, a total of 140 kg (309 lb) of mercuric nitrate were used in reprocessing the Al-alloy material. This quantity of Hg(II) nitrate corresponds to 409 moles of mercury or 82 kg (0.082 MT). For the purposes of this report, it is assumed that actual PUREX Plant dissolution of the Al-alloy fuel material was performed in close compliance with the published flowsheets.

5.14.2.2 Suppression of Radioiodine Volatilization. It was known before chemical reprocessing of irradiated fuels began at the Hanford Site that control of the release of radioactive iodine, e.g., ^{129}I and, especially, ^{131}I , would be important. Thus, plutonium separations processes were designed to minimize the environmental effects of any released radioiodine by allowing ^{131}I ($t^{1/2} = \text{ca. } 7 \text{ days}$) to decay to negligible levels before fuel reprocessing by maximizing the atmospheric dilution of any emitted radioiodine, and by performing fuel dissolution operations during optimal atmospheric conditions. To further suppress evolution of radioiodine, starting in about 1951, Hg(II) nitrate was routinely added to the nitric acid dissolvent in the B and T Plants and later, for a time at least, in the REDOX and PUREX Plants. Since Hg(II) forms a strong complex with iodide ion, it was expected (Holm 1951) that the addition of Hg(II) nitrate during the plutonium reduction step in the BiPO_4 process (B and T Plants) would react with radioactive iodine and, thereby, decrease its volatilization.

B and T Plant Experience. Schneider (1951) states that in the BiPO_4 process, 113.4 kg (250 lb) of 0.385 wt% $\text{Hg}(\text{NO}_3)_2$ solution were added during processing of 997.9 kg (2,200 lb) of uranium metal. Per Appendix B, 3,683.2 MT of uranium metal were processed through B and/or T Plants during the period of 1951 through 1956. This resulted in the use of 1611.4 kg of Hg(II) nitrate which contained 1.0 MT of mercury.

In the HDW model Rev. 3, Agnew et al. (1996) estimates that wastes generated in the BiPO_4 process over the time this process was used contained 0.48 MT of mercury.

REDOX and PUREX Plant Experience. Historical records (Beard 1970) indicate that Hg(II) nitrate was occasionally used at both the REDOX and PUREX Plants to suppress volatilization of radioiodine during dissolution of short-cooled fuels. During these periods, which are estimated to be about 25 percent of the time before 1968, 0.8 kg (1.7 lb) of $\text{Hg}(\text{NO}_3)_2$ were added for every six tons of uranium dissolved. The practice of adding Hg(II) nitrate for iodine evolution control was discontinued at the PUREX Plant in 1968.

Available data (Allen 1976) indicate 1,633 kg (3,600 lb) of $\text{Hg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$ were used at the REDOX and PUREX Plants during the period from 1952 through 1967. This amount of Hg(II) nitrate corresponds to 960 kg (0.96 MT) of mercury.

In the HDW model Rev. 3, Agnew et al. (1996) estimates that REDOX process wastes contained 1.21 MT of mercury and that PUREX process wastes contained 5.29 MT of mercury. These latter estimates appear to assume that Hg(II) nitrate was used continuously rather than occasionally in both REDOX and PUREX Plant operation. Furthermore, Agnew et al. (1996) also states that Hg(II) nitrate was used to suppress volatilization of radioiodine during the entire time the Hanford Site PUREX Plant was operated. Historical records indicate that such usage was discontinued in 1968.

5.14.2.3 Other Sources of Mercury. Personal communications with various active and retired Hanford Site scientific personnel discloses that it was once common practice to discard small amounts of contaminated and potentially-contaminated mercury metal from manometers and other instruments to aqueous wastes, which eventually were routed to the underground tanks. The best recollection of many of the involved personnel is that direct disposal of mercury metal occurred only infrequently and that only a very small amount of mercury was ever disposed of in this manner.

5.14.3 Best-Basis Estimate for Mercury Inventory Value

The evaluations described in Section 5.14.2 accounts for a total of 2.1 MT of mercury in all the Hanford Site tanks (Table 5.14-2). This latter value, although substantially lower than the 6.86 MT inventory figure carried in Agnew et al. (1996), is taken to be the best-basis global inventory value for mercury.

Table 5.14-2. Best-Basis Estimate of Mercury Inventory.

Source	Mercury
Dissolution of Al-alloy fuels	0.1 MT
Suppression of iodine emission:	
B and T Plants	1.0 MT
REDOX and PUREX Plants	0.96 MT
Total	2.1 MT (rounded)

PUREX = Plutonium-Uranium Extraction
REDOX = Reduction and oxidation.

5.15 NICKEL

Nickel is a component of many of the water-insoluble solids (sludges) present in Hanford Site tank wastes. Pretreatment of retrieved sludges before their vitrification with dilute NaOH washes to remove soluble aluminum, phosphate, etc., will not remove nickel. Thus, conditions, e.g., melt formulation, waste loading, etc., for vitrifying nickel-bearing sludges must be controlled to permit production of glass meeting current specifications for nickel content.

Nickel resulted from three principal sources:

- As an impurity or additive in fuel and cladding
- Corrosion of 304-L stainless steel equipment
- From precipitation of nickel ferrocyanides from aged bismuth phosphate process wastes from 1954 to 1957.

The latter operation was performed both in-plant (U Plant) and in-tank to reduce the ^{137}Cs concentration of the aged wastes to a level that would allow their disposal to the soil (under then-existing rules), thereby freeing up needed tank space. Nickel ferrocyanide solids were originally deposited in 20 separate SSTs. Recent analyses of samples taken from these tanks show that nickel is present in the expected (from historical records) amounts.

5.15.1 Identification and Comparison of Nickel Inventory Values

Two currently reported global inventory values for nickel in the Hanford Site tanks are listed in Table 5.15-1. The data sources list nickel inventories separately for SSTs and DSTs, and these separate inventory values are included in Table 5.15-1. The global nickel inventories from the two data sources agree to within about 18 percent. However, as noted in Table 5.15-1, the HDW model predicts the DSTs to contain about 27.3 MT more nickel than the TWRS data and about 59.2 MT less nickel in the SSTs.

Table 5.15-1. Currently Reported Inventories of Nickel in Hanford Site Tanks (MT).

Tanks	TWRS data ^a	HDW model data ^b
Double-shell tanks	10.9	38.2
Single-shell tanks	203.0	143.8
Total	214.0	182

HDW = Hanford Defined Waste

TWRS = Tank Waste Remediation System

^aShelton (1996)

^bAgnew et al. (1996).

Several questions concerning the validity of the HDW model and TWRS nickel inventories must be answered, namely:

- Do both inventory databases include data relating to introduction of nickel into the tanks from:
 - Impurities in fuel and constituents of cladding
 - Precipitation of nickel ferrocyanide
 - Corrosion of stainless steel equipment?
- Do both inventory data bases use the same input inventory information?
- Do both inventory data bases include information concerning the inventory of nickel that may have been transferred to underground cribs?
- What is the basis in each data base for distributing the tank nickel inventory between the DSTs and SSTs?

5.15.2 Nickel Inventory Evaluation

The following sections summarize the major sources of nickel in tank waste, and provide an estimate of nickel discharges to cribs.

5.15.2.1 Sources of Nickel Introduced into Tanks.

Fuels and Cladding. Section 7.0 of this report provides an in-depth analysis of the inventories of minor components, including nickel, introduced into the Hanford Site tanks as a result of decladding and dissolution of aluminum-clad and Zircaloy[®]-clad fuels. For convenience, these data are summarized in Table 5.15-2.

Table 5.15-2. Inventory of Nickel in Aluminum- and Zircaloy®-Clad Fuel.

Component	Aluminum-clad fuel inventory, kg (MT)	Zircaloy®-clad fuel inventory, kg (MT)
Uranium fuel	9.3 E+03 (9.3)	≤5.70 E+02 (≤0.57)
Aluminum-silicon bond layer	3.3 E+02 (0.33)	
Aluminum cladding	2.01 E+04 (20.1)	
Braze filler		≤1.4 E+00 (0.0014)
Zircaloy® cladding		2.3 E+02 (0.23)
Total	2.97 E+04 (29.7)	≤8.1 E+02 (≤0.81)

There is no indication in either the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) or in the HDW model (Agnew et al. 1996) that account was ever taken of the nickel in either aluminum-clad or Zircaloy®-clad fuel processed at the Hanford Site.

Precipitation of Nickel Ferrocyanide. Stedwell (1957) states that 1.22 E+05 kg (2.70 E+04 lb) of nickel sulfate were used to precipitate nickel ferrocyanide from 1.2 E+08 L of Metal Recovery Process waste on an "in-plant" basis. Chemical flowsheets issued during the period (1954 through 1957) when nickel ferrocyanide scavenging operations were underway indicate nickel was added to the waste as an aqueous solution of NiSO₄ · 6H₂O. Accordingly, 1.22 E+05 kg of NiSO₄ · 6H₂O correspond to 4.66 E+05 moles of nickel equivalent to 27.4 MT of nickel. Borsheim and Simpson (1991) estimate that 4.58 E+05 moles of ferrocyanide (either as the potassium or the sodium salt) were added during in-plant scavenging operations. The equivalency of the nickel and ferrocyanide mole quantities are in agreement with the current belief that the precipitated compound was Na₂Ni[Fe(CN)₆].

Stedwell (1957) also stated that as of January 15, 1957, 1.6 E+04 kg of NiSO₄ · 6H₂O had been used to precipitate nickel ferrocyanide from 1.2 E+07 L of Metal Recovery Plant waste on an in-farm basis. In-farm nickel ferrocyanide scavenging operations continued through treatment of approximately 1.2 E+07 gal of waste in the 200 East Area; contrary to early expectations, candidate wastes in the 200 West Area were not scavenged with nickel ferrocyanide. Jeppson (1993) provides data relating to the amount of nickel sulfate used for in-farm scavenging purposes in the 200 East Area. According to information in Jeppson (1993) approximately 16.3 MT of nickel were used for this purpose.

Overall, 27.4 and 16.3 MT of nickel were used in the in-plant and in-farm scavenging operations, respectively. Thus, assuming that none of the nickel was eventually sent to cribs, some 43.7 MT of nickel from nickel ferrocyanide scavenging operations are presently stored in the Hanford Site tanks. Agnew et al. (1996) predicts a total of 46.5 MT of nickel in the tanks containing uranium recovery process wastes. Also note, however, that Agnew

et al. (1996) states that 33.6 MT of nickel were sent to cribs with scavenged UR and 1C bismuth phosphate process wastes (see Section 5.15.2.2).

Of interest from a historical point of view, Allen (1976) estimated that a total of $2.3 \text{ E}+05 \text{ kg}$ ($1.5 \text{ E}+06 \text{ moles}$) of nickel sulfate were added to the Hanford Site wastes from 1944 through 1975. This amount of nickel sulfate corresponds to 88 MT of nickel. Allen based his estimate upon Stedwell's data for in-tank scavenging and assumed, erroneously that nickel was added as anhydrous nickel sulfate rather than as the hexahydrate. Allen also projected Stedwell's (1957) data for in-farm scavenging completed to January 15, 1957, to expected future scavenging in both candidate 200 East and 200 West Area tanks. (The 200 West Area tanks were not subjected to nickel ferrocyanide scavenging.)

Further, Allen (1976) estimated that the sludge in the tanks receiving nickel ferrocyanide solids contained 500 MT of $\text{Ni}_2[\text{Fe}(\text{CN})_6]$. Using this formula, the amount of nickel ferrocyanide assumed to be in the tanks contains 176 MT, not 88 MT, of nickel. Personnel who prepared the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) in the 1985 to 1987 timeframe mistakenly carried forward the 176 MT (rounded to 180 MT) of nickel value. As noted earlier, the correct compound for the solids precipitated in the scavenging operations is now thought to be $\text{Na}_2\text{Ni}[\text{Fe}(\text{CN})_6]$.

Corrosion of Stainless Steel Equipment. Reprocessing of irradiated fuel from Hanford Site reactors was performed in stainless steel, principally 304-L, equipment. Corrosion of this equipment over its service life introduced fairly large amounts of iron, chromium, and nickel into the HLW sent to the underground tanks. The exact quantity of stainless steel corrosion products routed to the underground tanks is still controversial.

Various estimates of the amounts of corrosion-derived nickel in the Hanford Site tanks are available. For example, Agnew et al. (1996) discusses both a "low" and a "high" estimate. The latest, and what is judged to be the most accurate estimate of the inventory of nickel resulting from corrosion of stainless steel, is based upon the recent calculations for iron corrosion documented in Appendix H of this report. Table 5.15-3 provides a summary of nickel corrosion values calculated from data reported in Section 5.10 for inventories of corrosion-derived iron; stainless steel 304-L contains 71 wt% iron and 10 wt% nickel.

Table 5.15-3. Estimates of Amounts of Corrosion-Generated Nickel in Hanford Site Tanks.^a

Process	Nickel inventory, MT
PUREX	25.1 ^b
REDOX	1.2
Bismuth Phosphate	4.6
Uranium Recovery	6.3
Total	37.2

PUREX = Plutonium-uranium extraction

REDOX = Reduction and oxidation

^aBased upon corrosion data reported in Appendix H of this report

^bOperating Period: 5.1 MT Ni (1983 to 1989) and 20.0 MT Ni (1956 to 1972).

Van der Cook and Walser (1970) estimated that in the PUREX process operation for each MT of fuel processed about 1.2 kg of iron, over and above that added as process chemicals, were introduced into the process waste. About one-half of the 1.2 kg was attributable to corrosion of various items of process equipment and piping. Over the life of fuel reprocessing activities at the Hanford Site, $1.08 \text{ E}+08$ MT of fuel were reprocessed. If the Van der Cook and Walser (1970) estimate prevailed during reprocessing of all the fuel, the amount of "corrosion-derived" iron introduced into the tank wastes was 64.8 MT. Since nickel is only 10 wt% of the metal content of stainless steel 304-L, about 9.1 MT of "corrosion-generated" nickel was introduced into the tank waste.

Agnew et al. (1996) provides a somewhat higher estimate of corrosion-derived nickel. To account for the large amount of iron found by analysis in sludges, Agnew et al. (1996) suggests that as much as $1.2 \text{ E}+07$ mol (673 MT) of iron in the tank waste is attributable to corrosion of stainless steel. On this basis, about 94.8 MT of nickel could have been introduced into the tank wastes.

There is no evidence in the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) that any account was taken of corrosion-generated nickel in either the DSTs or SSTs.

5.15.2.2 Nickel Sent to Cribs. Agnew et al. (1996) estimates that over the lifetime of the Hanford Site project 33.6 MT of nickel were sent to cribs constructed in the Hanford Site soil. Any nickel that was indeed sent to cribs was likely present in various forms, e.g., particulate nickel ferrocyanide, precipitated hydrated nickel oxide, and as aqueous-soluble nickel ion.

From historical records, Waite (1991) reports that approximately $9.0 \text{ E}+04$ kg of ferrocyanide were discharged directly to the soil at the Hanford Site. (Waite does not

provide any data concerning the amount of nickel, if any, that might have been discharged to the soil.) Waite (1991) does not state if the discharged ferrocyanide was present as the free ion or combined with nickel into a solid form or both. If all the $9.0 \text{ E}+04$ kg of ferrocyanide was discharged to the solid as $\text{Na}_2\text{NiFe}(\text{CN})_6$ (very unlikely situation), about 26 MT of nickel would have also been discharged to the soil.

There is no firm basis whatsoever for determining the amount of nickel, if any, that was discharged to the soil. For purposes of this report, it is assumed that all the nickel originally input to the tanks is still there. This assumption is a conservative one since it maximizes the amount of nickel-bearing material that will have to be vitrified.

5.15.3 Best-Basis Estimate for Nickel Inventory Value

Depending upon the choice of the datum used for the amount of nickel introduced into the underground tanks as a result of the corrosion of stainless steel piping and equipment, several inventories for nickel in Hanford Site waste tanks can be calculated (Table 5.15-4).

The Case 2 and 3 nickel inventories bracket those currently carried in the TWRS data base and those predicted by the HDW model. However, Case 3 is rejected in favor of Case 2 as providing the best-basis estimate of the total Hanford Site tank inventory of nickel. Case 2 incorporates what is now thought to be the most accurate estimate of the amount of nickel introduced to the tanks from corrosion, dissolution of fuel and cladding, and nickel ferrocyanide scavenging operations. The best-basis inventory of nickel in the Hanford Site waste tanks is 111 MT.

Table 5.15-4. Possible Inventories of Nickel in Hanford Site Waste Tanks.

Component	MT Nickel		
	Case 1	Case 2	Case 3
From fuel and cladding	30.5	30.5	30.5
Precipitated as nickel ferrocyanide	43.7	43.7	43.7
Corrosion of stainless steel	9.1 ^a	37.2 ^b	94.8 ^c
Disposed of to cribs	0.0	0.0	(33.6)
Total in tanks	83.3	111.4	135.0

^aBased on Van der Cook and Walser (1970)

^bBased on data in Appendix H

^cBased on the Hanford Defined Waste model Rev. 4 (Agnew et al. 1996).

5.16 NITRATE AND NITRITE

Nitric acid (HNO_3) was the largest source of nitrate. It was used in the dissolvers at the processing facilities and sodium nitrate (NaNO_3) was used to suppress hydrogen generation during the decladding step of the Bismuth Phosphate, REDOX, and PUREX processes. Aluminum nitrate nonahydrate (ANN) used in the REDOX process to extract plutonium and uranium was another major source of nitrate. Lesser sources include: (1) cadmium nitrate, $\text{Cd}(\text{NO}_3)_2$, which was used as a neutron poison in plutonium transfers between the processing areas and solvent extraction at the PUREX Plant; and (2) lead nitrate ($\text{Pb}(\text{NO}_3)_2$) which was used at the PUREX Plant and B Plants for fission product recovery. Sodium nitrite (NaNO_2) was used as an additive to alkaline wastes to inhibit corrosion of carbon steel storage tanks. Nitrite is generated as a product in the radiolysis of nitrate. It was also used in the bismuth phosphate and PUREX processes to oxidize plutonium for extraction.

Due to its high solubility, nitrate and nitrite reside mostly in the salt cake layers and in the liquid phase of the tank waste. Due to reduction/oxidation reactions in the processes and radiolysis in the waste tanks, it is difficult to estimate the respective concentrations of nitrate and nitrite. Therefore, only the combined nitrate/nitrite inventory will be discussed here for now. The uncertainty associated with not knowing the individual nitrate and nitrite masses does not impact facility design and subsequent facility costs.

5.16.1 Identification of Nitrate/Nitrite Inventory Values

The HDW model and TWRS inventory values are summarized in Table 5.16-1. The HDW model, TWRS inventory, and this report values for total nitrate/nitrite are 58,900 MT, 117,000 MT, and 85,700 MT, respectively. Table 5.16-1 summarizes the breakdown between nitrate and nitrite reported values.

Table 5.16-1. Reported Nitrate/Nitrite Values (MT).

Reference	Nitrate (MT)	Nitrite (MT)	Nitrate + Nitrite (MT)
HDW (Agnew et al. 1996)	45,300	13,600	58,900
TWRS (Shelton 1996)	107,000	9,530	117,000
This report	-	-	85,700

HDW = Hanford Defined Waste
 TWRS = Tank Waste Remediation System.

5.16.2 Nitrate/Nitrite Inventory Evaluation

The nitrate/nitrite inventory can be estimated by computing the total amount of nitrate and nitrite present in each of the separations process wastes and subtracting the amount of each component that was discharged to cribs and trenches.

5.16.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process operated at T Plant from December 1944 through 1956 and at B Plant from April 1945 through 1952. The Hanford Site production records indicate 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the BiPO_4 processing is developed in Appendix C based on the Schneider (1951) flowsheet. Comparable HDW nitrate values are obtained from the "waste volume out" and "nitrate and nitrite concentrations" of the HDW. The HDW nitrate and nitrite concentrations used are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

The nitrate usage in the BiPO_4 processing is shown in Table 5.16-2 for the Schneider (1951) flowsheet (Appendix C) and for the HDW model (Agnew et al. 1996).

Table 5.16-2. Amount of Nitrate/Nitrite in Bismuth Phosphate Wastes.

Waste stream	Nitrate + Nitrite (MT)		Volume (m ³)	
	Schneider 1951	Agnew et al. 1996	Schneider 1951	Agnew et al. 1996
Metal waste	4,825	1,680	124,900	135,900
First cycle (1C)	9,900	5,550 ^a	107,100	121,600 ^a
Second cycle (2C)	6,720	6,300	109,800	120,000
224 Waste	7,520	3,080	114,300	31,400
Coating Waste	675	-	8,100	-
Total	29,640	16,500	464,000	409,000

^aIncludes Coating Waste.

Metal Waste. After decladding, the first steps in the BiPO_4 process dissolved the uranium metal slugs and extracted the plutonium by carrier precipitation with bismuth phosphate. Sulfuric acid was added to the solution to complex the uranyl ion (UO_2^{+2}) and avoid uranyl phosphate precipitation. This resulted in the metal waste stream. The metal waste stream contained the bulk of the uranium and approximately 90 percent of the long-lived fission products (e.g., ¹³⁷Cs and ⁹⁰Sr). The metal waste was neutralized with sodium hydroxide and sodium carbonate and was sent to the SSTs for storage.

The stored metal waste containing the uranium from BiPO_4 processing was later retrieved as the feed for the UR process.

Cribbed BiPO_4 Wastes. Portions of the other BiPO_4 waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The total nitrate discharged to cribs and specific retention trenches was 11,100 MT nitrate/nitrite in 259,000 m^3 (68.5 Mgal) (Waite 1991). This 11,100 MT nitrate/nitrite is 44.7 percent of the BiPO_4 wastes excluding the metal wastes. The nitrate/nitrite values reported in Waite are suspect as the nitrate/nitrite is approximately 10 percent low to be charge balanced with the other reported components. The solubility behavior of nitrate in the alkaline wastes should be similar to sodium. The Section 5.20, Sodium, analysis of the Waite report indicates that 70.0 percent of the sodium in the BiPO_4 wastes excluding the metal wastes content was cribbed.

The HDW model estimates 9,130 MT nitrate/nitrite was cribbed from the BiPO_4 process in 173,000 m^3 (45.8 Mgal) (Agnew et al. 1996). This is equivalent to 61 percent of the nitrate/nitrite in the BiPO_4 wastes excluding the metal wastes. The HDW estimate for BiPO_4 wastes is obtained from the "crib volume" and the "supernatant nitrate and nitrite concentrations" of Agnew et al. (1996). The nitrate/nitrite concentrations are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989). The higher percentage cribbed in the HDW model is due to the lower nitrate/nitrite quantity assumed in the 1C and 224 wastes (Table 5.16-2).

This analysis assumes that the sodium concentrations presented by Waite are more accurate than the nitrate analyses and uses 70 percent as the fraction of nitrate cribbed from BiPO_4 wastes excluding the metal wastes. This is equivalent to 17,580 MT nitrate/nitrite cribbed from BiPO_4 wastes.

Summary for BiPO_4 Process Wastes. The residual MW, 1C, and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.16-3. This report estimates the residual BiPO_4 tank wastes contain 7,530 MT of nitrate versus the 5,710 MT estimated by the HDW model.

Table 5.16-3. Estimated Amount of Nitrate/Nitrite in BiPO₄ Tank Wastes, MT.

BiPO ₄ process waste	Nitrate/nitrite, MT	
	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes produced	29,640	16,520
BiPO ₄ wastes cribbed	17,580	9,130
BiPO ₄ wastes sent to uranium recovery	4,825	1,680
BiPO ₄ wastes in single- and double-shell tanks	7,530	5,710

HDW = Hanford Defined Waste.

The principal differences in the present estimate and the HDW model are the input chemicals in the flowsheets assumed and the volume of cribbed BiPO₄ wastes.

5.16.2.2 Uranium Recovery Wastes. The UR process began in 1952 to extract uranium from the BiPO₄ process MW stored in B, BX, BY, C, T, TX, and U tank farms.

The design basis flowsheet for the UR process is provided in the UR Technical Manual (GE 1951a). The process operated with a highly variable feed due to variations in the quality of retrieved metal waste sludges and supernatants. The variation of uranium content during the retrieval operation resulted in variation of required nitric acid for dissolution and resulting sodium hydroxide for subsequent neutralization. As a result, the UR flowsheet (GE 1951a) is not expected to provide an accurate estimate of UR chemical usage.

Cribbed Uranium Recovery Wastes. The HDW model analysis of transfer records from the UR operation indicates that 157,500 m³ (41.6 Mgal) containing 26,700 MT nitrate/nitrite was cribbed. This reflects 62.5 vol% of the total UR waste streams being routed to cribs and 37.5 vol% of the total UR waste streams remaining in the SSTs. The HDW model estimate for UR wastes is obtained from the crib volume and the supernatant nitrate/nitrite concentration of Agnew et al. (1996). The nitrate/nitrite concentrations are attributed to Borsheim and Simpson (1991) and Sloat (1954).

Waite (1991), states that 155,000 m³ (41 Mgal) of scavenged UR wastes containing 31,900 MT of nitrate/nitrite in a total of 433,800 m³ (114.6 Mgal) containing 43,000 MT total nitrate/nitrite was discharged to cribs and specific retention trenches. The nitrate values in the Waite report are suspect as an additional 4,000 MT of nitrate is required to attain charge balance with the other components listed in the Waite chemical data. With adjustment of the total nitrate in the Waite data by an additional 4,000 MT and subtraction of the 17,580 MT nitrate/nitrite in cribbed BiPO₄ wastes, a net 29,400 MT of nitrate/nitrite is calculated to have been cribbed from UR wastes as shown below:

Total cribbed (Waite, 1991)	43,000 MT NO ₃ /NO ₂
Charge balance correction	<u>4,000</u> MT NO ₃ /NO ₂
Corrected total crib discharge	47,000 MT NO ₃ /NO ₂
BiPO ₄ wastes cribbed (Table 5.16-3)	<u>17,580</u> MT NO ₃ /NO ₂
Net UR wastes cribbed	29,400 MT NO ₃ /NO ₂

This analysis assumes that the UR nitrate discharged to cribs and specific retention trenches is equal to the 62.5 vol% split determined by the HDW model.

Uranium Recovery Chemicals Added. The assumption of 62.5 percent of the UR waste nitrate/nitrite being cribbed (29,400 MT nitrate/nitrite) results in a total UR waste nitrate content of 47,000 MT. The nitrate content of the UR waste feed and UR chemicals added are 4,825 MT and 42,200 MT, respectively.

For the HDW model the amount of nitrate added to tank wastes during UR is the difference in the nitrate content between the UR wastes produced and the BiPO₄ wastes sent to UR. The HDW model nitrate and nitrite values in the UR wastes produced (50,300 MT nitrate/nitrite) are obtained from the "waste volume out" and "nitrate and nitrite concentrations" of the HDW model. The HDW nitrate and nitrite concentrations used are attributed to Borsheim and Simpson (1991) and Sloat (1954). Subtraction of the nitrate/nitrite inventory in the UR feed (1,680 MT nitrate/nitrite) gives a 48,600 MT nitrate/nitrite chemical addition in UR.

Summary for Uranium Recovery Process Wastes. Table 5.16-4 provides a summary nitrate/nitrite material balance for the UR process for this analysis and the HDW model. This report estimates the residual UR tanked wastes contain 17,600 MT of nitrate/nitrite versus the 23,600 MT estimated by the HDW model.

Table 5.16-4. Estimated Amount of Nitrate/Nitrite in Uranium Recovery Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes sent to UR	4,825	1,680
UR chemicals added	42,200	48,600
UR wastes produced	47,000	50,300
UR wastes cribbed	29,400	26,700
UR wastes in single-shell and double-shell tanks	17,600	23,600

HDW = Hanford Defined Waste
UR = Uranium recovery.

The difference in analysis that provided the nitrate/nitrite results in Table 5.16-4 are as follows:

1. UR feed - Values reported in Section 5.16.2.1.
2. UR wastes produced.
 - a. Present estimate of UR wastes produced equals nitrate reported cribbed (Waite 1991) corrected for charge balance, and BiPO_4 wastes cribbed (Section 5.16.2.1), divided by 0.625 (the volume fraction of UR wastes cribbed [Agnew et al. 1996]).
 - b. The HDW model basis for UR wastes produced is Sloat (1954) and Borsheim and Simpson (1991).
3. UR chemicals added - the difference between (2) and (1).
4. UR wastes cribbed.
 - a. Present estimate based on Waite (1991) corrected for charge balance and BiPO_4 wastes cribbed (Section 5.16.2.1).
 - b. The HDW basis uses transfer records and calculated nitrate/nitrite solubility.

Because of the lack of definitive data on the chemical usage in UR and the nitrate content of UR sludges, there is a significant uncertainty in the amount of nitrate/nitrite associated with residual UR wastes in the waste tanks. The HDW basis for nitrate and nitrite content in cribbed UR wastes result in a greater fraction of nitrate/nitrite in the residual UR tank wastes, 47 percent versus 37 percent. The uncertainty in nitrate/nitrite content in residual UR tank wastes could be several thousand MT of nitrate/nitrite.

5.16.2.3 REDOX Process Wastes. The REDOX process used solvent extraction and chemical salting methods to recover uranium and plutonium from irradiated fuel. This process started up in January 1952 in S Plant. According to the production records, 19,706 MTU were processed through the REDOX Plant from 1952 to 1966 (Appendix B). The REDOX process flowsheets are summarized in Appendix D and were used as a basis for estimating the total amount of nitrate/nitrite in the REDOX process wastes.

The total use of ANN by the REDOX process contained 4,210 MT of Al, Section 5.1. The Appendix D analysis of REDOX flowsheets and essential material use states that the average neutralized REDOX HLW was 0.98M aluminum and 4.31M nitrate. This results in a total sodium nitrate content in the REDOX HLW of 58,290 MT NaNO_3 containing 42,520 MT nitrate.

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In January, 1963 (Isaacson 1965), the use of commercial NaNO_3 for hydrogen suppression in the fuel decladding step was discontinued when NaNO_3 solutions were recovered from dissolved waste tank farm sludge for recycling back to the dissolvers. In the decladding process 1.05 mole of NaNO_3 is converted to 0.9 mole of NaNO_2 and 0.15 mole of NH_3 per mole of Al dissolved. This conversion of NaNO_3 to NaNO_2 is reflected in Table 5.16-5 (flowsheets 8, 9A, and 9B) by the negative values for net reduction of nitrate to nitrite and ammonia.

Table 5.16-5. Amount of NaNO_3 and NaNO_2 Sent to Underground Storage Tanks from the REDOX Process.

Flowsheet	Allocated MTU	Salt waste NaNO_3 (MT)	Cladding waste	
			NaNO_3 (MT)	NaNO_2 (MT)
No. 4 and 5	9,400	-	527	737
No. 6	4,000	-	140	203
No. 7	1,930	-	29	137
No. 8	1,690	-	-171	119
No. 9A	2,360	-	-256	178
No. 9B	320	-	-14	10
Total	19,706	58,290	255	1,384

A total of 58,500 MT of NaNO_3 were transferred to underground storage from the REDOX Plant, containing 42,700 MT of NO_3 . A total of 1,384 MT of NaNO_2 was produced by the decladding waste, containing 923 MT of NO_2 .

This analysis gives a total REDOX nitrate/nitrite waste inventory of 43,600 MT based primarily on reported ANN consumption (96 percent) and secondarily on flowsheet analysis of NaNO_3 usage (4 percent). The HDW model gives a total of 27,100 MT nitrate/nitrite based on transfer records and unreferenced flowsheet compositions. Agnew et al. (1996) does not provide references for the REDOX waste compositions used. Agnew (1995) indicates eight references (GE 1951b, Merrill 1955, Crawley 1960, Isaacson 1965, Allen 1976, Jenkins and Foster 1978, Jungfleisch 1984, and Anderson 1990) were used to define the compositions of the four REDOX waste streams but does not state the specific reference or methodology.

5.16.2.4 PUREX Process Wastes. There were three main production periods at the PUREX Plant: 1983 through 1989, 1965 through 1972, and 1956 through 1964.

1983 through 1989. The period from 1983 through 1989 processed Zircaloy®-clad fuel exclusively. The NCRW produced during this period contained nitrate and nitrite from ammonium fluoride/ammonium nitrate (AFAN) used in decladding the fuel.

Tank core samples and flowsheet information can be used for the 1983 to 1989 period to estimate the total nitrate/nitrite sent to underground storage. The neutralized cladding removal waste (NCRW) that left the PUREX Plant during this period was sent to DSTs 241-AW-103 and 241-AW-105 while the neutralized current acid waste (NCAW) was sent to DSTs 241-AZ-101 and 241-AZ-102.

Based on flowsheets and tank core samples, about 30 MT of NO₃ and 9 MT of NO₂ were sent to 241-AW-103 and 241-AW-105 during the 1983 to 1989 period when the PUREX Plant processed 3900 MTU from N reactor (Schofield 1991). The combined mass per MTU for nitrate/nitrite is:

$$(30,000+9,000)\text{kg}/3,900 \text{ MTU} = 10 \text{ kg NO}_3/\text{NO}_2 \text{ in NCRW per MTU irradiated fuel}$$

For NCAW, Schofield estimated the nitrate and nitrite masses from flowsheet information and his knowledge of deviations from the flowsheet. He compared these with the results of core samples taken in 1989 (Schofield 1991). The results, summarized in Table 5.16-6, show excellent agreement. The higher sample-based numbers were chosen for this evaluation. Using 6,350 MTU¹ for NCAW (Section 5.20) the combined mass per MTU is:

$$(476,000+333,000)\text{kg}/6,350 \text{ MTU} = 127.4 \text{ kg NO}_3/\text{NO}_2 \text{ in NCAW per MTU total solvent extraction feed}$$

Table 5.16-6. Comparison of Nitrate/Nitrite Estimates in Neutralized Current Acid Waste.

	Flowsheet-based estimate (kg) ^a	Sample-based estimate (kg) ^b
NO ₃	436,000	476,000
NO ₂	323,000	333,000

PUREX = Plutonium-uranium extraction

^aEstimates derived from the PUREX process flowsheet (Schofield 1991)

^bEstimates derived from 1989 core sample data (Schofield 1991).

Combining NCRW and NCAW together yields a total nitrate mass of 506 MT and a total nitrite mass of 342 MT.

¹During 1983 through 1989, PUREX processed 3,900 MTU of irradiated fuel through solvent extraction by recycling 2,450 MTU of cold uranium to maintain the extraction column throughput above the minimum processing rate.

1956 through 1982. The site production records indicate 67,490 MTU and 629 MT of thoria were processed in the PUREX plant from 1956 through 1972 (Appendix B). The PUREX plant was maintained in standby from 1973 through 1982. From purchase records, the chemical requirements for the PUREX process and the resulting neutralized wastes were estimated (Allen 1976). From these records, Allen estimated the following quantities of nitrate and nitrite salts in PUREX tanked wastes from 1956 through 1972:

Table 5.16-7. Nitrate/Nitrite Salts Contained in PUREX Plant Tanked Wastes from 1965 through 1972^a.

Chemical	MT
NaNO ₃	4,590
Pb(NO ₃) ₂	13
NaNO ₂	11,730

PUREX = Plutonium-uranium extraction

^aAllen (1976).

The chemicals in Table 5.16-7 contain an equivalent to 3,350 MT of nitrate and 7,820 MT of nitrite. The combined mass per MTU is:

$$(3,350,000 + 7,820,000) \text{ kg} / 67,490 \text{ MTU} = 165.5 \text{ kg/MTU}$$

which is 20 percent higher than the 137.4 kg/MTU total solvent extraction feed calculated for the 1983 through 1989 period.

The contribution of nitrate and nitrite from the PUREX process is summarized in Table 5.16-8.

Table 5.16-8. Amount of Nitrate/Nitrite Sent to Underground Storage from the PUREX Process.

Period	1956 through 1972 (MT)	1973 through 1982 (MT)	1983 through 1989 (MT)	Total (MT)
NO ₃	3,350	0	506	3,860
NO ₂	7,820	0	342	8,160
NO ₃ /NO ₂	11,170	0	848	12,020

PUREX = Plutonium-uranium extraction.

5.16.2.5 B Plant Process Wastes B Plant and Semiworks processed tank wastes for strontium and cesium removal in the 1965 through 1982 time period. This processing involved: (1) processing PUREX and REDOX alkaline tank waste supernatants for ^{137}Cs recovery, (2) processing acidic wastes from PUREX, (3) the dissolution of retrieved tank farm sludges in nitric acid, and (4) subsequent neutralization of the raffinates before returning the wastes to the tank farms. The resulting sodium and nitrate additions by B Plant fission product removal operations for the 1965 to 1972 period was included in the PUREX wastes by Allen. The combined nitrate production determined by Allen is retained in Table 5.16-8.

The nitrate content of B Plant process wastes for the 1973 through 1982 period when no processing of PUREX acid waste was performed can be estimated from the B Plant NaOH use, section 5.20. The B Plant NaOH use from 1973 through 1982 was 3,588 MT NaOH. This amount of NaOH could neutralize 5,650 MT of nitric acid in dissolved tank sludges. Allowing for excess NaOH to control the neutralized waste pH, this analysis assumes that 5,000 MT of nitrate was in the B Plant process waste during the 1973 to 1982 period.

5.16.3 Best-Basis Estimate for Nitrate/Nitrite Inventory Value

Table 5.16-9 provides the results from this evaluation for nitrate/nitrite, together with comparable estimates from other sources. The best-basis global estimate for Hanford Site tank waste is about 85,700 MT of nitrate/nitrite. This estimate is 45 percent higher than HDW model estimate of 58,900 MT nitrate/nitrite (Agnew et al. 1996) and 25 percent lower than the current value of 117,000 MT in the TWRS reference chemical inventory (Shelton 1996). The current TWRS reference inventory is based on 1986 EIS estimates for tank wastes, corrected for known additions to the waste tanks.

The present estimate is based primarily on reported chemical consumption for REDOX, PUREX, and B Plant; reported nitrate/nitrite crib discharges for BiPO_4 and UR; and flowsheet values of chemical consumption for the BiPO_4 process. The HDW model is based on flowsheet waste compositions and waste transfer records from Hanford Site processing plants. The TWRS inventory is based on the 1986 EIS estimate of Hanford Site tank inventory in 1983 plus additions to the tank system after 1983.

Table 5.16-9. Estimated Amount of Total Nitrate/Nitrite in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
BiPO ₄ wastes in SST/DSTs	7,530	5,710	NA
UR wastes in SST/DSTs	17,600	23,600	NA
REDOX	43,600	27,100	NA
PUREX and B Plant	17,000	2,490	NA
Total	85,700	58,900	117,000

DST = Double-shell tank
 HDW = Hanford Defined Waste
 NA = Not attributed to individual waste streams
 PUREX = Plutonium-uranium extraction
 REDOX = Reduction oxidation
 SST = Single-shell tank
 TWRS = Tank Waste Remediation System
 UR = Uranium recovery.

The process wastes of PUREX and B Plant wastes in the period from 1956 through 1989 are combined in Table 5.16-10 for comparison purposes. The individual process contributions in the HDW model (Agnew et al. 1996) were not developed by this report. The 1956 through 1989 total PUREX + B Plant value of 2,490 MT nitrate/nitrite was determined as the difference of the total nitrate/nitrite (58,900 MT nitrate/nitrite, Agnew et al. 1996) and the sum of the BiPO₄, UR, and REDOX tanked wastes discussed in Sections 5.16.2.1 through 5.16.2.3 (56,400 MT nitrate/nitrite).

5.17 PHOSPHATE

The following were the primary sources of phosphate in tank wastes:

1. Phosphoric acid (H_3PO_4) used to separate and decontaminate plutonium from uranium and fission products in the $BiPO_4$ process.
2. Solvent losses from the PUREX and B Plant solvent extraction processes.
3. Phosphoric acid (H_3PO_4) used for solvent treatment in the Uranium Recovery (UR) process (GE 1951a).
4. Fission product recovery using a phosphotungstic acid precipitation process.
5. Decontamination solutions containing phosphoric acid or trisodium phosphate (Na_3PO_4).

Phosphate is a trivalent anion. The sodium salt is soluble in acidic and alkaline solutions. The calcium, strontium, and barium salts are insoluble in alkaline solution, thermodynamically stable, and resist metathesis to the soluble trisodium phosphate by sodium hydroxide. In the Hanford wastes, approximately 60 percent of the total phosphate sodium is expected to be water soluble. The remaining 40 percent of phosphates in the sludges are primarily aluminum and iron phosphates of which three fourths are metathesized to the soluble trisodium phosphate by sodium hydroxide leaching. This results in approximately 90 percent of the tank waste phosphate inventory being routed to the low activity waste (LAW) and 10 percent to the HLW.

5.17.1 Identification of Phosphate Inventory Values

The reported global phosphate inventory values are 4,030¹ MT (HDW model, Agnew et al. 1996), 4,970 MT (TWRS, Shelton 1996), and 6,000 MT by this report.

The methodology and analysis to obtain a total global phosphate inventory of 6,000 MT phosphate is contained in Section 5.17.2.

¹This values is taken from the Table ES-1a of the HDW model, Rev. 4 (Agnew et al. 1996). It is inconsistent with the value obtained from summing the different process contributions to the phosphate global inventory as defined by Agnew et al. (1996). Refer to Table 5.17-6.

5.17.2 Phosphate Inventory Evaluation

The Hanford Site tank phosphate inventory is estimated by computing the total amount of phosphate added to each of the separations processes and subtracting the amounts of phosphate in the wastes that were discharged to cribs and trenches. Crib transfers are especially important because these transfers are used to estimate the average phosphate concentration and total volume of BiPO_4 and UR process wastes that were left in the tanks (Waite 1991, Anderson 1990). Other sources of useful information include the following:

1. Process flowsheets (Schneider 1951 and GE 1951b).
2. Tank transaction records and estimates from the HDW model (Agnew et al. 1995a and Agnew et al. 1996, respectively).
3. Partial chemical usage summaries (McDonald 1968, McDonald and Hogan 1977, and Hogan 1992).
4. Tank samples and tank inventory estimates derived from supernatant and core samples (Hodgson 1995).

These sources are used to develop phosphate inventory estimates for each of the chemical separations processes.

5.17.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process started up at T Plant in December 1944, and at B Plant in April 1945, and continued until 1952 in B Plant and until 1956 in T Plant. The site production records indicate that 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the BiPO_4 processing is developed in Appendix C based on the flowsheet by Schneider (1991). Comparable HDW phosphate values are obtained from the "waste volume out" and "phosphate concentrations" of the HDW. The HDW phosphate concentrations used are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

The phosphate usage in the BiPO_4 processing is shown in Table 5.17-1 for the Schneider (1991) flowsheet and for the HDW model (Agnew et al. 1996).

Table 5.17-1. Amount of Phosphate in BiPO₄ Process Wastes, MT.

Waste stream	Present estimate	HDW model (Agnew et al. 1996)
Metal waste (MW)	3,050	2,320
First-cycle and coating waste (1C/CW)	2,820	3,540
Second-cycle waste (2C)	2,570	1,580
224 Waste	350	150
Total	8,790	7,590

Metal Waste. The first steps in the BiPO₄ process dissolved the uranium metal slugs and extracted the plutonium by carrier precipitation with bismuth phosphate. This resulted in the metal waste stream. The metal waste stream contained the bulk of the uranium and approximately 90 percent of the long-lived fission products (e.g., ¹³⁷Cs and ⁹⁰Sr) and was sent to the SSTs for storage.

The stored metal waste containing the uranium from BiPO₄ processing was later retrieved as the feed for the UR process. Approximately 73 percent of the uranium is thought to have been associated with the sludge, as Na₄UO₂(CO₃)₃ and NaUO₂PO₄, while the remainder was dissolved in the supernatant (GE 1951b). Furthermore, it appears that 5.5 percent of the metal waste sludge was not recovered from the tanks during the sluicing campaign (Rodenhizer 1987). Two-thirds of the uranium in the metal waste sludge was in the form of Na₄UO₂(CO₃)₃ and remainder as NaUO₂PO₄, the following calculation indicates the amount of phosphate that might be in the tanks with the residual metal waste.

$U(MW) = (0.73)(0.055)(7,800 \text{ MTU}) = 310 \text{ MTU}$, where U(MW) is the amount of residual metal waste uranium in the tanks.

$PO_4(MW) = (U)[(0.34)(1/0.6134)(0.245)]$, where:

0.34 equals the fraction of uranium in the form of NaUO₂PO₄,

0.6134 equals the fraction of uranium as in NaUO₂PO₄,

0.245 equals the fraction of phosphate in NaUO₂PO₄.

$PO_4(MW) = 42 \text{ MT}$ (from residual MW in the tanks).

Thus, 3,010 MT of phosphate in the metal wastes was transferred to the UR process from the total 3,050 MT of phosphate initially produced in the metal waste.

Cribbed BiPO₄ Wastes. Portions of the other BiPO₄ waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The total phosphate discharged to cribs and specific retention trenches was 2,610 MT phosphate in 259,000 m³ (68.5 Mgal) (Waite 1991).

The HDW model estimates 2,110 MT phosphate was cribbed from the BiPO₄ process in 173,000 m³ (45.8 Mgal) (Agnew et al. 1996). The HDW estimate for BiPO₄ wastes is obtained from the crib volume and the supernatant phosphate concentration (Agnew et al. 1996). The phosphate concentrations are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

Summary for BiPO₄ Process Wastes. The residual MW, 1C, and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.17-2. This report estimates the residual BiPO₄ tank wastes contain 3,170 MT of phosphate versus the 3,070 MT estimated by the HDW model.

Table 5.17-2. Estimated Amount of Phosphate in BiPO₄ Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes produced	8,790	7,590
BiPO ₄ wastes cribbed	2,610	2,110
BiPO ₄ wastes sent to uranium recovery	3,010	2,410
BiPO ₄ wastes in single-shell and double-shell tanks	3,170	3,070

HDW = Hanford Defined Waste.

The principle differences in the present estimate and the HDW model are the input chemicals in the flowsheets assumed and the volume of cribbed BiPO₄ wastes.

5.17.2.2 Uranium Recovery Process Wastes. The UR process began in 1952 to extract uranium from the BiPO₄ process MW stored in B, BX, BY, C, T, TX, and U tank farms.

Uranium Recovery Chemicals Added. The design basis flowsheet for the UR process is provided in the *Uranium Recovery Technical Manual* (GE 1951a). Phosphate use in the UR process was limited to solvent treatment. The design basis flowsheet states that organic treatment could use sodium sulfate, or sulfuric acid, or phosphoric acid for solvent washing. The phosphoric acid alternate solvent wash is stated as less than 12.5 flows of 1.08M phosphoric acid. This is equivalent to less than 0.2 MT phosphate per MTU in the UR feed. For the 7,500 MTU processed by UR the potential phosphate use is less than 1,500 MT.

As a result, the UR flowsheet (GE 1951a) is not expected to provide an accurate estimate of UR phosphate usage. For this analysis, an allowance of 800 MT phosphate is assumed.

For the HDW model the amount of phosphate added during UR is the difference in the phosphate content between the UR wastes produced and the BiPO_4 wastes sent to UR. The HDW model phosphate values in the UR wastes produced (3,260 MT PO_4) are obtained from the "waste volume out" and "phosphate concentrations" of the HDW. The HDW phosphate concentrations used are attributed to Borsheim and Simpson (1991) and Sloat (1954). Subtraction of the phosphate inventory to UR (2,410 MT PO_4) gives an 850 MT PO_4 chemical addition in UR.

Cribbed Uranium Recovery Wastes. The HDW model (Agnew et al. 1996) analysis of transfer records from the UR operation indicates that 155,000 m^3 (41 Mgal) or 62.5 vol% of the total UR waste streams were cribbed and 37.5 vol% of the total UR waste streams were routed to the SSTs. The HDW model UR waste phosphate distribution is 61 percent (1,990 MT phosphate) to the cribs and 39 percent (1,270 MT phosphate) to the SSTs. The HDW estimate for UR wastes is obtained from the crib volume and the supernatant phosphate concentration of Agnew et al. (1996). The phosphate concentrations are attributed to Borsheim and Simpson (1991) and Sloat (1954).

Waite (1991), states that 155,000 m^3 (41 Mgal) of scavenged UR wastes containing 1,520 MT of phosphate was discharged to cribs and specific retention trenches. This is less than 50 percent of the phosphate content in the UR wastes.

Summary for Uranium Recovery Process Wastes. Table 5.17-3 provides a summary phosphate material balance for the UR process for this analysis and the HDW model. This report estimates the residual UR tank wastes contain 2,300 MT of phosphate versus the 1,270 MT phosphate estimated by the HDW model.

Table 5.17-3. Estimated Amount of Phosphate in Uranium Recovery Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO_4 wastes sent to UR	3,010	2,410
UR chemicals added	800	850
UR wastes produced	3,810	3,260
UR wastes cribbed	1,520	1,990
UR wastes in single-shell /double-shell tanks	2,300	1,270

HDW = Hanford defined waste

UR = Uranium recovery.

The difference in analysis that provided the results in Table 5.17-3 are

1. UR feed - Values calculated in section 5.17.2.1, above.
2. UR Wastes Produced
 - a. Present estimate of UR wastes produced equals the amount in the UR feed plus UR chemicals added, (3)(a) below)
 - b. The HDW model basis for UR wastes produced is Sloat (1954) and Borsheim and Simpson (1991).
3. UR Chemicals Added
 - a. An allowance of 800 MT phosphate for solvent washing.
 - b. The HDW model is the difference between 2. and 1, above.
4. UR Wastes Cribbed
 - a. Present estimate based on Waite (1991).
 - b. The HDW basis uses transfer records and calculated phosphate solubility.

Due to the lack of definitive data on the chemical usage in UR and the phosphate split between UR waste sludges and cribbed UR waste supernatants, there is little definitive basis for the amount of phosphate associated with residual UR wastes in the waste tanks. The HDW basis for phosphate content in cribbed UR wastes result in a lower fraction of phosphate in the residual UR tank wastes, 39 percent versus 60 percent for this analysis. The uncertainty in phosphate content in residual UR tank wastes could be hundreds of MT.

5.17.2.3 REDOX Process Wastes. Phosphates were not used in the REDOX process. This statement is confirmed by examining the composition of tank 241-S-104 waste. This waste contains 480 kg of phosphate in 409,000 m³ (108 kgal) of R1 (REDOX process high-level) waste and 91,000 m³ (24 kgal) of CWR1 (REDOX process cladding waste) (Agnew et al. 1995b).

If the phosphate concentration in tank 241-S-104 waste can be used as a basis for judging the total amount of phosphate in all of the REDOX process waste, the total amount of phosphate would be only 6.26 MT (less than 0.15 percent of the phosphate in BiPO₄ and UR waste).

This calculation does not include phosphate associated with the 241-S-104 supernatant, most of which was transferred tank 241-TY-103 in the first quarter of 1970 and eventually incorporated into T2SlTck (T2 salt cake) (Anderson 1990, Agnew et al. 1995a). T2SlTck contains about 285 MT of phosphate from a variety of sources, but none of this, other than incidental amounts, can be attributed to the REDOX process.

5.17.2.4 PUREX Process Wastes. The phosphate content in PUREX wastes originates from TBP solvent losses to the wastes. Table 5.17-4 shows the PUREX process phosphate contribution to the tank wastes given by this report and the HDW model.

Table 5.17-4. Estimated Amount of Phosphate in PUREX Tank Wastes, MT.

Process	Present estimate (Sederburg and Reddick 1994)	HDW model (Agnew et al. 1996)
PUREX (1956-1982), PL1, TH1, TH2	243	90
PUREX (1983-1989), PL2	25	287
Total PUREX (1956-1989)	268	377

HDW = Hanford Defined Waste

PL1 = PUREX process low-level waste (1956 to 1976)

PL2 = PUREX process low-level waste (1983 to 1988)

PUREX = Plutonium-Uranium Extraction

TH1 = Thorium high-level waste or cladding waste (1966)

TH2 = Thorium high-level waste or cladding waste (1970).

HAPO/Chemical Processing Department Monthly Reports and monthly essential material usage records were used to define the annual PUREX solvent losses to tanks, cribs, and the atmosphere for the period 1955 through 1991 (Sederburg and Reddick 1994). Sederburg and Reddick identified 773 m³ of TBP reporting to the waste tanks. The 773 m³ of TBP contains the equivalent of 268 MT phosphate. The Sederburg and Reddick analysis indicates that approximately one percent of the TBP content is in the NCAW and the remainder in Organic Wash Waste (OWW).

The HDW model phosphate values in the PUREX tank wastes produced (377 MT phosphate) are obtained from the "waste volume out" and "phosphate concentrations" of the HDW for streams PL1, TH1, TH2, and PL2.

5.17.2.5 B Plant Process Wastes. Monthly essential material usage records can be used to define the chemical consumption for B Plant and tank farms during the 1965 to 1982 period (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.20-12. The TBP, and di-2-ethyl hexylphosphoric acid (D2EHPA) use in 1968 through 1979 was used as the solvent for strontium recovery. Solvent losses and the associated phosphorus reported to the waste tanks.

Table 5.17-5 shows that the total estimated phosphate use by B Plant and tank farms processing in the period of 1965 through 1977 is 12 MT phosphate. This analysis did not determine if the HDW model attributed any phosphate input to the tanks from B Plant fission product recovery operations.

Table 5.17-5. Phosphate Use in B Plant from 1965 through 1977. (2 Sheets)

Year	Solvent component, MT	
	TBP, (C ₄ H ₉ O) ₃ PO	D2EHPA, (C ₈ H ₁₇ O) ₂ POOH
1965 ^a		17.7
1966 ^a		1.4
1967 ^a		
1968 ^b	6.0	10.0
1969 ^b	5.5	5.4
1970 ^b	4.7	8.4
1971 ^b	3.2	5.5
1972 ^b	7.4	9.7
1973 ^b	3.2	6.5
1974 ^b	2.6	6.3
1975 ^b	2.1	2.3
1976 ^b	5.7	6.3
1977 ^b	1.8	6.8
Total	42	70
Subtot PO ₄ , MT	4.9	7.0
Total PO ₄ , MT	12 ^c	

D2EHPA = Di(2-ethylhexyl)phosphoric acid

TBP = Tri-butyl phosphate

^aMcDonald 1968

^bMcDonald and Hogan 1977

^cTotal B Plant and tank farms phosphate use from 1965 through 1977.

5.17.2.6 Other Cribbed Wastes. Agnew et al. (1996) defines portions of waste streams other than the BiPO₄ and UR process wastes discussed earlier as having been cribbed. These streams are identified in Agnew et al. (1996) as P1, CWP1, OWW1, DW, T1-SltCk, and WTR. The fraction of the streams cribbed is given in Agnew et al. (1996) in Appendix D. The total phosphate in the streams is given by the volume and the concentration. The summed products of these three values give a total calculated amount of phosphate cribbed of 33 MT phosphate.

This report's analysis is based on the reported cribbed phosphate given by Waite (1991). The cribbed phosphate content in BiPO_4 and UR wastes given in the previous sections includes all phosphate cribbed in the common crib and specific retention sites. Therefore, the "other cribbed wastes" identified in Agnew et al. (1996) are included in the BiPO_4 or UR values identified by Waite (1991) and no separate value is identified by this report's analysis.

5.17.2.7 N Reactor Decontamination Wastes. Agnew et al. (1996) defined the residual amount of N Reactor decontamination wastes in the DSTs after a single vault of "Phosphate/Sulfate Waste" grout was produced. The N Reactor decontamination waste stream, "N", as defined in Agnew et al. (1996) Appendix D, with composition attributed to Lucas (1989). The resulting residual N Reactor Decontamination Hanford Site tank inventory is 190 MT phosphate.

5.17.2.8 Other Phosphate Sources. Decontamination agents were used by processing facilities, tank farm operations, and for equipment reclamation by T Plant. The decontamination agents frequently contain phosphate compounds. The decontamination wastes and laboratory wastes are routed to tank farms. The expected total phosphate content in decontamination chemicals and laboratory wastes is expected to be less than 100 MT phosphate. This study uses an allowance of 40 MT phosphate contained in decontamination agents.

5.17.3 Best-Basis Estimate for Phosphate Inventory

Table 5.17-6 provides the results from this evaluation for phosphate, together with comparable estimates from other sources. The global best-basis estimate for Hanford Site tank waste is about 6,000 MT of phosphate. This estimate is about 20 percent higher than HDW model estimate of 4,870 MT phosphate (Agnew et al. 1996) and the current value of 4,970 MT in the TWRS reference chemical inventory (Shelton 1996). The current value is based on 1986 EIS estimates for tank wastes, corrected for known transfers to cribs.

Table 5.17-6. Estimated Amount of Phosphate in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
BiPO ₄ wastes in SST/DST's	3,170	3,070	NA
UR wastes in SST/DST's	2,300	1,270	NA
REDOX	6	0	NA
PUREX	277	377	NA
B Plant	12	-	NA
Other cribbed wastes	-	(33)	NA
N Reactor decontamination	190	190	NA
Other phosphate sources	40	-	NA
Total	6,000	4,870 ^a	4,970

HDW = Hanford Defined Waste

NA = Not attributed to individual waste streams

TWRS = Tank Waste Remediation System

^aSummary Table E-1a of HDW model, Rev. 3 (Agnew et al. 1996) indicates 4,028 MT PO₄.

The current Hanford Site tank waste inventory estimate of 6,000 MT phosphate has the highest uncertainty in the residual wastes produced by the BiPO₄ and UR processes. These wastes represent 90 percent of the estimated total, thus total uncertainty in the phosphate inventory is high. Both the current estimate and the HDW model are based on flowsheets for additions and estimated crib discharges. Tank sampling and analysis of sludges will not fully resolve the uncertainty because the alkaline soluble trisodium phosphate in the supernatants has resulted in a fairly wide distribution of residual phosphate in tank supernatants and salt cakes.

It is noted that the addition and subtraction of the individual streams in the HDW model by this report's analysis resulted in a total of 4,870 MT phosphate versus a total of 4,028 MT phosphate given in the HDW model summary (Agnew et al. 1996) for the residual tank waste phosphate inventory. This apparent discrepancy requires further investigation to resolve.

5.18 POTASSIUM

There are two classic sources of potassium in Hanford Site tank wastes: (1) potassium compounds that were purposely added in the separations processes; and (2) potassium contamination from other process chemicals.

Potassium was first added in 224-B/T Buildings operations (final plutonium decontamination and concentration cycles in the bismuth phosphate processes). Three potassium compounds were added: KMnO_4 as an oxidant, KOH to effect the metathesis of LaF_3 solids, and KNO_3 in rework solutions from the plutonium isolation process. In the REDOX process, KMnO_4 was added during early years as part of the headend ruthenium oxidation/volatilization step.

During the 1950's some potassium was added in the Uranium Recovery process as $\text{K}_4(\text{FeCN})_6$ (along with NiSO_4) to effect the scavenging precipitation of ^{137}Cs from waste supernatants.

In the PUREX Plant, potassium was routinely added as KMnO_4 to wash process solvents. Later, during 1966 and 1970, KF was a process chemical for the special thorium- ^{233}U campaigns. More recently, KOH was added to metathesize uranium tetrafluoride produced in chemical decladding of Zircaloy®-clad N Reactor fuel elements.

Certain amounts of potassium were also introduced as a minor impurity in other essential chemicals used in separations processes. Of these materials, the sodium hydroxide used to neutralize acidic waste solutions is likely the chief source of potassium as a contaminant.

The chemistry of potassium and sodium compounds is generally very similar. Both are group 1A alkali metals. Most compounds of both potassium and sodium are very water soluble, although significant amounts of water insoluble or partially soluble sodium and potassium compounds also exist in Hanford Site waste sludges.

The chief importance of potassium in the tanks chemical inventory is its impact on future waste pretreatment and immobilization processes. The majority of the potassium will exist in tank waste supernatants that will eventually be vitrified and disposed as LLW waste. Prior to vitrification, these waste supernatants will be processed using ion exchange methods to remove radio-caesium (^{137}Cs) which is also an alkali metal. Potassium ions compete with cesium ions for uptake by both organic ion exchange resins and inorganic sorbents. Specification of process conditions for satisfactory removal of ^{137}Cs from Hanford Site alkaline waste solutions requires a good knowledge of the tank potassium inventory both on a global and a tank-by-tank basis.

Major amounts of potassium also will contribute towards increasing the volume of vitrified LLW since the glass volume is controlled by the alkali metal content in the wastes.

5.18.1 Identification and Comparison of Potassium Inventory Values

A comparison of TWRS and HDW model estimates of the global inventory for potassium (see Table 5.0-1, 619 MT versus 481 MT) shows a minor degree of disagreement. Comparison of SST and DST potassium inventories from the same table shows even greater disagreement. In addition, the early estimate of Allen (1976) for potassium in pre-1976 wastes is an order of magnitude less than the HDW model and TWRS values. In an attempt to understand and reconcile these differences, an independent global estimate was calculated, as described, below.

5.18.2 Potassium Inventory Evaluation

The potassium inventory can best be reestimated by assessing several sources of useful information: (1) process flowsheets for the three primary fuels separations processes as well as for waste scavenging processes, (2) essential material records, (3) records of potassium contamination in essential materials, and (4) tank samples.

The source of potassium as a contaminant in other process chemicals was investigated by looking at specifications for several grades of caustic soda. So far this path of investigation has found no data on which to base an estimate of the "contaminant potassium" source term.

Results of the reestimate for addition of potassium compounds are summarized in Table 5.18-1. Here, the potassium usage rate (kg of potassium per MTU of fuel processed) is listed for each potassium bearing essential chemical by plant and by operating period (in certain cases where flowsheet consumption rates changed with time). The fourth column lists tonnages of fuel processed while various flowsheets were in effect. Column five lists an estimated "overage factor" to account for inefficiencies caused by rework, or the recycle of cold uranium through the solvent extraction processes. These "overage factors" are consistent with cold uranium percentages discussed in Section 4.3.1. Lastly, Table 5.18-1 (column six) gives calculated values for total potassium added to tanks, corresponding to each plant/flowsheet/chemical consumed. Exceptions to this flowsheet methodology are the U Plant $K_4Fe(CN)_6$ addition and the PUREX Plant Thorium- ^{233}U Campaign additions where historical consumption values were used instead of flowsheet values.

Table 5.18-1. Estimates of Total Potassium Added to Hanford Waste Tanks.

Plant	Chemical added	Flowsheet usage rate (Kg K/MTU)	Uranium processed (MTU)	Overage ratio	Total potassium (MT)	References
Bismuth phosphate	KOH	1.25 E+02	7.80 E+03	1.00	9.73 E+02	Schneider 1951
	KMnO ₄	2.90 E+00	7.80 E+03	1.00	2.26 E+01	Schneider 1951
	KNO ₃	3.30 E+00	7.80 E+03	1.00	2.57 E+01	Schneider 1951
REDOX (01/52 - 09/59)	KMnO ₄ (High)	2.73 E+00	1.13 E+04	1.15	3.55 E+01	GE 1951b
	KMnO ₄ (Low)	2.00 E+00	1.13 E+04	1.15	2.60 E+01	GE 1951b
Scavenging Process (U Plant)	K ₄ Fe(CN) ₆	--	--	--	2.25 E+01	Stedwell 1957
PUREX (U-Al)	KMnO ₄	2.30 E-01	6.11 E+04	1.06	1.49 E+01	Matheison and Nicholson 1968
PUREX (U-Zr through 1972)	KMnO ₄	3.20 E-01	1.34 E+03	1.09	4.67 E-01	RHO 1985b
	KOH	6.60 E+01	1.34 E+03	1.00	8.83 E+01	RHO 1985b
PUREX (U-Zr) 1983 - 1989	KMnO ₄	3.20 E-01	3.89 E+03	1.63	2.03 E+00	RHO 1985b
	KOH	6.60 E+01	3.89 E+03	1.00	2.57 E+02	RHO 1985b
PUREX (Th)	KF	--		Included in Totals	5.03 E+00	Isochem 1967 Jackson and Walser 1977
	KMnO ₄	--		Included in Totals	1.43 E+00	Isochem 1967 Jackson and Walser 1977
Total Potassium Added to Tanked Waste					1.45 E+03	
Estimated Potassium Discharged to Cribs					1.04 E+03	
Net Potassium Remaining in Waste Tanks					4.04 E+02	

PUREX = Plutonium-Uranium Extraction
REDOX = Reduction and oxidation.

Further details are given below for each plant operation.

5.18.2.1 Bismuth Phosphate Process. Schneider 1951 indicates that two potassium compounds (KOH, and KMnO₄) were introduced into the 224-B/T Buildings operations while a third potassium chemical, KNO₃, added in 231 Building operations, was back cycled to the 224-B/T Buildings. Of these, the consumption of KOH (used for metathesizing lanthanum/plutonium fluoride precipitates) dominates all other sources of potassium added to tanks. However, it is likely that this large quantity of potassium (970 MT), being soluble in

the 224-B/T Buildings waste supernatants, was subsequently routed to cribs as were the 224-B/T wastes

5.18.2.2 REDOX Process. The REDOX process consumed KMnO_4 in the headend ruthenium oxidation process step from plant startup in 1952 until September 1959, when the troublesome headend oxidation step was finally eliminated from the process. During this period the permanganate consumption ratio was somewhat variable according to monthly reports. Unfortunately, no actual records of direct chemical consumption have been found for potassium permanganate, and even the two flowsheets documents issued during this period are very ambiguous as to the permanganate consumption ratio. To deal with this uncertainty, Table 5.18-1 shows an estimated range for the likely consumption ratio during this period.

5.18.2.3 Uranium Recovery Scavenging Processes. According to flowsheet records, potassium was only introduced during in-plant scavenging operations. Stedwell (1957) recorded data on actual chemical consumption in all of these operations. The consumption value for $\text{K}_4\text{Fe}(\text{CN})_6 \cdot 3\text{H}_2\text{O}$ (22,500 kg of K) is directly listed in Table 5.18-1.

In subsequent sludge settling and supernatant disposal operations, it is likely that essentially all of this added potassium remained soluble and was routed to cribs. This assumption is consistent with Sloat (1954).

5.18.2.4 PUREX Process. Potassium consumption values in Table 5.18-1 are listed for several fuel types and operating periods.

During the processing of aluminum-clad uranium fuel, the only intentional potassium input was in the form of KMnO_4 , used along with sodium carbonate for solvent washing. According to Matheison and Nicholson (1968) the use of permanganate didn't begin until sometime around 1957, and then it was only added to the #1 solvent wash system. Later, permanganate was used in both solvent washing operations, along with the introduction of spent wash recycle from the #2 system to the #1 system. In Table 5.18-1 these variations have been approximated by using the consumption factor from the 1968 flowsheet as representative of operations 10/57 through 1972, during which 61,000 MTU of uranium fuels were processed.

The processing of thorium oxide targets in two separate campaigns (1966 and 1970) introduced potassium in two chemical forms, KF and KMnO_4 . The KF was primarily used to accelerate dissolution of the thorium oxide fuel in nitric acid. In addition, significant quantities of KF were consumed during pre-campaign flushing operations. For the 1966 campaign, actual chemical consumption data are given in Isochem (1967). The accounting of the 1970 campaign (Jackson and Walser 1977) gives a processing flowsheet but no information on quantities of KF consumed during flushing. In the absence of actual consumption data for 1970 campaign flushing data from the first campaign (Isochem 1967) were assumed to be representative of 1970 operations. Processing consumption factors for

the 1970 campaign were very similar to the 1966 campaign (8.15 kg K/MT Th from KF and 2.61 kg K/MT Th from KMnO_4).

The processing of Zircaloy®-clad fuel from N-reactor called for the use of KMnO_4 (for solvent washing) and KOH (for metathesis of UF_4 solids generated during decladding operations). Consumption factors were derived from the flowsheet (RHO 1985b). Table 5.18-1 shows consumption factors and fuel tonnage values for two time periods: (1) pre-1973 when the overage factor was estimated to be 9 percent; and (2) 1983 through 1989, when the overage factor was significantly higher (63 percent).

5.18.2.5 Reevaluation of Potassium Discharged from Tanks and Net Tank Inventory.

Table 5.18-1 indicates that a total of $1.45\text{E}+03$ MT of potassium was added to tanked waste. As mentioned, above, essentially all potassium originating from the bismuth phosphate 224-B/T Buildings and from in-plant scavenging operations (the addition of $\text{K}_4\text{Fe}(\text{CN})_6$) would have remained soluble in waste supernatants and been routed to cribs in supernatant discharges from the tanks. Thus, Table 5.18-1 shows a total of $1.04\text{E}+03$ MT of potassium estimated to have been discharged to cribs. This leaves an estimated $4.04\text{E}+02$ MT of potassium as the net global inventory in tanks.

5.18.3 Best-Basis Estimate for Potassium Inventory Value

The reestimated potassium inventory value (404 MT) is lower than the HDW model value (481 MT) by a relative percent difference of 17 percent. This difference is small enough to suggest a "conclusion of confirmation:" either value or the average could be adopted as a best estimate of the real inventory. Given the fact that the reevaluation assumed 100 percent solubility and crib discharges of a major source term of potassium (and therefore, may be slightly low), coupled with the fact that the HDW model may have more accurately accounted for supernatants entrainment in sludge solids, it is suggested that the HDW model value (481 MT) be adopted as the best-basis global inventory estimate for potassium.

5.19 SILICON

Five sources for silicon in the tanks have been identified:

1. Fuel fabrication
2. Chemical process operations
3. Process chemical impurities
4. Direct addition by tank farms
5. Blowsand.

This evaluation has established an independent estimate for each of these sources. The independent values are compared to the HDW model prediction and TWRS values for silicon.

5.19.1 Identification of Silicon Values

Estimates of silicon to date have primarily focused on discharge records and process flowsheets of the chemical processes. Table 5.19-1 compares some of the reported values.

Table 5.19-1. Reported Silicon Inventory Values.

Reference	Inventory (MT)	Comment
TWRS Baseline (Shelton 1996)	692	
HDW model (Agnew et al. 1996)	614	
<i>Hanford Defense Waste Final Environmental Impact Statement</i> (DOE 1987)	447	Silicon absent from double-shell tank waste compositions.
TRAC (Jungfleish 1984)	317	Calculated waste generated through 1980 only.
Allen (1976)	418	Calculated waste generated through 1972 only.

HDW = Hanford Defined Waste
TRAC = Track radioactive components
TWRS = Tank Waste Remediation System.

5.19.2 Silicon Inventory Evaluation

Previous estimates of silicon have primarily focused on developing estimates from process flowsheets and discharge records of the major chemical processes. The present evaluation has identified additional sources of silicon to be evaluated. These additional sources include: (1) fuel fabrication, (2) minor chemical processes, (3) process chemical impurities, (4) direct addition by tank farms, and (5) blowsand.

5.19.2.1 Fuel Fabrication. Approximately 99,000 MTU was processed by various separation facilities at the Hanford Site (Appendix B). This value includes the fuel processed by the PUREX Plant between 1983 and 1989. Fuel elements were clad in either aluminum or zirconium to protect the uranium from corrosion during irradiation in the reactors. Zirconium-clad fuel was used in N Reactor exclusively, whereas aluminum-clad fuel was used in all the older single-pass reactors.

The primary source of silicon from fuel fabrication was the Al-Si braze layer of the aluminum-clad fuel. Small contributions of silicon came from the silicon impurities in the aluminum alloy fuel jackets and in the uranium metal. The zirconium alloy jackets also contain trace quantities of silicon.

Al-Si Braze Alloy. The hypoeutectic Al-Si alloy used in the triple-dip and lead dip processes in general ranged from 10.6 to 11.5 wt% silicon (Burley 1958). The nominal silicon content of Al-Si was 10.8 wt%. The silicon attributable to the Al-Si bond layer was estimated from gap allowance between the uranium metal core and the aluminum alloy jacket. Gap volumes were derived and calculated from drawings of the various fuel types fabricated. An allowance for erosion of the aluminum components experienced during canning operations, as reported by Weakley (1958) was included. Assumptions detailed in Section 4.2.1 included design differences between natural uranium fuel and enriched uranium fuel. The specifications of both solid fuel, and Internal and External fuel were also used. These factors, when combined with the quantities of the various fuel types processed, result in an estimated quantity of silicon added by the Al-Si bond layer of 118.3 MT.

Silicon from the Uranium Core. Weakly (1958) reports a maximum silicon specification for uranium metal of 75 $\mu\text{g/g}$. Using this specification and a value of 93,200 MTU of aluminum-clad fuel yields a maximum estimate of 7 MT of silicon.

Silicon from the Aluminum Jacket Alloy. At least two types of aluminum alloy were used for the fuel element jacket. The 1245 (C-64) alloy was introduced in 1944 and the M-388 (X-8001) alloy was introduced to fuel fabrication in mid 1957 (Weakley 1958). Full scale use of the X-8001 alloy was in effect in December 1960 (DeNeal 1970). Both of these alloys had a maximum silicon specification of 0.17 wt%. Using the value of 93,200 MTU of aluminum-clad fuel yields a maximum estimate of 4.3 MT silicon.

Silicon Introduced by Zirconium-Clad Fuel Fabrication. The maximum silicon specification on the N Reactor fuel components was 124 $\mu\text{g/g}$ in the uranium 601 alloy, 100 $\mu\text{g/g}$ in the Zircaloy-2[®] alloy, and 250 $\mu\text{g/g}$ in the braze metal. The separations plant reconstructed fuel process history in Appendix B indicates that 1,820 MT of zirconium-clad fuel was processed between 1963 and 1972, and 3,890 MT was processed between 1983 and 1989. Using these values, a maximum estimate of 0.8 MT silicon is obtained. Almost all of this silicon is in the uranium 601 alloy.

5.19.2.2 Chemical Process Operations. Chemical processes introduced silicon to the waste as intentional routine and non-routine chemical additions.

Routine Additions of Silicon During Chemical Process Operations. A number of chemical processes were operated at Hanford to recovery plutonium, uranium, strontium, cesium as well as other isotopes. Evaluation of these processes is in progress. The major processes to be evaluated are as follows:

- Bismuth Phosphate
- TBP/Uranium Recovery
- In-Farm Cesium/Strontium Scavenging
- REDOX
- PUREX
- Waste Fractionization at B Plant
- Plutonium Finishing Plant Operations.

Bismuth Phosphate Process. Silicon from the aluminum fuel jackets and the Al-Si bond layer are expected to have been in the CW stream. It is estimated that the CW stream contained 8.1 MT of silicon. The bismuth phosphate process introduced $(\text{NH}_4)_2\text{SiF}_6$ into the First-Cycle and Second-Cycle waste streams. Based on fuel process records and the flowsheet (Schneider 1951), it is estimated that 91.4 MT of silicon entered the First-Cycle waste and 79 MT entered the Second-Cycle waste. The flowsheet does not appear to include the 0.6 MT of silicon present as impurities in the uranium metal that would end up in either the metal waste or the First-Cycle waste.

Uranium Recovery Process. The Uranium Recovery Process was reviewed, and no silicon additions by the process were identified. Any silicon present in the bismuth phosphate metal waste (0.6 MT) will also show up in the uranium recovery feed and waste streams.

In-Farm Processing. The In-Farm Cesium Scavenging flowsheets and process documents do not indicate any additional sources of silicon.

REDOX Process. The REDOX process flowsheets and process documents were reviewed and do not indicate any additional sources of silicon other than fuel cladding. Silicon from the aluminum fuel jackets and the Al-Si bond layer are expected to have been in the cladding waste stream. Using fuel fabrication documents, it is estimated that the cladding

waste stream contained 25.1 MT of silicon. Another 1.5 MT of silicon was present as impurity in the uranium metal and would have ended up in the solvent extraction waste.

PUREX Process. The PUREX process flowsheets and process documents were reviewed and do not indicate any additional sources of silicon other than fuel cladding. Silicon from the aluminum fuel jackets and the Al-Si bond layer are expected to have been in the cladding waste stream. Using fuel fabrication documents, it is estimated that the CW stream contained 89.4 MT of silicon. Another 4.9 MT of silicon was present as impurities in the uranium metal and would have ended up in the solvent extraction waste.

Waste Fractionization (B Plant). The cesium and strontium waste fractionization and purification flowsheets do not indicate any additional sources of silicon.

Plutonium Finishing Plant. Silicon is a known constituent of the Plutonium Reclamation Facility (PRF) high salt waste, though at a low level, 42 mg/L (90 mg/L SiO₂). The PRF began operation in 1964 and ran through 1977, then ran again between 1983 and 1986. However, before May of 1973, waste from the Plutonium Finishing Plant was sent to cribs rather than to tank farms. The PRF does not significantly contribute to the silicon inventory.

Non-Routine Additions of Silicon During Chemical Process Operations. The REDOX and PUREX processes used SiO₂ gel beds to capture radoruthenium during the uranium purification process. The beds were regenerated and were not routinely replaced, if at all. It is not known if the SiO₂ beds were disposed of as tank waste or as solid waste, however, disposal as solid waste is considered more likely. The SiO₂ beds are considered to be a small and unlikely source of silicon.

Since the 1970's, spent ion exchange media from the 242-S and 242-A evaporators has been discharged to the Hanford Site underground waste tanks. The evaporators used a zeolite-based ion exchange media designed to physically degrade at a pH above 12. The 242-S Evaporator was started up in November 1971 and was shut down in 1981. The 242-A Evaporator was started up in March 1977 and was shut down in 1989. After completion of upgrades, the 242-A Evaporator was restarted in 1995. A partial inventory for this silicon source can be determined from essential material consumption records.

Material consumption records for post-1981 are not available; however, the annual volume of waste concentrated post-1981 is substantially lower than pre-1981 rates. Essential material consumption records for 1977 through 1981 indicate that the evaporators used 14.25 MT of Zeolon 900.¹ Zeolon 900 has a SiO₂/Al₂O₃ ratio of 10, which results in a partial estimate of 5.7 MT silicon from this source. The ion exchange media from the 242-S

¹Zeolon 900 was a synthetic zeololite used for ion exchange. It was used in the evaporator because it would polish ¹³⁷Cs. Because it was an inorganic, it could be dumped or flushed to the tanks. Zeolon 900 was a product of the Norton Co. and was later sold to the Philadelphia Quartz Corporation, Baltimore, M.D.

Evaporator was first dumped to tank 241-S-102, then later to tank 241-SY-102. Prior to 1981, the 241-A Evaporator dumped to tank 241-A-102 and later to tank 241-AW-102.

5.19.2.3 Process Chemical Impurities. Unintentional additions of silicon may have occurred due to silicon impurities in purchased chemicals. Perusal of the process facility technical manuals does not indicate that Hanford process facilities had unique silicon specifications for purchase of process chemicals. Since there is no evidence of a unique silicon specification, it is assumed that silicon content of purchased chemicals would be in compliance with industrial standards. The major process chemicals used at Hanford were NaOH, HNO₃, and ANN.

The industry specification for 50 wt% commercial grade caustic soda is 0.04 wt% SiO₂. The specification for 50 wt% rayon grade caustic soda is 0.002 wt% SiO₂. Section 5.20 estimates the total sodium discharged from the chemical process plants at 54,200 MT. Agnew et al. (1996) estimates 40,300 MT of sodium discharged from the plants. If it is assumed that all the sodium estimated in Section 5.20 was obtained as 50 wt% NaOH, then up to 75.4 MT silicon could have been introduced as a chemical impurity if commercial caustic soda was used, whereas only 4.0 MT silicon would have been used if the purer rayon grade was used. The incentive for using a better grade of caustic soda (rayon) would be to control the chloride content in the waste. Commercial grade caustic soda can contain up to 1.0 wt% NaCl. It is known that the chloride content of purchased chemicals has been a procurement specification for some time. Evidence of rayon grade caustic soda usage at the Hanford Site, however, has not been established.

The SiO₂ impurity level of other purchased chemicals is not well documented. Review of essential material reports (Palmer 1978 and Isochem 1966) provided little information on SiO₂ impurities beyond a 0.01 wt% SiO₂ limit for aluminum nitrate solution. Based on the estimate of aluminum introduced by ANN, as discussed in Section 5.1, the ANN could have introduced 5.6 MT of silicon. The essential material reports do not include a specification for SiO₂ in nitric acid. In addition, the inventory of virgin nitric acid used has not been established. The essential material reports do include a nitric acid limit for residue after evaporation of 0.01 wt%. This residue would come from a number of sources during nitric acid manufacture, and would more likely be iron and heavy metals than silicon.

Values for silicon introduced by corrosion and process water have been determined for PUREX Plant processing of N Reactor fuel. The values are 0.95 gmol/MTU and 0.81 gmol/MTU, respectively. If these values are applied to all of the fuel processed on the Hanford Site, the silicon introduced by these sources totals 2.6 MT and 2.3 MT, respectively. These values indicate that corrosion and process water are likely to be small contributors to the silicon inventory.

5.19.2.4 Addition by Tank Farm Activities. In 1966, tank farm operations added 57,000 kg of Portland cement to tank 241-BY-105 (Krieg et al. 1990). Portland cement contains 22 wt% SiO₂ (Kirk-Othmer 1979). During 1970 to 1972, a total of 354,000 kg of diatomaceous earth was added to tanks 241-BX-102, SX-113, TX-116, TX-117, TY-106, and

U-104 (Krieg et al. 1990). Chemical analysis for typical diatomaceous earth indicates a SiO_2 content of 92.3 wt%, dry basis (Buckingham and Metz 1974). The moisture content of the diatomaceous earth that was used is unknown. Using the masses of Portland cement and diatomaceous earth, and the respective weight fractions, an estimate of 159 MT of silicon is obtained for direct addition by tank farm activities.

5.19.2.5 Blowsand. Blowsand is dust or dirt that entered the tanks as material suspended in air. The following three pathways for entry of blowsand into the tanks were identified:

1. By the tank vents
2. From removal of cover blocks from pits
3. By direct addition.

The following analysis was developed from personal communications with Messrs. Russ Knight, Rex Bendixsen, and Fay Boyd, all of whom worked in or with Tank Farm Operations for many years and have first hand knowledge of blowsand accumulation during operations.

Tanks Vents. Tank vents were not equipped with HEPA filters until the mid 1970's. The SX, SY, A, AX, AY, AZ, AN, AW, and AP tanks were provided with forced ventilation. All other tanks were ventilated by breathing, i.e., through atmospheric pressure changes. The average dust loading of air on the Hanford Site according to Schreckhise et al. (1993) is $1.0\text{E}-04 \text{ g/m}^3$. This dust burden includes the contribution of dust storms. The SiO_2 content of blowsand is assumed to be the same as Hanford Site soil, 61 wt% SiO_2 .

Tanks with Forced Ventilation. The following additional parameters were assumed:

- Forced ventilation places a negative pressure on the tank. Air enters a tank with forced ventilation by various in-leakage paths and is unfiltered.
- Unfiltered forced ventilation continued for about 40 years in the SX and A farms based on completion of construction of SX farm in 1954 and A farm in 1955. All other farms with forced ventilation were completed after these dates.
- There are 53 tanks in the SX, SY, A, AX, AY, AZ, AN, AW, and AP farms.¹
- A conservatively high air flow rate of $1,000 \text{ ft}^3/\text{min}$ was assumed for each tank.

¹The A, AX, and AY tank farms were equipped with air-lift circulators. Thus, much of the inlet air for these tanks would have come from compressors. Although the compressors did not provide instrument quality air, the compressors were housed in buildings that would have protected the air supply from high dust levels.

- Limited forced ventilation was provided during sluicing operations and during certain maintenance activities. This contribution is considered small and has been ignored.

For the 15 oldest tanks in SX farm,

$$\frac{(15 \text{ tanks})(40 \text{ yr})(365 \text{ d/yr})(24 \text{ hr/d})(60 \text{ min/hr})(1,000 \text{ ft}^3/\text{min/tk})(1.0\text{E-}4 \text{ g/m}^3)}{(35.314 \text{ ft}^3/\text{m}^3)(1,000 \text{ g/kg})}$$

= 890 kg of soil could have collected.

Repeating and summing this calculation for all 9 ventilated tank farms results in an estimate of 2,000 kg of soil entering the tanks. The silicon load of the soil is 2.0 E+04 mol or 0.6 MT.

Tanks with natural ventilation. The following additional parameters were assumed:

- Unfiltered natural ventilation continued for about 30 years based on construction of the first farm being completed in 1944, and installation of HEPA filters in the mid 1970's.
- Garfield (1975) calculates the daily natural air breathing rate of a tank to be 0.00667 times the tank volume.
- The total volume of all DST and SST tanks including dome space is 7.24 E+05 m³ (RHO 1985a). This volume was not reduced to correct for either tanks under forced ventilation or for the tank volume occupied by waste.

$$\frac{(7.24 \text{ E+}5 \text{ m}^3)(0.00667/\text{d})(30 \text{ yr})(365 \text{ d/yr})(1.0\text{E-}4 \text{ g/m}^3)}{(1,000 \text{ g/kg})}$$

= 5.3 kg

These assumptions result in an estimate of 5.3 kg of soil entering the tanks. The silicon load of the soil is 54 mol or 1.5 kg.

Cover Block Removal. Tank pits associated with the tanks and the tank farm system are nearly flush with the ground and can accumulate sand in the joints between the cover blocks. The pits are used to house pumps, condensers, sluicing equipment, valves and jumpers. Sand enters the pit when the cover blocks are removed.

Pits on the tank or within a tank farm drain to the tank whereas pits outside the farms drain to a catch tank. Sand collected in a catch tank may or may not have been transferred by the catch tank pump to the waste tanks. Observations of the catch tank liquid level rates

of change during transfers indicate that solids heels formed a cone around the pump suction and were not readily transferred to the tanks.

Before completion of SX farm in 1954, the tanks and tank farms were not equipped with pump pits or valve pits. Many older tanks were retro-fitted in the early 1950's with pits to support sluicing operations. After the mid 1970's, attempts were initiated to seal the cover blocks to prevent infiltration of water. Initial efforts used metal foil tape. Sealing of the cover blocks would also minimize collection of sand between the cover block joints.

During tank sluicing operations (1953 to 1957) removal of cover blocks was an extremely frequent operation, on the order of once per shift. A second period of frequent cover block removal occurred during the late 1970's in support of saltwell pumping. The amount of blowsand accumulated by this activity has not been considered due to uncertainty in the sealing of pits. Typical removal rates for cover blocks during times with more routine operations were much less. A removal frequency of twice per week is assumed.

Estimates of the typical volume of blowsand found in a pit range from 15 to 50 lb (0.15 to 0.5 ft³). The estimate assumes the volume of blowsand introduced during removal of cover blocks to be 0.25 ft³ (about 7 L).

Cover Block Removal for Sluicing Operations. The time period of active sluicing was 5 years, from 1953 to 1957.

$$(5 \text{ yr})(365 \text{ d/yr})(3 \text{ shift/d})(1 \text{ removal/shift})(0.25 \text{ ft}^3/\text{removal}) \times$$

$$(100 \text{ lb/ft}^3)(453 \text{ g/lb})/(1000 \text{ g/kg}) = 62,000 \text{ kg.}$$

Cover Block Removal for Routine Operations. The time period for the less active routine operations was 20 yrs., from 1957 to the mid 1970's.

$$(20 \text{ yr})(52 \text{ wk/yr})(2 \text{ removals/wk})(0.25 \text{ ft}^3/\text{removal}) \times$$

$$(100 \text{ lb/ft}^3)(453 \text{ g/lb})/(1,000 \text{ g/kg}) = 24,000 \text{ kg.}$$

These assumptions result in a total estimate of 86,000 kg of blowsand entering from cover block removal. The silicon load of the soil is 8.7 E+05 mol or 24 MT. This estimate is highly sensitive to amount of blowsand assumed to collect in the pits. Contributions from pits where the cover blocks were left off for an extended period of time may not fall within this assumption. Distribution of the blowsand among the tanks would depend on the frequency of pit activity.

Direct Addition. One occasion is known where soil was placed in the SX-101 pump pit for shielding during maintenance. When the maintenance was completed, the soil was washed into the tank. A volume of one cubic meter is assumed. This assumption result is

an estimate of 1,600 kg of blowsand entering by this single event. The silicon load of the soil is $1.6 \text{ E}+04$ mol or 0.45 MT.

5.19.2.6 Silicon Inventory Evaluation Summary. Table 5.19-2 summarizes the sources of silicon identified in this section. Estimates of silicon identified in Table 5.19-2 have primarily focused on discharge records and process flowsheets of the chemical processes. It does not appear that any of these previous sources included silicon from chemical impurities, materials added to the tanks or from blowsand. These sources could add as much as 230 MT to the current inventory.

The bismuth phosphate process flowsheet appears to base the silicon values on chemical usage. There is no evidence that silicon impurities were considered. The REDOX and PUREX process flowsheets do not consistently track the silicon introduced by the fuel. The following points are noted:

- The major source of SiO_2 appears to be from fuel fabrication and process chemicals.
- Direct addition of diatomaceous earth and Portland cement to the tanks appears to be the next most significant source.
- The most significant source of blowsand is entry through crevices in the cover blocks. This estimate is highly dependant on the estimate of typical blowsand accumulation and the frequency of cover block removal. The assumptions used are based on interviews with experienced tank farm operating and engineering personnel. Using maximum estimates for blowsand accumulation and cover block removal frequency could easily increase this impact by a factor of four.

A parallel concern from blowsand might be the impact on the aluminum inventory. Typical Hanford Site soil contains 13.6 wt% Al_2O_3 . The 88,000 kg of blowsand estimated to enter the tanks would contain $1.17 \text{ E}+05$ mol or 3.2 MT of aluminum. This is less than 0.1 percent of the aluminum thought to be in the tanks from fuel fabrication and process chemicals.

Table 5.19-2. Summary of Silicon Sources. (2 Sheets)

Source	Mass of Silicon (MT)	Remarks
Fuel Fabrication		
Al-Si Braze Metal	118.3	
Uranium metal	7.0	
Aluminum Jacket	4.3	
Zirconium Clad Fuel	0.8	

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Table 5.19-2. Summary of Silicon Sources. (2 Sheets)

Source	Mass of Silicon (MT)	Remarks
Chemical Processing		
BiPO ₄ 1st Cycle	91.4	
BiPO ₄ 2nd Cycle	79.0	
Uranium Recovery	--	No sources of silicon identified
In-Farm Processing	--	No sources of silicon identified
REDOX Plant	--	No sources of silicon identified
PUREX Plant	--	No sources of silicon identified
Waste Fractionization	--	No sources of silicon identified
Plutonium Finishing Plant	--	No significant source of silicon identified
Non-Routine Process Additions	5.7	
Chemical Impurities		
NaOH	75.4	
HNO ₃	--	Not a likely source of silicon
ANN	5.6	
Others	--	
Corrosion	2.6	
Process water	2.3	
Direct Addition by Tank Farms		
Diatomaceous Earth and Portland Cement	159	
Blowsand		
Forced Ventilation	0.6	
Natural Ventilation	<0.1	
Cover Block Removal	24	
Soil Addition	0.45	
Total Silicon Sources	570.8	

Hanford Defined Waste Inventory. The HDW model exhibits several discrepancies with the independent evaluation compiled in this report. For the REDOX process, the HDW model report assumes a value of 650 g Si/MTU coming from the fuel and 1,550 g Si/MTU from unidentified sources. The unidentified sources assumption (1,550 g Si/MTU) was included in the HDW model to account for the high silicon inventories found in the REDOX waste tanks. The silicon introduced by fuel is derived from REDOX Flowsheet 6 (Crawley and Harmon 1960b). Fuel fabrication records and fuel process history, used in the independent evaluation, however, indicate that the silicon from fuel ranged from 1,010 to 1,610 g Si/MTU. The bulk of the fuel processed through the REDOX Plant were enriched I&E elements with a silicon value of 1,610 g Si/MTU. Sources such as chemical impurities and blowsand could account for the rest of the discrepancy with the silicon found in the REDOX Plant waste tanks.

For the PUREX process, the HDW model report assumes 4,025 g Si/MTU for all of the years that the PUREX Plant operated, including the years when only zirconium-clad fuel was processed. This value appears to be excessive as most of the fuel run in the PUREX Plant was either natural I&E fuel at 1,350 g Si/MTU or zirconium-clad N Reactor fuel at 131 g Si/MTU.

The HDW inventory does not appear to have, as yet, incorporated the diatomaceous earth and Portland cement additions made by Tank Farms Operations.

Tank Waste Remediation System Inventory. Review of the TWRS inventory identified a calculation error that decreases the silicon estimate from 693 to 603 MT silicon. In addition, it was noted that 96 MT of silicon is attributed to Tank 241-AY-101. This value is based on a sample result and cannot be reconciled by the tank fill history. Although the sample result cannot be ruled out, it is considered suspect.

5.19.3 Best-Basis Estimate for Silicon Value

The best-basis value for the silicon inventory is the value developed by the independent evaluation, 570 MT. The reasons for using the independent evaluation value are as follows:

- The assumptions in the HDW inventory cannot be reconciled with fuel fabrication records, and several sources, diatomaceous earth, Portland cement, and blowsand have not been included.
- The technical basis for the TWRS SST inventory is limited to process knowledge of the reprocessing facilities. The basis for silicon in the DSTs may be biased toward a high value because of the Tank 241-AY-101 sample.
- The values for the majority of the silicon sources are based, to a large degree on subjective reasoning and have a large element of uncertainty. Use of a large uncertainty value (± 25 percent) is recommended.

5.20 SODIUM

The following were the primary sources of sodium, the primary nonvolatile constituent in tank wastes:

1. Sodium hydroxide (NaOH) used as follows:
 - a. Dissolve the aluminum cladding from irradiated reactor fuel and targets.
 - b. Neutralize acidic wastes resulting from separations of Pu, U, Th, and other byproducts from acid dissolved reactor fuel and targets.
 - c. Neutralize the acidic cladding waste from Zircaloy®-clad fuels.
2. Sodium nitrate (NaNO₃), which was used in the NaOH dissolution of aluminum cladding to suppress hydrogen evolution.
3. Sodium carbonate (Na₂CO₃), which was used to treat solvent extraction organic solvents for recycle and to maintain product quality.
4. Sodium nitrite (NaNO₂), which was used as an additive to neutralized alkaline wastes to inhibit stress corrosion cracking in the carbon steel waste storage tanks and to oxidize Pu(III) to Pu(IV) in the separations plants.
5. Sodium bisulfate and sodium sulfate, which were used for fission product recovery and strontium product purification.

Sodium is widespread throughout the waste tanks. Sodium is a monovalent alkaline metal and essentially all sodium salts are soluble in acidic and alkaline solutions. In the Hanford Site wastes, approximately 99 percent of the total sodium is expected to be water soluble. The insoluble 1 percent of the sodium is found in the sludges as complex mixed metal compounds. Soluble sodium in the tank wastes is associated with alkaline soluble compounds such as sodium nitrate (NaNO₃), sodium nitrite (NaNO₂), sodium hydroxide (NaOH), sodium phosphate (Na₃PO₄), sodium sulfate (Na₂SO₄), sodium fluoride (NaF), sodium chloride (NaCl), sodium aluminate (NaAlO₂), sodium chromite (NaCrO₂), and sodium dichromate (Na₂Cr₂O₇).

The wastes were processed for solids removal (i.e., settling) and/or volume reduction after they were routed to the tanks. The volume reduction processes included scavenging, evaporation, and crystallization processes.

The scavenging process treated supernatants containing sodium for cesium and strontium removal and discharged the treated supernatants to cribs for disposal of the decontaminated sodium salt solutions.

The evaporation processes using the 242-B and 242-T evaporators removed water to reduce tank waste volume.

The later crystallization processes evaporated additional water from the supernatants to crystallize sodium nitrate salts and further reduce tank waste volume. The saturated mother liquor from this crystallization in the tanks is drained from the salt crystals by the process of "saltwell pumping" and transferred to the DSTs to become DSSF. The residual SST interstitial liquor and DSSF contains the remaining uncrystallized sodium nitrate and the other soluble sodium salts defined earlier.

As a result, the SSTs currently contain approximately 60 percent of the total waste sodium as crystallized sodium nitrate, 10 percent associated with SST sludges, and 10 percent as interstitial solutions with high sodium concentrations. The DSTs contain the remaining approximately 20 percent of the total waste tank inventory of sodium as supernatants.

The crystallized sodium nitrate redissolves into solution with the addition of water. The retrieval of SST tank wastes is planned to use "past practice" sluicing with water to dissolve the crystallized salt cake and remaining interstitial liquor.

5.20.1 Identification of Sodium Inventory Values

The reported global sodium inventory values are 40,300 MT (HDW model, Agnew et al. 1996), 68,500 MT (TWRS, Shelton 1996), and 54,200 MT by this report. A separate analysis of current SST waste salt cake, drainable liquids, and sludge volumes, combined with the DST sodium inventory, gives a potential total sodium inventory in the tank wastes of 54,900 MT.

The methodology and analysis to obtain a global sodium inventory of 54,200 MT sodium is contained in the following sections.

5.20.2 Sodium Inventory Evaluation

The Hanford Site tank sodium inventory is estimated by computing the total amount of sodium added to each of the separations processes and subtracting the amounts of sodium in the wastes that were discharged to cribs and trenches. Crib transfers are especially important because these transfers are used to estimate the average sodium concentration and total volume of BiPO_4 and UR process wastes that were left in the tanks (Waite 1991, Anderson 1990). Other sources of useful information include the following:

1. Process flowsheets (Schneider 1951, GE 1951b, Merrill and Stevenson 1955, Crawley 1960, Isaacson 1965, Boldt 1966, Schofield 1991).
2. Tank transaction records and estimates from the HDW model (Agnew et al. 1995a and Agnew et al. 1996, respectively).
3. Partial chemical usage summaries (McDonald 1968, McDonald and Hogan 1977, Hogan 1992, and Jenkins and Foster 1978).
4. Process production records (Appendix B).
5. Tank samples and tank inventory estimates derived from supernatant and core samples (Hodgson 1995, Ryan 1995).

These sources are used to develop sodium inventory estimates for each of the chemical separations processes.

5.20.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process operated at T Plant from December 1944 to 1956 and at B Plant from April 1945 to 1952. The site production records indicate that 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the BiPO_4 processing is developed in Appendix C based on the flowsheet by Schneider (1991). Comparable HDW sodium values are obtained from the "waste volume out" and "sodium concentrations" of the HDW. The HDW model sodium concentrations used are attributed to Jungfleisch (1980, 1983, and 1984) and Lucas (1989).

The sodium usage in the BiPO_4 processing is shown in Table 5.20-1 for the Schneider (1991) flowsheet and for the HDW model (Agnew et al. 1996).

Table 5.20-1. Amount of Sodium in BiPO_4 Process Wastes, MT.

Waste stream	Present estimate (Schneider 1991)	HDW model (Agnew et al. 1996)
Metal waste (MW)	10,430	7,590
First-cycle and coating waste (1C/CW)	5,740	6,110
Second decontamination cycle waste (2C)	4,010	4,280
224 Waste	4,260	1,300
Total	24,400	19,280

HDW = Hanford Defined Waste.

Metal Waste. The first steps in the BiPO_4 process dissolved the uranium metal slugs and extracted the plutonium by carrier precipitation with bismuth phosphate. This resulted in the metal waste stream. The metal waste stream contained the bulk of the uranium and approximately 90 percent of the long-lived fission products (e.g., ^{137}Cs and ^{90}Sr) and was sent to the SSTs for storage.

The stored metal waste containing the uranium from BiPO_4 processing was later retrieved as the feed for the UR process. Approximately 73 percent of the uranium is thought to have been associated with the sludge, as $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$ and NaUO_2PO_4 , while the remainder was dissolved in the supernatant (GE 1951b, page 203). Furthermore, it appears that 5.5 percent of the metal waste sludge was not recovered from the tanks during the sluicing campaign (Rodenhizer 1987). Two-thirds of the uranium in the metal waste sludge was in the form of $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$ and the remainder as NaUO_2PO_4 . The following calculation indicates the amount of sodium that might be in the tanks with the residual metal waste.

$$U(\text{MW}) = (0.73)(0.055)(7,800 \text{ MTU}) = 310 \text{ MTU}$$
, where $U(\text{MW})$ is the amount of residual metal waste uranium in the tanks.

$$\text{Na}(\text{MW}) = (U)[(0.66)(1/0.432)(0.170) + (0.34)(1/0.6134)(0.0593)]$$
, where:

0.66 equals the fraction of uranium in the form of $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$,

0.432 equals the fraction of uranium in $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$,

0.170 equals the fraction of sodium in $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$,

0.34 equals the fraction of uranium in the form of NaUO_2PO_4 ,

0.6134 equals the fraction of uranium in NaUO_2PO_4 ,

0.0593 equals the fraction of sodium in NaUO_2PO_4 .

$$\text{Na}(\text{MW}) = 91.5 \text{ MT}$$
 (from residual MW in the tanks).

Thus, 10,340 MT of sodium in the metal wastes was transferred to the Uranium Recovery process from the total 10,430 MT of sodium initially produced in the metal waste.

Cribbed BiPO_4 Wastes. Portions of the other BiPO_4 waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The total sodium discharged to cribs and specific retention trenches was 9,850 MT sodium in 259,000 m^3 (68.5 Mgal) (Waite 1991).

The HDW model estimates 6,420 MT sodium was cribbed from the BiPO_4 process in 173,000 m^3 (45.8 Mgal) (Agnew et al. 1996). The HDW estimate for BiPO_4 wastes is obtained from the crib volume and the supernatant sodium concentration of Agnew et al. (1996).

Summary for BiPO₄ Process Wastes. The residual MW, 1C and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.20-2. This report estimates the residual BiPO₄ tank wastes contain 4,210 MT of sodium versus the 5,180 MT estimated by the HDW model.

Table 5.20-2. Estimated Amount of Sodium in BiPO₄ Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes produced	24,400	19,280
BiPO ₄ wastes cribbed	9,850	6,420
BiPO ₄ wastes sent to uranium recovery	10,340	7,680
BiPO ₄ wastes in SSTs/DSTs	4,210	5,180

DST = Double-shell tank
HDW = Hanford Defined Waste
SST = Single-shell tank.

The principal differences in the present estimate and the HDW model are: (1) the input chemicals in the flowsheets (assumed), (2) the volume of cribbed BiPO₄ wastes, and (3) a significant difference in the amount of sodium contained in the wastes sent to UR.

5.20.2.2 Uranium Recovery Process Wastes. The UR process began in 1952 to extract U from the BiPO₄ process MW stored in B, BX, BY, C, T, TX, and U Tank Farms.

The design basis flowsheet for the uranium recovery process is provided in the *Uranium Recovery Technical Manual* (GE 1951a). The process operated with a highly variable feed due to variations in the quality of retrieved metal waste sludges and supernatants. The variation of uranium content during the retrieval operation results in variation of required nitric acid for dissolution and resulting sodium hydroxide for subsequent neutralization. As a result, the UR flowsheet (GE 1951a) is not expected to provide an accurate estimate of uranium recovery chemical usage.

Cribbed Uranium Recovery Wastes. The HDW model analysis of transfer records from the UR operation indicates that 62.5 vol% of the total UR waste streams were cribbed and 37.5 vol% of the total UR waste streams were routed to the SSTs. The HDW model estimate for UR wastes is obtained from the crib volume and the supernatant sodium concentration of Agnew et al. (1996). The sodium concentrations are attributed to Borsheim and Simpson (1991) and Sloat (1954).

Waite (1991) states that 155,000 m³ (41 Mgal) of scavenged UR wastes containing 13,100 MT of sodium was discharged to cribs and specific retention trenches. This analysis assumes that the sodium discharged to cribs and specific retention trenches is equal to the vol% split determined by the HDW model.

Uranium Recovery Chemicals Added. The assumption of 62.5 percent of the UR waste sodium being cribbed results in a total UR waste sodium content of 20,960 MT. The sodium content of the residual in tank UR wastes and UR chemicals added are 7,860 MT and 10,620 MT, respectively. The uranium chemical sodium addition of 10,620 MT is approximately 20 percent greater than the flowsheet predicted value (GE 1951a) of 8,880 MT, as expected.

For the HDW model the amount of sodium added during UR is the difference in the sodium content between the UR wastes produced and the BiPO₄ wastes sent to Uranium Recovery. The HDW model sodium values in the UR wastes produced (24,600 MT sodium) are obtained from the "waste volume out" and "sodium concentrations" of the HDW model. The HDW model sodium concentrations used are attributed to Borsheim and Simpson (1991) and Sloat (1954). Subtraction of the sodium inventory to UR (7,680 MT sodium) gives a 16,920 MT sodium chemical addition in UR.

Summary for Uranium Recovery Process Wastes. Table 5.20-3 provides a summary sodium material balance for the UR process for this analysis and the HDW model. This report estimates the residual UR tank wastes contain 7,860 MT of sodium versus the 11,300 MT estimated by the HDW model.

Table 5.20-3. Estimated Amount of Sodium in Uranium Recovery Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes sent to UR	10,340	7,680
UR chemicals added	10,620	16,920
UR wastes produced	20,960	24,600
UR wastes cribbed	13,100	13,300
UR wastes in SST/DST's	7,860	11,300

DST = Double-shell tank

HDW = Hanford Defined Waste

SST = Single-shell tank

UR = Uranium Recovery.

The difference in analysis that provided the results in Table 5.20-3 are as follows:

1. UR feed - Values calculated in Section 5.20.2.1.
2. UR wastes produced.
 - a. Present estimate of UR wastes produced equals sodium reported cribbed (Waite 1991) divided by 0.625 (the volume fraction of UR wastes cribbed [Agnew et al. 1996]).
 - b. The HDW model basis for UR wastes produced is Sloat (1954) and Borsheim and Simpson (1991).
3. UR chemicals added - the difference between (2) and (1).
4. UR wastes cribbed.
 - a. Present estimate based on Waite (1991).
 - b. The HDW basis uses transfer records and calculated sodium solubility.

Due to the lack of definitive data on the chemical usage in uranium recovery and the sodium content of uranium recovery sludges, there is a significant uncertainty in the amount of sodium associated with residual uranium recovery wastes in the waste tanks. The HDW basis for sodium content in cribbed UR wastes result in a greater fraction of sodium in the residual UR tank wastes, 46 percent versus 32.5 percent. The uncertainty in sodium content in residual UR tank wastes could be several thousand MT of sodium.

5.20.2.3 REDOX Process Wastes. The REDOX process used solvent extraction and aluminum nitrate as a salting agent to recover uranium, plutonium, and neptunium from irradiated fuel. This process started up in January 1952. During the period from 1952 to 1966, 19,706 MTU were processed through the REDOX Plant (Appendix B). Sodium usage in the REDOX process included: (1) sodium hydroxide to de-clad irradiated fuel and neutralize the solvent extraction wastes for SST storage, (2) sodium nitrate to suppress hydrogen during fuel de-cladding, and (3) sodium dichromate to adjust plutonium valence for solvent extraction recovery.

Sodium Hydroxide. The synopsis of REDOX Plant operations (Jenkins and Foster 1978) was used as a basis for estimating the total amount of sodium from sodium hydroxide in the REDOX process wastes. Table 5.20-4 provides the annual sodium hydroxide usage for REDOX (Jenkins and Foster 1978).

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Table 5.20-4. NaOH Added to REDOX Process. (2 Sheets)

Year	MTU processed	NaOH, kg	Na, kg/MTU	Na, MT
1952	745.8	3,262,500	2,520	1,876
1953	1,297.8	4,225,000	1,872	2,430
1954	1,432.7	4,367,000	1,753	2,510
1955	2,200.5	4,852,000	1,269	2,790
1956	2,118.5	3,609,000	980	2,070
1957	1,658.8	2,329,000	807	1,340
1958	1,274.0	1,631,000	736	938
1959	831.5	1,051,000	727	604
1960	755.9	1,081,000	822	622
1961	1,144.6	1,312,000	659	754
1962	892.1	1,108,000	714	637
1963	1,054.8	1,202,000	656	691
1964	1,695.0	1,770,000	601	1,018
1965	1,407.6	1,670,000	683	960
1966	1,196.8	1,657,000	796	953
Total	19,706	35,130,000	-	20,200

MT = Metric ton

MTU = Metric ton uranium

REDOX = Reduction and oxidation.

A comparison of flowsheet derived sodium estimates to reported REDOX Plant operating usage (Jenkins and Foster 1988) is provided in Table 5.20-5.

Table 5.20-5. Comparison of REDOX Flowsheets to REDOX Plant Operations Synopsis for Sodium Hydroxide.

Year	Flowsheet	Na from NaOH, kg/MTU ^a	Na from NaOH, kg/MTU ^b	Ratio
1952	No. 4	1,970	2,520	1.28
1956	No. 5	638	980	1.54
1961	No. 6	664	659	1.01
1964	No. 7	357	601	1.68
1965	No. 8	443	683	1.54
1966	No. 9	409	741	1.81

MTU = Metric ton uranium

REDOX = Reduction and oxidation

^aFlowsheets No. 4, 5, 6, 7, 8, and 9 (GE 1951b, Merrill 1955, Crawley 1960, Isaacson 1965, Boldt 1966).

^bSynopsis of REDOX Plant Operations (Jenkins and Foster 1978).

Because the REDOX product streams were frequently reworked, the plant experienced frequent start ups and shut downs (five day a week operation for flowsheets 4 and 5), and because of the specialty processing campaigns for neptunium and nonstandard fuel (performed during the periods represented by Flowsheets 7, 8, and 9), the REDOX flowsheets cannot be used to accurately account for all of the chemicals added to the process. The flowsheet provides chemical usage at equilibrium operation. The nonstandard operations result in chemical use higher than predicted by the flowsheets. Therefore, the reported usage of sodium hydroxide versus flowsheet values shown in Table 5.20-5 is reasonable.

It also appears from REDOX history (personal communication, R. A. Watrous), that Flowsheet 5 was probably issued in advance of certain changes in the plant configuration, and Flowsheets 7, 8, and 9 well after completion of the indicated changes. The REDOX operations summarized in the synopsis of Jenkins and Foster (1978), therefore, seem to be a more appropriate basis for defining the composition of the REDOX process waste.

Sodium Nitrate. Tank waste supernatants were used after January 1963 to supply all of the sodium nitrate needed for decladding operations. This change was documented in Flowsheets 7 and 8 (Isaacson 1965).

During the period of 1952 through 1962, 14,350 MT of U was processed at the REDOX Plant. Appendix D shows that REDOX used about 150 kg NaNO₃ per MTU for the decladding operation. This results in an estimated REDOX use of 2,150 MT NaNO₃ from 1952 through 1962 containing 580 MT of sodium.

Sodium Dichromate. Sodium dichromate use at Hanford is discussed in Section 5.7 with an estimate of 749 MT of sodium dichromate used at the REDOX Plant. There is 330 MT sodium associated with the 749 MT sodium dichromate used by the REDOX process.

Summary for REDOX Process Wastes. Sodium use by the REDOX Plant is summarized in Table 5.20-6 for this analysis and the HDW model.

Table 5.20-6. Estimated Amount of Sodium in REDOX Wastes, MT.

Chemical Source	Present estimate	HDW model (Agnew et al. 1996)
Sodium hydroxide	20,200	NA
Sodium nitrate	580	NA
Sodium dichromate	330	NA
Total	21,100	11,970

HDW = Hanford Defined Waste

NA = Not attributed to individual chemicals.

This analysis gives a total REDOX sodium usage of 21,100 MT sodium based primarily on reported NaOH consumption (95 percent) and secondarily on flowsheet analysis (5 percent) of NaNO_3 and $\text{Na}_2\text{Cr}_2\text{O}_7$ usage. The HDW model gives a total of 11,970 MT sodium based on transfer records and unreferenced flowsheet compositions. Agnew et al. (1996) does not provide references for the REDOX waste compositions used. Agnew (1995) indicates eight references (GE 1951b, Merrill 1955, Crawley 1960, Isaacson 1965, Allen 1976, Jenkins and Foster 1978, Jungfleisch 1984, and Anderson 1990) were used to define the compositions of the four REDOX waste streams but does not state the specific reference or methodology.

5.20.2.4 PUREX Process Wastes. The PUREX process came on line in January 1956. The PUREX Plant processing can be broken down into three main periods: 1983 through 1989, 1965 through 1982, and 1956 through 1964.

1983 through 1989. The period from 1983 through 1989 processed Zircaloy®-clad fuel exclusively. The NCAW during this period was effectively segregated in two DSTs and the sodium content can be determined by sample analysis. The NCRW was distributed throughout the DSTs by decanting and concentrating. The NCRW sodium content can be estimated by analysis of the flowsheet.

The NCAW sodium content is determined by samples (Hodgson 1995, Ryan 1995) and summarized in Shelton (1996). The NCAW produced from 1983 through 1989 contains 577 MT total sodium in the combined sludge and supernatant as shown in Table 5.20-7.

Table 5.20-7. Estimated Amount of Sodium in 1983 through 1989 PUREX Neutralized Current Acid Waste, MT.

Tank	Present estimate ^a	HDW model (Agnew et al. 1996)	
		P3 waste	PL2 waste
241-AZ-101	384	170	-
241-AZ-102	193	97	-
Other DSTs	-	17	641
Total	577	922	

DST = Double-shell tanks

HDW = Hanford Defined Waste

P3 = Aging waste from PUREX process high-level waste (1983 to 1988)

PL2 = PUREX process low-level waste (1983 to 1988)

^aShelton 1996.

During 1983 through 1989, PUREX processed 3,900 MTU of irradiated fuel through solvent extraction by recycling 2,450 MTU of cold uranium to maintain the extraction column throughput above the minimum processing rate. The total 577 MT sodium in the NCAW from processing a total of 6,350 MTU equals a sodium usage of 91 kg Na/MT total U. Schofield (1991) estimated that 102 kg Na/MTU as NaOH was used to neutralize NCAW from the PUREX process.

The basis for the PUREX 1983 through 1989 wastes in Agnew et al. (1996) is not defined. The quantities and distribution of sodium in the P3 and PL2 wastes are defined by volume, concentration, and distribution of Agnew et al. (1996).

The quantity of sodium in the NCRW stream from PUREX Plant from 1983 through 1989 was assessed by Schofield (1991). Schofield reviewed core samples and flowsheet estimates to determine the amount of sodium in the PUREX process wastes produced from reprocessing N Reactor fuel. Schofield estimated that 704 MT of sodium as NaOH was used to neutralize the cladding waste from decladding 3,900 MTU of irradiated fuel from 1983 through 1989.

The quantity of sodium in NCRW stream from the PUREX Plant from 1983 through 1989 is defined by Agnew et al. (1996) by the volume and the sodium concentration for the CWZr2 stream. These assumptions give a total of 492 MT sodium in the PUREX Plant NCRW from 1983 through 1989.

The total sodium use by the PUREX Plant from 1983 through 1989 is summarized in Table 5.20-8. Altogether, 1,280 MT of sodium are estimated to have been used in the PUREX process from 1983 through 1989.

Table 5.20-8. Estimated Amount of Sodium in 1983 through 1989
PUREX Plant Wastes, MT.

Stream	Present estimate	HDW model (Agnew et al. 1996)
NCAW (PL3/PL2)	577	922
NCRW (CWZr2)	704	492
Total	1,280	1,414

CWZr2 = REDOX process coating waste, zirconium cladding

PL2 = PUREX process low-level waste (1956 to 1976)

PL3 = PUREX process low-level waste (1983 to 1988)

HDW = Hanford Defined Waste

NCAW = Neutralized Current Acid waste

NCRW = Neutralized Cladding Removal Waste.

1965 through 1982. The site production records indicate 25,179 MT of U and 629 MT of Th were processed in the PUREX Plant from 1965 to 1972 (Appendix B). The PUREX Plant was maintained in standby from 1973 through 1982. Monthly essential material usage records can be used to define the chemical consumption for the PUREX Plant during the 1965 to 1982 period (McDonald 1968, McDonald and Hogan 1977, and Hogan 1992). Data from these essential material use records are summarized by year in Table 5.20-9.

Table 5.20-9 shows that the total sodium use by PUREX Plant processing in the period of 1965 through 1982 is 5,590 MT sodium. The sodium sulfate use in 1965 through 1967 was for fission product recovery by sulfate strike precipitation.

1956 through 1964. The third period covers the PUREX production process from 1956 through 1964. During this period, 42,320 MTU were processed in the PUREX Plant (Appendix B). From 1956 through 1964, only aluminum-clad natural uranium fuel was processed through PUREX.

Examination of the PUREX processing history in Table 5.20-9 shows that aluminum-clad fuel was predominately process during 1965 and 1967. The average chemical usage in 1965 and 1967 was 230 kg of NaOH/MTU, 14 kg of Na₂CO₃/MTU, 140 kg of NaNO₃/MTU and 11 kg of NaNO₂/MTU.

Since sugar denitration was not used until the early 1960's, more sodium was probably required for waste neutralization during this era. Sugar denitration destroys about two-thirds of the nitric acid in current acid waste (Willett 1981). Based on these values, about 60 kg of additional sodium per MTU would have been needed for acid neutralization for a total of 290 kg NaOH/MTU during the 1956 through 1964 period.

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Table 5.20-9. Sodium Chemical Use in the PUREX Process from 1965 through 1982.

Year	MT processed			Chemical use, MT				
	Al clad U	Zr clad U	ThO ₂	NaOH	NaNO ₃	Na ₂ CO ₃	NaNO ₂	Na ₂ SO ₄
1965 ^a	5,385		5	1,190	746	77	62	142
1966 ^a	3,806		197	1,225	605	54	42	119
1967 ^a	4,359	123		1,067	630	58	43	23
1968 ^b	3,393	427		1,031	557	38	34	
1969 ^b	2,568	228		529	538	29	32	
1970 ^b	555	92	427	749	240	71	11	
1971 ^b	2,988	287		742	476	30	18	
1972 ^b	552	416		591	95	40	13	
1973 ^b								
1974 ^b								
1975 ^b								
1976 ^b								
1977 ^b								
1978 ^c				24				
1979 ^c				39				
1980 ^c				37				
1981 ^c				14	1			
1982 ^c				48			2	
Total	23,706	1,572	629	7,286	3,887	396	258	284
Subtotal Na, MT	-	-	-	4,190	1,052	172	86	92
Total Na, MT	5,590 ^d							

^aMcDonald (1968)

^bMcDonald and Hogan (1977)

^cHogan (1992)

^dTotal PUREX process sodium from 1965 through 1982.

Agnew (1995) identified a sulfate strike operation in the PUREX Plant in 1960 and 1961 based on examination of PUREX Plant monthly reports. This sulfate strike operation is similar to the operation performed in 1965 through 1967 that used 284 MT of sodium sulfate. An allowance of 150 MT of sodium sulfate is assumed for the sulfate strike operation in 1960 and 1961.

The resulting estimated chemical use in PUREX Plant for 1956 through 1964 is shown in Table 5.20-10. The total PUREX process sodium use from 1956 through 1964 is estimated at 9,120 MT sodium. This is a use of 215 kg Na/MTU. The PUREX process use should be 204 kg Na/MTU, based on the original flowsheet (GE 1955). The estimated use is higher than the flowsheet value due to inefficiencies of rework of products and startups/shutdowns.

Table 5.20-10. Sodium Chemical Use in the PUREX Process from 1956 through 1964.

Year	MTU processed	Estimated chemical use, MT				
		NaOH, 265 kg/MTU	NaNO ₃ , 140 kg/MTU	Na ₂ CO ₃ , 14 kg/MTU	NaNO ₂ , 11 kg/MTU	Na ₂ SO ₄ , allowance
1956	2,286	663	320	32	25	
1957	3,989	1,157	558	56	44	
1958	4,707	1,365	659	66	52	
1959	4,635	1,344	649	65	51	
1960	5,599	1,624	784	78	62	75
1961	5,438	1,577	761	76	60	75
1962	5,483	1,590	778	77	60	
1963	5,024	1,457	703	70	55	
1964	5,157	1,496	722	72	57	
Total	42,318	12,270	5,925	592	465	150
Subtotal Na, MT	-	7,060	1,603	257	155	50
Total Na, MT	9,120 ^a					

^aTotal estimated PUREX process sodium from 1956 through 1964.

Summary for PUREX Process Wastes. Sodium use by the PUREX Plant is summarized in Table 5.20-11. This analysis gives a total PUREX Plant sodium usage of 16,000 MT sodium based on the following:

1. Extrapolation of reported 1965 and 1967 usage (9,120 MT sodium, 57 percent).
2. Reported 1965 through 1982 usage (5,590 MT sodium, 35 percent).
3. DST analytical results for 1983 through 1989 (1,280 MT sodium, 8 percent).

Sodium use by the PUREX Plant for comparable periods in the HDW model (Agnew et al. 1996) was not determined in this report. The HDW model PUREX Plant sodium use was combined with B Plant fission product recovery wastes for comparison. This comparison is shown in a later section, Section 5.20.3.

Table 5.20-11. Estimated Amount of Sodium in the PUREX Plant Wastes, MT.

Years	Estimated sodium content from chemical use, MT Na					
	NaOH	NaNO ₃	Na ₂ CO ₃	NaNO ₂	Na ₂ SO ₄	Total
1956 - 1964	7,060	1,603	257	155	50	9,120
1965 - 1982	4,190	1,052	172	86	92	5,590
Subtotal, 1956 - 1982	11,250	2,650	429	241	142	14,710
Subtotal, 1983 - 1989	-	-	-	-	-	1,280
Total 1956 - 1989	16,000					

PUREX = Plutonium-uranium extraction.

5.20.2.5 B Plant Process Wastes. Monthly essential material usage records can be used to define the chemical consumption for B Plant and Tank Farms during the 1965 to 1982 period (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.20-12. The sodium bisulfate and sodium sulfate use in 1968 through 1979 was for strontium product purification before encapsulation.

Table 5.20-12 shows that the total estimated sodium use by B Plant and Tank Farms processing in the period of 1965 through 1982 is 4,910 MT sodium.

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Table 5.20-12. Chemical Use in B Plant and Tank Farms from 1965 to 1982.

Year	Chemical use, MT								
	NaOH		NaNO ₃		Na ₂ CO ₃ B Plant	NaNO ₂		NaHSO ₄ B Plant	Na ₂ SO ₄ B Plant
	B Plant	Tank Farms	B Plant	Tank Farms		B Plant	Tank Farms		
1965 ^a									
1966 ^a									
1967 ^a									
1968 ^b	789		17		32	5		7	
1969 ^b	624		55		54			41	
1970 ^b	707		4		38			24	
1971 ^b	982				221			2	33
1972 ^b	732				148				39
1973 ^b	495	76		45	2				29
1974 ^b	551	99		11	18				13
1975 ^b	516	69		12	14				5
1976 ^b	541	69		1	7				8
1977 ^b	654	78		1	5				1
1978 ^c	416	96			6				6
1979 ^c	141				27		2		1
1980 ^c	86			11			7		
1981 ^c	188		5				4		
1982 ^c									
Total	7,910		165		572	18		75	137
Subtotal Na, MT	4,550		44		248	6		14	44
Total Na, MT	4,910 ^d								

^aMcDonald (1968)

^bMcDonald and Hogan (1977)

^cHogan (1992)

^dTotal B Plant and Tank Farms sodium use from 1965 through 1982.

5.20.2.6 Other Cribbed Wastes. Agnew et al. (1996) defines portions of waste streams other than the BiPO_4 and UR process wastes discussed earlier as having been cribbed. These streams are identified in Agnew et al. (1996) as P1, CWP1, OWW1, DW, T1-SltCk, and WTR. The fraction of the streams cribbed is given in Agnew et al. (1996) Appendix D. The total sodium in the streams is given by the volume and the concentration. The summed products of these three values give a total calculated amount of sodium cribbed of 250 MT sodium.

This report's analysis is based on the reported cribbed sodium given by Waite (1991). The cribbed sodium content in BiPO_4 and Uranium Recovery wastes given in the previous sections includes all sodium cribbed in the common crib and specific retention sites. Therefore, the "other cribbed wastes" identified in Agnew et al. (1996) are included in the BiPO_4 or UR values identified by Waite (1991) and no separate value is identified by this report's analysis.

5.20.2.7 N Reactor Decontamination Wastes. Agnew et al. (1996) defined the residual amount of N Reactor decontamination wastes in the DSTs after a single vault of "Phosphate/Sulfate Waste" grout was produced. The N Reactor decontamination waste stream, "N," is defined in Agnew et al. (1996), Appendixes B and D, with composition attributed to Lucas (1989). The resulting residual N Reactor Decontamination Hanford tank inventory is 162 MT sodium.

5.20.3 Best-Basis Estimate for Sodium Inventory

Table 5.20-13 provides the results from this evaluation for sodium, together with comparable estimates from other sources. The best-basis global inventory of sodium in Hanford Site tank waste is about 54,200 MT. This estimate is significantly higher than the HDW model estimate of 40,300 MT sodium (Agnew et al. 1996) and significantly lower than the current value of 68,500 MT in the TWRS reference chemical inventory (Shelton 1996).

The present estimate is based primarily on reported chemical consumption for REDOX, PUREX, and B Plant; reported sodium crib discharges for BiPO_4 and Uranium Recovery; and flowsheet values of chemical consumption for the BiPO_4 process. The HDW model is based on flowsheet waste compositions and waste transfer records from Hanford Site processing plants. The TWRS inventory is based on the 1986 EIS estimate of Hanford Site tank inventory in 1983 plus additions to the tank system after 1983.

Table 5.20-13. Estimated Amount of Sodium in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
BiPO ₄	4,210	5,180	NA
Uranium recovery	7,860	11,300	NA
REDOX	21,100	11,970	NA
PUREX, B Plant, and Tank Farms (1956-82)	19,600	11,110	NA
Other cribbed wastes	-	(250)	NA
N Reactor decontamination	162	162	NA
PUREX (1983-89)	1,280	1,410	1,280
Total	54,200	40,300	68,500

HDW = Hanford Defined Waste
 NA = Not attributed to individual waste streams
 PUREX = Plutonium-Uranium extraction
 REDOX = Reduction and Oxidation
 TWRS = Tank Waste Remediation System.

The process wastes of PUREX, B Plant, and tank farms wastes in the period from 1956 through 1982 are combined in Table 5.20-13 for comparison purposes. The individual process contributions in the HDW model (Agnew et al. 1996) were not developed by this report. The 1956 through 1983 value of 11,110 MT sodium is determined by the difference of the total sodium (40,300 MT sodium, Agnew et al. 1996) and the sum of the individual streams discussed in Sections 5.20.2.1 through 5.20.2.7 (29,190 MT sodium).

An alternate method of determination of the Hanford Site waste tank sodium inventory based on reported waste volumes is available. The SSTs contain about 88,000 m³ of salt cake, 2,090 m³ of supernatant, 22,800 m³ of drainable liquid (based on 45 volume percent drainable liquid in the salt cake and 15 vol% drainable liquid in the sludge) and 45,400 m³ of sludge (Hanlon 1995). Hanlon (1995) uses reported liquid levels and assumes a 45 vol% salt cake porosity or void volume as equivalent to drainable liquid volume to conservatively project future saltwell pumping volumes. Based on historical saltwell pumping experience, it has been determined that the drainable liquid fraction of salt cake varies from 19 to 43 percent, with most salt cake tanks being in the range of 35 percent drainable liquid (DeWeese 1988).

Table 5.20-14 shows an estimated Hanford Site waste tank inventory of 54,900 MT sodium based on adjusting the reported waste volumes to 35 vol% salt cake porosity. For this analysis, porosity is considered equal to void volume in salt cake and filled with either supernatant or air. The volume-based estimate of 54,900 MT sodium compares favorably with the estimated 54,200 MT sodium inventory obtained by analysis of chemical use records, reported discharges to cribs and specific retention trenches, and flowsheet analysis.

The best-basis global inventory of 54,200 MT sodium has the highest uncertainty in the residual wastes produced by the BiPO_4 and Uranium Recovery processes. These wastes represent 20 percent of the estimated total, thus total uncertainty in the sodium inventory is low.

Also, the estimated total of 54,200 MT sodium may be biased high because the NaOH use in processing facilities included NaOH for regeneration of water demineralizers. Regeneration solutions of sodium hydroxide and sulfuric acid used in regeneration of demineralized water ion exchange beds was routed to chemical sewers in the regeneration process. The chemical consumption records quoted in the earlier sections are high for NaOH by this undefined amount of regeneration NaOH. The total amount of regeneration NaOH is expected to be low (less than 200 MT of contained sodium).

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Table 5.20-14. Estimated Amount of Sodium in the Hanford Site Tank Wastes based on Reported Waste volumes, MT.

Waste	Volume at 45 vol% salt cake drainable liquid, m ³ (Hanlon 1995)	Volume at 35 vol% salt cake drainable liquid, m ³	Assumed sodium concentration	Sodium, MT, at 35 vol% salt cake porosity ^a
SST sludge	45,400	45,400	-	3,800 ^b
SST salt cake volume including voids	88,000	88,000	-	-
SST salt volume without voids	48,400	57,200	2.26 g NaNO ₃ /cc 0.611 MT Na/M ³	35,000
SST sludge drainable liquid	6,810	6,810	-	-
SST salt cake drainable liquid	15,950	12,400	-	-
SST supernatant	2,090	2,090	-	-
Total SST drainable liquid	24,850	21,300	10M NaNO ₃ 0.230 MT Na/M ³	4,900
DST inventory ^c without future saltwell pumping or facility decommissioning	-	-	-	11,200
Total	-	-	-	54,900

DST = Double-shell tank

SST = Single-shell tank

^aFor this analysis, porosity is considered equal to void volume in salt cake

^bColton (1996)

^cShelton (1996).

5.21 SULFATE

The following are the primary sources of sulfate in tank wastes:

1. Sulfuric acid (H_2SO_4) used in the bismuth phosphate precipitation processes to separate uranium from plutonium and for plutonium decontamination.
2. Ferrous sulfamate ($Fe(NH_2SO_3)_2$) and ferrous ammonium sulfate ($Fe(NH_4)_2(SO_4)_2$) used to reduce plutonium valence for separation from uranium in the fuel reprocessing plants and the uranium recovery plant.
3. Sulfamic acid (NH_2SO_3H) used as a stabilizer to prevent oxidation and excessive use of ferrous ion reductant.
4. Sodium sulfate (Na_2SO_4) and sodium bisulfate ($NaHSO_4$) used for recovery and purification of fission products.

Sulfate is a divalent anion. The sodium salt is soluble in acidic and alkaline solutions. The calcium, strontium, and barium salts are insoluble in alkaline solution and resist metathesis to the soluble sodium sulfate by sodium hydroxide. In the Hanford Site wastes, it is expected that greater than 90 percent of the total sulfate will be solubilized by water or metathesized to the soluble sodium sulfate by sodium hydroxide leaching. This results in greater than 90 percent of the tank waste sulfate inventory being routed to the LAW and less than 10 percent to the HLW.

5.21.1 Identification of Sulfate Inventory Values

The reported global inventory values for sulfate are 3,220 MT (HDW model, Agnew et al. 1996), 2,040 MT (TWRS, Shelton 1996), and 5,000 MT by this report.

The methodology and analysis to obtain a total global sulfate inventory of 5,000 MT sulfate is contained in the following sections.

5.21.2 Sulfate Inventory Evaluation

The Hanford Site tank sulfate inventory is estimated by computing the total amount of sulfate added to each of the separations processes and subtracting the amounts of sulfate in the wastes that were discharged to cribs and trenches. Crib transfers are used to estimate the average sulfate concentration and total volume of residual bismuth phosphate and UR process wastes that were left in the tanks (Waite 1991, Anderson 1990). Other sources of information include the following:

1. Process flowsheets (Schneider 1951, GE 1951b, Crawley 1960, Isaacson 1965, Boldt 1966, Schofield 1991).
2. Tank transaction records and estimates from the HDW model (Agnew et al. 1995a and Agnew et al. 1996, respectively)
3. Partial chemical usage summaries (McDonald 1968, McDonald and Hogan 1977, and Hogan 1992).
4. Tank samples and tank inventory estimates derived from supernatant and core samples (Hodgson 1995, Ryan 1995).

These sources are used to develop sulfate inventory estimates for each of the chemical separations processes.

5.21.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process operated at T Plant from December 1944 into 1956 and at B Plant from April 1945 into 1952. The Hanford Site production records indicate that 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the bismuth phosphate processing is developed in Appendix C based on the Schneider (1991) flowsheet. Comparable HDW sulfate values are obtained from the "waste volume out" and "sulfate concentrations" of the HDW. The HDW sulfate concentrations used are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

The sulfate usage in the bismuth phosphate processing is shown in Table 5.21-1 for the Schneider (1991) flowsheet and for the HDW model (Agnew et al. 1996).

Table 5.21-1. Amount of Sulfate in Bismuth Phosphate Process Wastes, MT.

Waste stream	Sulfate, MT	
	Present estimate (Table C-3, Appendix C)	HDW model (Agnew et al. 1996)
Metal waste (MW)	3,030	2,760
First decontamination cycle and coating waste (1C/CW)	700	662
Second decontamination cycle (2C)	635	384
224 waste	15	5
Total	4,380	3,810

Metal Waste. After decladding, the bismuth phosphate process dissolved the uranium metal slugs and extracted the plutonium by carrier precipitation with bismuth phosphate. Sulfuric acid was added to the solution to complex the uranyl ion (UO_2^{+2}) and avoid uranyl phosphate precipitation. This resulted in the metal waste stream. The metal waste stream contained the bulk of the uranium and approximately 90 percent of the long-lived fission products (e.g. ^{137}Cs and ^{90}Sr). The metal waste was neutralized with sodium hydroxide and sodium carbonate and was sent to the SSTs for storage.

The stored metal waste containing the uranium from bismuth phosphate processing was later retrieved as the feed for the UR process.

Cribbed Bismuth Phosphate Wastes. Portions of the other bismuth phosphate waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The estimated total sulfate discharged to cribs and specific retention trenches was 460 MT sulfate in 259,000 m³ (68.5 Mgal) (Waite 1991).

The HDW model estimates 690 MT sulfate was cribbed from the bismuth phosphate process in 173,000 m³ (45.8 Mgal) (Agnew et al. 1996). The HDW estimate for bismuth phosphate wastes is obtained from the crib volume and the supernatant sulfate concentration of Agnew et al. (1996). The sulfate concentrations are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

Summary for Bismuth Phosphate Process Wastes. The residual MW, 1C, and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.21-2. This report estimates the residual bismuth phosphate tank wastes contain 890 MT of sulfate versus the 370 MT estimated by the HDW model.

Table 5.21-2. Estimated Amount of Sulfate in Bismuth Phosphate Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
Bismuth Phosphate wastes produced	4,380	3,810
Bismuth Phosphate wastes cribbed	460	690
Bismuth Phosphate wastes sent to uranium recovery	3,000*	2,750
Bismuth Phosphate wastes remaining in single-shell tanks/double-shell tanks	920	370

HDW = Hanford defined waste

*Assumes 99 percent.

The principal differences in the present estimate and the HDW model are the input chemicals in the flowsheets assumed and the volume of cribbed bismuth phosphate first-cycle and second-cycle wastes.

5.21.2.2 Uranium Recovery Wastes. The UR process began in 1952 to extract uranium from the bismuth phosphate process MW stored in B, BX, BY, C, T, TX, and U tank farms.

Uranium Recovery Chemicals Added. The design basis flowsheet for the uranium recovery process is provided in the *Uranium Recovery Technical Manual* (GE 1951a). The flowsheet value for sulfate addition via ferrous sulfamate and sulfamic acid is 1,120 MT sulfate for 7,500 MTU recovered. This 1,120 MT sulfate does not include potential use of sulfuric acid or sodium sulfate to treat the UR solvent. The flowsheet lists sulfuric acid or sodium sulfate as alternate solvent treatment chemicals to the phosphoric acid assumed in this analysis (Section 5.17.2.2). The potential exists that the UR solvent treatment ultimately used sodium carbonate.

The UR process operated with a highly variable feed due to variations in the quality of retrieved metal waste sludges and supernatants. As a result, the UR flowsheet (GE 1951a) is not expected to provide an accurate estimate of UR sulfamic acid, ferrous sulfamate, and sulfuric acid use. This is still the best information available.

Cribbed Uranium Recovery Wastes. The HDW model analysis of transfer records from the UR operation indicates that 62.5 vol% of the total UR waste streams were cribbed and 37.5 vol% of the total UR waste streams remained in the SSTs. Waite (1991) states that 155,000 m³ (41 Mgal) of scavenged UR wastes containing 2,150 MT of sulfate was discharged to cribs and specific retention trenches. The 2,150 MT of cribbed sulfate represents 52 percent of the projected sulfate content in the UR wastes.

Summary for Uranium Recovery Process Wastes. Table 5.21-3 provides a summary sulfate material balance for the UR process for this analysis and the HDW model.

Table 5.21-3. Estimated Amount of Sulfate in Uranium Recovery Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
Bismuth Phosphate wastes sent to Uranium Recovery	3,000	2,750
Uranium Recovery chemicals added	1,120	640
Uranium Recovery wastes produced	4,120	3,390
Uranium Recovery wastes cribbed	2,150	2,120
Uranium Recovery wastes remaining in single-shell tank/ double-shell tanks	2,000	1,270

5.21.2.3 REDOX Process Wastes. According to the site production records, 19,706 MTU were processed through the REDOX process from 1952 through 1966 (Appendix B). The REDOX process flowsheets can be used to estimate the total amount of sulfate produced during this production (Merrill and Stevenson 1955, Crawley 1960, Isaacson 1965, and Boldt 1966). Based on these flowsheets, it appears that 138.4 MT of sulfate was produced in the REDOX process from consumption of ferrous sulfamate and sulfamic acid.

Chemical purchase records can also be used to generate a separate estimate of net sulfate consumption (McDonald 1968). According to these records, 30,000 kg (66,000 lb) of Fe(NH₂SO₃)₂ were consumed in the REDOX Plant from 1965 through 1966. This is equivalent to 23.2 MT of sulfate produced from oxidation of the sulfamate. The site production records indicate 2,604 MTU were processed through the REDOX Plant from 1965 to 1966 (Appendix B). This results in a recorded production of 8.9 kg sulfate in REDOX process wastes per MTU processed.

From 1952 to 1966, a total of 19,706 MTU were processed through the REDOX Plant (Appendix B). Based on these figures and the assumption of 8.9 kg sulfate/MTU, it is calculated that 175 MT of sulfate was produced by the REDOX process from ferrous sulfamate and sulfamic acid. This sulfate content is 126 percent of the value predicted by the REDOX process flowsheets.

Because the REDOX Plant product streams were frequently reworked, the plant experienced frequent startups and shutdowns (five day a week operation for flowsheets 4 and 5), and specialty processing campaigns for neptunium and nonstandard fuel (performed during the periods represented by Flowsheets 7, 8, and 9), the REDOX process flowsheets can not be used to accurately account for all of the chemicals added to the process. The flowsheet provides chemical usage at equilibrium operation. The nonstandard operations result in chemical use higher than predicted by the flowsheets. Therefore, the reported production of sulfate being 126 percent of the flowsheet values is consistent (Section 4.3.1).

The HDW model (Agnew et al. 1996) calculates a total of 311 MT SO₄ in the neutralized REDOX high level wastes (R1 and R2) and REDOX cladding wastes (CWR1 and CWR2).

5.21.2.4 PUREX Process Wastes. The PUREX process came on line in January 1956. The PUREX Plant processing can be broken down into three main periods: 1983 through 1989, 1965 through 1982, and 1956 through 1964.

1983 through 1989. The period from 1983 through 1989 processed Zircaloy®-clad fuel exclusively. The NCAW during this period was effectively segregated in two DSTs and the sulfate content can be determined by sample analysis.

The total sulfate use by the PUREX Plant in the 1983 through 1989 period is summarized in Table 5.21-4 for this analysis and the HDW model.

Table 5.21-4. Estimated Amount of Sulfate in 1983 Through 1989 PUREX Process Neutralized Current Acid Waste, MT.

Tank	Present estimate	HDW model (Agnew et al. 1996)	
		P3 waste	PL2 waste
241-AZ-101	62 ^a	33	-
241-AZ-102	50 ^a	19	-
Other double-shell tanks	-	3	17
Total	134 ^b	72	

HDW = Hanford Defined Waste

PUREX = Plutonium-uranium extraction

^aShelton (1996), (from tank waste sample analyses).

^bJ. J. Hogan (personal communication), (from essential material records).

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Essential material usage records can be used to define the chemical consumption for the PUREX Plant from 1983 to 1989 (Hogan 1992, and personal communication). J. J. Hogan, per personal communication, also stated that 103 MT (227,800 lb) of ferrous sulfamate and 54.5 MT (120,100 lb) of sulfamic acid was used at the PUREX Plant from 1983 through 1991. This material would result in 134 MT of sulfate in the PUREX Plant tank wastes.

The basis for the PUREX process 1983 through 1989 wastes in Agnew et al. (1996) is not defined. The quantities and distribution of sulfate in the P3 and PL2 wastes are defined by volume, concentration, and distribution of P3 and PL2 wastes in the HDW model (Agnew et al. 1996).

The NCAW sulfate content and potential sulfate distribution in the waste tanks is also determined by samples (Hodgson 1995, Ryan 1995) and summarized in Shelton (1996). The NCAW produced from 1983 through 1989 contains $62 + 50 = 112$ MT total sulfate in the combined sludge and supernatant (Table 5.21-4).

1965 through 1982. The site production records indicate 25,179 MTU and 629 MT of thoria were processed in the PUREX Plant from 1965 through 1972 (Appendix B). The PUREX Plant was maintained in standby from 1973 through 1982. Monthly essential material usage records can be used to define the chemical consumption for the PUREX Plant from 1965 through 1982 (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.21-5.

Table 5.21-5 shows that the total sulfate produced from essential material use by PUREX Plant processing from 1965 through 1982 is 655 MT sulfate. The sodium sulfate use in 1965 through 1967 was for fission product recovery by sulfate strike precipitation.

Table 5.21-5. Sulfate Use in PUREX Process From 1965 Through 1982.

Year	MT processed			Chemical use, MT		
	Aluminum-clad Uranium	Zirconium-clad Uranium	ThO ₂	Fe(NH ₂ SO ₃) ₂	NH ₂ SO ₃ H	Na ₂ SO ₄
1965 ^a	5,385		5	90.7	16.3	142
1966 ^a	3,806		197	80.8	12	119
1967 ^a	4,459	123		69	17.4	23
1968 ^b	3,393	427		84.6	17.9	
1969 ^b	2,568	228		63.2	12.8	
1970 ^b	555	92	427	33.6	5.1	
1971 ^b	2,988	287		32.7	10.7	
1972 ^b	552	416		17.9	6.2	
1973-1982	0	0	0	0	0	0
Total	23,706	1,572	629	472	98	284
Subtotal SO ₄ , MT	-	-	-	366	97	192
Total SO ₄ , MT	655 ^c					

PUREX = Plutonium-uranium extraction

^aMcDonald (1968)

^bMcDonald and Hogan (1977)

^cTotal PUREX process sulfate from 1965 through 1982.

1956 through 1964. The third period covers the PUREX production process from 1956 through 1964. During this period, 42,320 MTU were processed in the PUREX Plant (Appendix B). From 1956 through 1964, only aluminum-clad natural uranium fuel was processed through the PUREX Plant.

Examination of the PUREX Plant processing history in Table 5.21-5 shows that 1965 and 1967 processed predominately aluminum-clad fuel. The thoria campaign in 1966 and zirconium clad fuel after 1967 used a disproportionately high amount of ferrous sulfamate. The average chemical usage in 1965 and 1967 was 16 kg of ferrous sulfamate/MTU and 3.4 kg of sulfamic acid/MTU.

Agnew (1995) identified a sulfate strike operation in the PUREX Plant during 1960 and 1961 based on examination of PUREX Plant monthly reports. This sulfate strike operation is similar to the operation performed in 1965 through 1967 that used 284 MT of sodium

sulfate. An allowance of 150 MT of sodium sulfate is assumed for the sulfate strike operation in 1960 and 1961.

The resulting estimated chemical use in PUREX Plant from 1956 through 1964 is shown in Table 5.21-6. The total PUREX process sulfate generation from 1956 through 1964 is estimated at 770 MT sulfate.

Table 5.21-6. Sulfate Use in PUREX Process From 1956 Through 1964.

Year	MTU processed	Estimated chemical use, MT		
		Fe(NH ₂ SO ₃) ₂ , @ 16 kg/MTU ^a	NH ₂ SO ₃ H, @ 3.4 kg/MTU ^a	Na ₂ SO ₄ , allowance
1956	2,286	36.6	7.8	
1957	3,989	63.8	13.7	
1958	4,707	75.3	16.1	
1959	4,635	74.2	15.9	
1960	5,599	89.6	19.2	75
1961	5,438	87	18.7	75
1962	5,483	87.7	18.8	
1963	5,024	80.4	17.2	
1964	5,157	82.5	17.7	
Total	42,318	677	145	150
Subtotal SO ₄ , MT	-	524	144	100
Total SO ₄ , MT	770 ^a			

MTU = Metric ton uranium

PUREX = Plutonium-Uranium Extraction

^aTotal estimated PUREX process sulfate from 1956 through 1964.

Summary for PUREX Process Wastes. Sulfate use by the PUREX Plant is summarized in Table 5.21-7. This analysis gives a total PUREX Plant sulfate generation of 1,560 MT sulfate based on the following:

1. Extrapolation of reported 1965 and 1967 usage for years 1956 through 1964 (770 MT sulfate, 50 percent of total).
2. Reported 1965 through 1982 usage (655 MT sulfate, 41 percent of total).
3. Reported 1983 through 1989 usage (134 MT sulfate, 9 percent of total).

Sulfate use by the PUREX Plant for comparable periods in the HDW model (Agnew et al. 1996) was not determined in this report. The HDW model PUREX Plant sulfate use was combined with B Plant fission product recovery wastes for comparison. This comparison is shown in Section 5.21-3.

Table 5.21-7. Estimated Amount of Sulfate in PUREX Process Wastes, MT.

Years	Estimated Sulfate Content from Chemical Use, MT SO ₄			
	Fe(NH ₂ SO ₃) ₂	NH ₂ SO ₃ H	Na ₂ SO ₄	Total
1956 - 1964	524	144	100	770
1965 - 1982	366	97	192	655
Subtotal, 1956 - 1982	890	241	292	1,425
1983 - 1989	80	54	-	134
Total, 1956 - 1989	1,560 ^a			

^aTotal estimated PUREX process sulfate from 1956 through 1989.

5.21.2.5 B Plant Process Wastes. Monthly essential material usage records can be used to define the chemical consumption for B Plant and Tank Farms from 1965 through 1982 (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.21-8. The sodium bisulfate and sodium sulfate used from 1968 through 1979 was for strontium product purification before encapsulation. B Plant also purified strontium in 1983-1984 and used an estimated 45 MT sodium sulfate (personal communication, D. E. Place).

Table 5.21-8 shows that the total estimated sulfate use by B Plant and Tank Farms processing from 1965 through 1982 is 152 MT sulfate.

Table 5.21-8. Sulfate Chemical Use in B Plant from 1965 to 1984. (2 Sheets)

Year	Estimated Sulfate Content from Chemical Use, MT SO ₄	
	NaHSO ₄	Na ₂ SO ₄
1965 - 1967 ^a		
1968 ^b	7	
1969 ^b	41	
1970 ^b	24	
1971 ^b	2	33
1972 ^b		39

Table 5.21-8. Sulfate Chemical Use in B Plant from 1965 to 1984. (2 Sheets)

Year	Estimated Sulfate Content from Chemical Use, MT SO ₄	
	NaHSO ₄	Na ₂ SO ₄
1973 ^b		29
1974 ^b		13
1975 ^b		5
1976 ^b		8
1977 ^b		1
1978 ^c		6
1979 ^c		1
1980 - 1982 ^c		
1983 - 1984 ^d		45
Total	75	182
Subtotal SO ₄ , MT	60	122
Total SO ₄ , MT	182 ^e	

^aMcDonald (1968)

^bMcDonald and Hogan (1977)

^cHogan (1992)

^dPersonal communication, D. E. Place.

^eTotal B Plant sulfate use from 1965 through 1982.

5.21.2.6 Other Cribbed Wastes. Agnew et al. (1996) defines portions of waste streams other than the bismuth phosphate and UR process wastes discussed above as having been cribbed. These streams are identified in Agnew et al. (1996) as P1, CWP1, OWW1, DW, T1-SltCk, and WTR. The fraction of the streams cribbed is from Agnew et al. 1996, pages D-49 and D-53. The total sulfate in the streams is given by the volume and the concentration. The summed products of these three values give a total calculated amount of sulfate cribbed of 32 MT sulfate.

This report's analysis is based on the reported cribbed sulfate given by Waite (1991). The cribbed sulfate content in bismuth phosphate and UR wastes given in the previous sections includes all sulfate cribbed in the common crib and specific retention sites. Therefore, any other cribbed wastes identified in Agnew et al. (1996) are included in the bismuth phosphate or UR process values identified by Waite (1991) and no separate value is identified by this report's analysis.

5.21.2.7 N Reactor Decontamination Wastes. Agnew et al. (1996) defined the residual amount of N Reactor decontamination wastes in the DSTs after a single vault of phosphate/sulfate waste grout was produced. The N Reactor decontamination waste stream, (N) is defined in Agnew et al. (1996), pages B-4, B-20, and D-53, with compositions attributed to Lucas (1989). The resulting Hanford Site residual N Reactor decontamination sulfate inventory is 2 MT sulfate.

5.21.3 Best-Basis Estimate for Sulfate Inventory Value

Table 5.21-9 provides the results from this evaluation for sulfate, together with comparable estimates from other sources. The best-basis global inventory for Hanford Site tank waste is 5,000 MT of sulfate. This estimate is significantly higher than both the HDW model estimate of 3,220 MT sulfate (Agnew et al. 1996) and the TWRS inventory value of 2,040 MT sulfate (Shelton 1996).

Table 5.21-9. Estimated Amount of Sulfate in Hanford Site Tank Wastes, MT.

Process Wastes	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
Bismuth Phosphate	920	370	NA
UR wastes	2,000	1,270	NA
REDOX	170	311	NA
PUREX (1956-82) and B Plant (1956-84)	1,740	1,230	NA
Other cribbed wastes	-	(32)	NA
N Reactor decontamination	2	2	NA
PUREX (1983-89)	134	72	112
Total	5,000	3,220	2,040

DST = Double-shell tank
 HDW = Hanford defined waste
 NA = Not attributed to individual waste streams
 PUREX = Plutonium-uranium extraction
 REDOX = Reduction and oxidation
 SST = Single-shell tank
 TWRS = Tank Waste Remediation System
 UR = Uranium recovery.

The present estimate is based on reported chemical consumption for REDOX, PUREX, and B Plants, reported sulfate crib discharges for bismuth phosphate and UR processes, and flowsheet values of chemical consumption for the bismuth phosphate process. The HDW model is based on flowsheet waste compositions and waste transfer records from the Hanford

Site processing plants. The TWRS inventory is based on the 1987 EIS estimate (DOE 1987) of Hanford Site tank inventory in 1983 plus additions to the tank system after 1983.

The process wastes of PUREX and B Plant wastes from 1956 through 1982 and 1984 are combined in Table 5.21-9 for comparison purposes. The individual process contributions in the HDW model (Agnew et al. 1996) are not cited in this report. The 1956 through 1983 value of 1,230 MT sulfate is determined by the difference of the total sulfate (3,220 MT sulfate [Agnew et al. 1996]) and the sum of the individual streams discussed in previous sections (1,990 MT sulfate).

The current Hanford Site tank waste inventory estimate of 5,000 MT sulfate has the highest uncertainty in the residual wastes produced by the bismuth phosphate and UR processes. These wastes represent 60 percent of the estimated total. Both the current estimate and the HDW model are based on flowsheets for additions and estimated crib discharges for the bismuth phosphate and UR processes. The remaining 40 percent of the estimated sulfate inventory is based on reported use of sulfate precursor chemicals resulting in significantly lower uncertainty.

Tank sampling and analysis of sludges will not fully resolve the uncertainty because the alkaline soluble sodium sulfate in the supernatants has resulted in a fairly wide distribution of residual sulfate in tank supernatants and salt cakes.

5.22 STRONTIUM

Other than the "in-plant" scavenging operation at U Plant, no Hanford Site process is known to have used strontium as a flowsheet material. Strontium was preferred to calcium as a supplemental scavenging agent but was not always available. The bulk of the strontium inventory is expected to be present in four SSTs that received UR waste treated by "in-plant" scavenging. The localization of high strontium concentrations in these four tanks is noteworthy.

Radioactive ^{90}Sr is, of course, widely distributed in the tanks but at low concentration. An exception to this is 241-C-106, the "high-heat" tank. Past operations resulted in an unusual concentration of ^{90}Sr in this tank.

Like calcium, strontium is frequently present as an impurity in many commercial chemicals but strontium is normally a trace component relative to calcium.

The extremely low solubility of compounds like SrCO_3 and especially $\text{Sr}_3(\text{PO}_4)_2$ limits the removal of strontium from sludges by proposed sludge washing methods.

5.22.1 Identification and Comparison of Strontium Inventory Values

The current revision of the HDW model (Agnew et al. 1996) distributes 167 MT of strontium to SSTs and only 0.03 MT (30 kgs) to DSTs, with about 1 MT lost to the cribs. By comparison, the TWRS inventory (Shelton 1996) accounts for 36 MT in SSTs and about 0.6 MT (600 kgs) in DSTs.

Table 5.22-1. Current Reported Values for Strontium.

	All tanks (MT)	Single-shell tanks (MT)	Double-shell tanks (MT)
HDW model (Agnew et al. 1996)	167	167	0.03
Tank Waste Remediation System Inventory (Shelton 1996)	37	36	0.6

HDW = Hanford Defined Waste.

5.22.2 Strontium Inventory Evaluation

The addition of strontium to UR waste scavenged ^{90}Sr by co-precipitation with $\text{Sr}_3(\text{PO}_4)_2$. The table of "chemicals added" in Appendix B of the HDW model document (Agnew et al. 1996) assigns the source of strontium to the 224 waste stream. The HDW model bears out in its tank-by-tank results (Appendix E) that virtually all of the strontium is routed to SSTs containing 224 waste. This apparently is an error in defining stream compositions because the document referenced by Agnew et al. for the composition (Lucas 1989) does not show strontium in 224 waste, and furthermore, the HDW "in-plant" scavenged stream shows no strontium, which is where it would be expected.

5.22.2.1 Fuel Fabrication. The strontium content of fuel is not known, but it is reasonable to assume that fuel made no significant contribution to the inventory.

5.22.2.2 Fission Product Strontium. Four primary isotopes of fission product strontium were produced in Hanford's reactors. Table 5.22-2 shows that radioactive ^{90}Sr and the stable ^{88}Sr persist; the short-lived ^{89}Sr and ^{91}Sr were essentially gone within two years of discharge from the reactor. The Hanford Site produced approximately 100 MCi ^{90}Sr . About 95.4 percent of the ^{90}Sr remained after two years of decay, and at that point in time, accounted for 60 wt% of the fission product strontium. The ^{90}Sr continued to decay over the years but ^{88}Sr remained constant. Multiple ORIGEN2 code runs which simulate the time integrated generation and decay of radionuclides in Hanford reactor fuels indicate that as of January 1, 1994 (the decay date basis used in Section 6), ^{90}Sr would account for only 43 wt% of total fission product strontium. This document estimates that as of January 1, 1994, approximately 72 MCi ^{90}Sr reside in the tanks, another 26 MCi are encapsulated, and roughly 7 MCi are distributed to offsite facilities, plant residuals, and solid waste. There is, therefore, 0.52 MT of ^{90}Sr in the tanks. Assuming that isotopic ratios for

strontium remaining in tanks are the same as for fuels before chemical separations, the total fission product strontium in tanks is estimated to be 1.2 MT ($0.52/0.43 = 1.2$).

Table 5.22-2. Fission Product Strontium in 9 Percent Mark IV Fuel (Hedengren 1985).

Sr isotope	Grams of Sr per MT of fuel	
	At discharge	After 2 years
⁸⁸ Sr	22.57	22.59
⁸⁹ Sr	10.30	0.00046
⁹⁰ Sr	35.56	33.91
⁹¹ Sr	0.12	nil
Total Fission Sr	68.55	56.50

5.22.2.3 Chemical Process Operations. Chemical processing introduced strontium to the waste through intentional routine additions. Initially, UR wastes were scavenged only by ferrocyanide to remove ¹³⁷Cs. Calcium was introduced later as a supplemental scavenging agent to remove ⁹⁰Sr, eventually to be replaced by scavenging with nonradioactive strontium. Four SSTs (241-BY-106, 107, 108, and 110) received 57 batches of "in-plant" scavenged waste. An interim report on the scavenging process, covering the filling of the first 30 tanks, shows that twelve were treated with strontium (Abrams 1956). The total strontium usage for those twelve tanks was 9.28 MT. Assuming that the remainder of the "in-plant" batches was scavenged at the same flowsheet conditions (0.004M Sr), total strontium consumption would be 29.14 MT. This compares favorably with the reported usage of 140,000 lb of Sr(NO₃)₂ (Stedwell 1957), which is equivalent to 26.70 MT of strontium. Stedwell's report was issued four months prior to the last "in-plant" batch so it may not account for all the strontium.

As a general observation, analytical results for Tanks 241-BY-108 and 241-BY-110 confirm a high concentration of strontium in the waste compared to other tanks.

The operating procedure for "in-farm" scavenging (Sloat 1955) indicates that scavenging with strontium was tested in the laboratory but the processing records show that strontium was never used (Jeppson 1993). "In-farm" scavenging was implemented for 200 East Area waste, but not in 200 West Area. The only known use of strontium, therefore, was for "in-plant" scavenging.

Klem (1990) indicates that Sr(NO₃)₂ was used in the 300 Area laboratories, but the quantities were most certainly small. There is also some question about the final destination of lab wastes.

5.22.2.4 Process Chemical Impurities. It is reasonable to assume that whenever calcium is present as an impurity, strontium would also be present at about 1/10 the concentration. Based on the discussion of calcium impurity in NaOH (see Section 5.3.2.3), one would project less than 1 MT of strontium from commercial grade caustic. This amount appears to be consistent with the isotopic dilution that occurred between the strontium in two year old fuel (60 wt% ^{90}Sr) and the strontium recovered in B Plant operations (49 wt% ^{90}Sr).

5.22.2.5 Summary of Strontium Evaluation. The sources evaluated in Sections 5.22.2.1 through 5.22.2.4 account for 31.3 MT of strontium. This is in reasonable agreement with the TWRS Inventory value of 37 MT, whereas the HDW model estimate of 167 MT was based on an erroneous defined waste composition as noted above.

5.22.3 Best-Basis Estimate for Strontium Inventory

For the best-basis estimate it is recommend that 31.3 MT of strontium be used. Also, it is important to note that virtually all of the strontium probably resides in four tanks (241-BY-106, 107, 108, and 110) that received "in-plant" scavenging waste. Fission product strontium, a small fraction of the total, is widely distributed.

5.23 TOTAL ORGANIC CARBON

The following were the primary sources of total organic carbon (TOC) in tank wastes:

1. Solvent losses from the UR, PUREX, and B Plant solvent extraction processes.
2. Equipment flushing reagents used at the PUREX Plant.
3. Fission product recovery using organic complexants.
4. Decontamination solutions.

TOC exists as several anions. Multivalent metal salts of oxalate are generally insoluble; monovalent salts are sparingly soluble. Metal salts of the organic complexants are generally soluble. The solvent losses from UR, PUREX, and B Plant solvent extraction processes can form a separate immiscible organic phase if the organic exceeds the solubility limit in a waste supernatant aqueous phase. In the Hanford wastes, greater than 90 percent of the TOC is expected to be water soluble upon retrieval.

5.23.1 Identification of Total Organic Carbon Inventory Values

The reported global TOC inventory values are 1,805 MT (HDW model, Agnew et al. 1996), 1,142 MT (TWRS, Shelton 1996), and 4,000 MT by this report.

The methodology and analysis to obtain a total global TOC inventory of 4,000 MT TOC is contained in Section 5.23.2.

5.23.2 Total Organic Carbon Inventory Evaluation

The Hanford Site tank TOC inventory is estimated by computing the total amount of TOC added to each of the separations processes and subtracting the amounts of TOC in the wastes that were discharged to cribs and trenches. Crib transfers are used to estimate the average TOC concentration and total volume of BiPO₄ and UR process wastes that were left in the tanks (Waite 1991, Anderson 1990). Other sources of useful information include the following:

1. Process flowsheets (Schneider 1951 and GE 1951b).
2. Tank transaction records and estimates from the HDW model (Agnew et al. 1995a and Agnew et al. 1996, respectively).
3. Partial chemical usage summaries (McDonald 1968, McDonald and Hogan 1977, and Hogan 1992).

These sources are used to develop TOC inventory estimates for each of the chemical separations processes.

The Hanford Site tank TOC inventory is estimated by computing the total amount of TOC added to each of the separations processes. The primary process sources of TOC were from the PUREX and fission product recovery solvent extraction processes. The BiPO₄ recovery process limited organic use to oxalate precipitation. The UR process and REDOX process also used organic solvents.

Organic discharges to process condensate cribs were assumed to be 80 percent of the solvent extraction diluent losses to tank farms from the PUREX and B Plant fission product recovery operations. The actual evaporation of diluent may approach 100 percent. The resulting 20 percent residual diluent in tank wastes results in a conservative basis to calculate TOC.

5.23.2.1 Bismuth Phosphate Process Wastes. The bismuth phosphate process started up at T Plant in December 1944, and at B Plant in April 1945, and continued until 1952 in B Plant and until 1956 in T Plant. The site production records indicate that 7,800 MTU were processed in the bismuth phosphate plants from 1944 to 1956 (Appendix B).

An estimate of chemical usage in the BiPO₄ processing is developed in Appendix C based on the flowsheet by Schneider (1951). Comparable HDW TOC values are obtained from the "waste volume out" and "TOC concentrations" of the HDW. The HDW TOC concentrations used are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

The TOC usage in the BiPO₄ processing is shown in Table 5.23-1 for the Schneider (1951) flowsheet (Appendix C) and for the HDW model (Agnew et al. 1996).

Table 5.23-1. Amount of Total Organic Carbon in BiPO₄ Process Wastes, MT.

Waste stream	Present estimate	HDW model (Agnew et al. 1996)
Metal waste (MW)	0	0
First decontamination cycle and coating waste (1C/CW)	0	0
Second decontamination cycle (2C)	0	0
224 Waste		
as oxalate ion, MT	461	83
as Total Organic Carbon, MT	126	23
Total Organic Carbon, MT	126	23

HDW = Hanford Defined Waste.

Cribbed BiPO₄ Wastes. Portions of BiPO₄ waste streams were disposed to cribs and specific retention trenches following settling of solids in a cascaded series of SSTs (Waite 1991). The solubility of oxalate in cribbed supernatants is assumed to be zero. The resulting total TOC discharged to cribs and specific retention trenches is 0 MT TOC in 259,000 m³ (68.5 Mgal) (Waite 1991).

The waste from the 244-T and 244-B Buildings was originally routed to tanks 241-T-361 and 241-B-361, respectively, and in turn overflowed to dry wells or cribs. Oxalate would have been present as an insoluble compound, and most should have settled out in the tanks. Tank 241-T-361 was active from 1944 to 1947, and tank 241-B-361 was active from 1945 to 1947 (Freeman-Pollard 1994). These concrete tanks are not part of the DST or SST systems. An estimate based on the MTU processed during those years indicate that up to 25 percent of the oxalate or TOC (31 of the 126 MT TOC) could remain in these two tanks or the associated dry wells/cribs. The remaining 95 MT TOC would be in the SSTs and DSTs.

The HDW model estimates 0.7 MT TOC were cribbed from the BiPO₄ 224 wastes in 5,530 m³ (1.46 Mgal) (Agnew et al. 1996). The HDW estimate for BiPO₄ wastes is obtained from the crib volume and the supernatant TOC concentration of Agnew et al. (1996). The TOC concentrations are attributed to Jungfleisch (1980, 1983, 1984) and Lucas (1989).

Summary for BiPO₄ Process Wastes. The residual MW, 1C and 224 wastes in the tanks determined by this estimate and the HDW model are presented in Table 5.23-2. This report estimates the residual BiPO₄ tank wastes contain 126 MT of TOC versus the 22 MT estimated by the HDW model.

Table 5.23-2. Estimated Amount of Total Organic Carbon in BiPO₄ Tank Wastes, MT.

BiPO ₄ waste stream	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes produced	126	23
BiPO ₄ wastes cribbed	0	0.7
BiPO ₄ wastes sent to uranium recovery	0	0
BiPO ₄ wastes in 241-B-361 and 241-T-361 tanks	31	0
BiPO ₄ wastes in single-shell and double-shell tanks	95	22

HDW = Hanford Defined Waste.

The principal differences in the present estimate and the HDW model for BiPO₄ wastes are the volume of 224 wastes, 114.3 ML and 31.4 ML, and concentration, respectively (Appendix C and Agnew et al. 1996).

5.23.2.2 Uranium Recovery Process Wastes. The UR process began in 1952 to extract uranium from the BiPO₄ process MW stored in B, BX, BY, C, T, TX, and U tank farms.

Uranium Recovery Chemicals Added. The design basis flowsheet for the UR process is provided in the *Uranium Recovery Technical Manual* (GE 1951a). TOC use in the UR process was limited to solvent losses to the tank wastes. The design basis flowsheet does not give a basis for solvent losses.

The PUREX process losses to tanked wastes were evaluated by Sederbug and Reddick (1994) as 747 MT TBP and 1,300 MT diluent during the PUREX life while processing 71,400 MTU. With the assumption of 20 percent residual diluent in the tank waste after evaporation, this results in 8.7 kg TOC in tanked wastes per MTU. This analysis uses the PUREX value of 8.7 kg TOC/MTU for the 7,500 MTU processed at UR for a total of 65 MT TOC.

For the HDW model the amount of TOC added during UR is the difference in the TOC content between the UR wastes produced and the BiPO₄ wastes sent to UR. The HDW model TOC values in the UR wastes produced (7.1 MT TBP containing 3.8 MT TOC) are obtained from the "waste volume out" and "TOC concentrations" of the HDW model. Subtraction of the TOC inventory to UR (0 MT TOC) gives a 3.8 MT TOC chemical addition in UR.

Cribbed Uranium Recovery Wastes. The HDW model (Agnew et al. 1996) analysis of transfer records from the UR operation indicates that 157,000 m³ (41.6 Mgal) or 62.5 vol% of the total UR waste streams were cribbed and 37.5 vol% of the total UR waste streams were routed to the SSTs. This analysis assumes 25 MT TOC cribbed in UR wastes and 40 MT TOC in residual UR wastes.

The HDW model UR waste TOC distribution is 62 percent (2.4 MT TOC) to the cribs and 38 percent (1.4 MT TOC) to the SSTs. The HDW model estimate for UR wastes is obtained from the crib volume and the supernatant TOC concentration of Agnew et al. (1996). The TOC concentrations are attributed to Borsheim and Simpson (1991) and Sloat (1954).

Summary for Uranium Recovery Process Wastes. Table 5.23-3 provides a summary TOC material balance for the UR process for this analysis and the HDW model. This report and the HDW model estimate the residual UR tank wastes contain 25 and 1.4 MT of TOC, respectively.

Table 5.23-3. Estimated Amount of Total Organic Carbon in Uranium Recovery Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)
BiPO ₄ wastes sent to UR	0	0
UR chemicals added	65	3.8
UR wastes produced	65	3.8
UR wastes cribbed	40	2.4
UR wastes in single-shell/double-shell tanks	25	1.4

HDW = Hanford Defined Waste
UR = Uranium recovery.

5.23.2.3 REDOX Process Wastes. The REDOX process used hexone as a process solvent. Hexone has a high volatility and no hexone is assumed to have been discharged to the tank farms in process wastes. The other use of organic at REDOX was an oxalic acid regenerant of silica gel columns. The silica gel columns provided a final decontamination of the REDOX uranium product stream. The flowsheets show oxalate use of 2.5 to 2.7 kg/MTU. This use for 19,700 MTU at REDOX results in a calculated 51 MT oxalate or 18 MT TOC.

This is supported by examining the composition of tanks 241-S-104 and 241-SX-108. Tank 241-S-104 contains 2,840 kg of TOC in 408.8 m³ (108 kgal) of R1 (REDOX process high-level) waste and 90.8 m³ (24 kgal) of CWR1 waste (REDOX process cladding waste)

(Agnew et al. 1995b). Tank 241-SX-108 contains about 500 kg of TOC in 333.1 m³ (88 kgal) of R1 and R2 (REDOX process high-level) waste.

This concentration of TOC could result in several 10s of MT of TOC in the REDOX sludges. Additional potential sources of TOC in REDOX wastes could be degraded solvent in the hexone still bottoms that were routed to the HLW, organics discharged from the 222-S laboratory, process equipment lubricants lost to the process cells, degraded paints and coatings, decontamination agents, and ion exchange resins.

This analysis assumes 30 MT of TOC in the REDOX wastes. The HDW model assumes zero TOC in the REDOX wastes.

5.23.2.4 PUREX Process Wastes. The TOC content in PUREX wastes originates from TBP/diluent solvent losses to the wastes, equipment flushes using oxalic-nitric acids and caustic-tartaric solution, and residual carbon resulting from sugar denitration of HLW prior to (1) neutralization and transfer to the tank farms or (2) transfer of Current Acid Waste (CAW) to B Plant. Table 5.23-4 shows the PUREX process TOC contribution to the tank wastes given by these sources.

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Table 5.23-4. Total Organic Carbon Discharged to Tank Farms by PUREX Process.
(2 Sheets)

Year	MT processed			Chemical use, MT				
	Al clad U	Zr clad U	ThO ₂	TBP ^a (C ₄ H ₉ O) ₃ PO ₄	Diluent ^a C ₁₂ H ₂₆	Oxalic Acid HO ₂ CCO ₂ H	Tartaric Acid HO ₂ C(CHOH) ₂ CO ₂ H	Sucrose C ₁₂ H ₂₂ O ₁₁
1955				6.1	10.2			
1956	2,286			37.8	63.8	16.4 ^b	4.0 ^c	
1957	3,989			51.7	87.1	28.6 ^b	6.9 ^c	
1958	4,707			49.9	84.1	33.7 ^b	8.2 ^c	
1959	4,635			40.1	67.6	33.2 ^b	8.0 ^c	
1960	5,599			47.8	80.5	40.1 ^b	9.7 ^c	
1961	5,438			73.7	124.2	39.0 ^b	9.4 ^c	
1962	5,483			54.1	91.1	39.3 ^b	9.5 ^c	
1963	5,024			34.5	60.0	36.0 ^b	8.7 ^c	90 ^d
1964	5,157			36.7	63.8	36.9 ^b	8.9 ^c	90 ^d
1965 ^a	5,385		5	54.0	99.9	27.0 ^e	6.4 ^e	89 ^e
1966 ^a	3,806		197	49.7	58.3	44.4 ^e	12.2 ^e	105 ^e
1967 ^a	4,359	123		27.1	49.4	34.0 ^e	6.4 ^e	113 ^e
1968 ^b	3,393	427		23.5	25.3	23.6 ^f	5.9 ^f	136 ^f
1969 ^b	2,568	228		18.0	31.7	15.9 ^f	4.0 ^f	89 ^f
1970 ^b	555	92	427	33.9	82.4	25.2 ^f	7.1 ^f	95 ^f
1971 ^b	2,988	287		27.7	53.8	14.2 ^f	3.0 ^f	88 ^f
1972 ^b	552	416		10.6	19.7	7.5 ^f	1.2 ^f	57 ^f
1973 ^b						0.6 ^f		
1974 ^b						5.7 ^f	0.4 ^f	
1975 ^b						1.3 ^f	1.1 ^f	
1976 ^b						3.2 ^f	0.7 ^f	
1977 ^b						0.5 ^f	0.3 ^f	
1978 ^c								
1979 ^c					0.6	0.4 ^g	0.3 ^g	
1980 ^c								
1981 ^c						0.1 ^g		
1982 ^c				1.5		0.6 ^g	3.0 ^g	1 ^g
1983 ^c		89		8.6	26.1	5.7 ^g	1.7 ^g	22 ^g

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Table 5.23-4. Total Organic Carbon Discharged to Tank Farms by PUREX Process.
(2 Sheets)

Year	MT processed			Chemical use, MT				
	Al clad U	Zr clad U	ThO ₂	TBP ^a (C ₄ H ₉ O) ₃ PO ₄	Diluent ^f C ₁₂ H ₂₆	Oxalic Acid HO ₂ CCO ₂ H	Tartaric Acid HO ₂ C(CHOH) ₂ CO ₂ H	Sucrose C ₁₂ H ₂₂ O ₁₁
1984 ^c		1,092		22.4	37.8	0.8 ^g		15 ^g
1985		1,142		13.5	26.4	4 ^h	1 ^h	30 ^h
1986		856		10.1	21.7	4 ^h	1 ^h	30 ^h
1987		125		7.5	18.5	4 ^h	1 ^h	30 ^h
1988		587		6.6	17.1	4 ^h	1 ^h	30 ^h
1989								
1990				0.4	3.7			
1991					0.7			
Total	65,925	5,463	629	747	1,300	530	130	1,100
TOC, MT	-	-	-	404	221 ⁱ	141	42	22 ^j
Total TOC, MT	830 ^k							

PUREX = Plutonium-uranium extraction

TBP = Tri-butyl phosphate

TOC = Total Organic Carbon

^aSederburg and Reddick (1994)

^bAssumes 7.2 kg oxalic acid per MT uranium average in period 1965 through 1969.

^cAssumes 1.7 kg tartaric acid per MT uranium average in period 1965 through 1969.

^dAssumes 18 kg sucrose per MT uranium

^eMcDonald (1968)

^fMcDonald and Hogan (1977)

^gHogan (1992)

^hAllowance of 4 MT oxalic acid, 1 MT tartaric acid, and 30 MT sucrose per year

ⁱAssumes 80 percent evaporation of diluent from tank farm operations

^jAssumes 5 percent residual carbon as oxalate

^kTotal PUREX process tank waste TOC from 1955 through 1991.

Hanford Atomic Products Operations (HAPO)/Chemical Processing Department Monthly Reports and monthly essential material usage records were used to define the annual PUREX solvent losses to tanks, cribs, and the atmosphere for the period 1955 through 1991 (Sederburg and Reddick 1994). Sederburg and Reddick identified 773 m³ of TBP and 1,770 m³ (468 kgal) of diluent reporting to the waste tanks. This analysis assumes subsequent evaporation of 80 percent of the diluent occurred in the tank farms reducing the diluent content to 350 m³ (92.5 kgal). The 773 m³ (204 kgal) of TBP and 350 m³ (92.5 kgal)

of diluent contain the equivalent of 372 and 221 MT TOC, respectively. The HDW model (Agnew et al. 1996) assumes evaporation of 100 percent of diluent originally containing 955 MT TOC. The HDW model basis for TBP losses to the tank wastes is 514 MT TOC.

5.23.2.5 B Plant Process Wastes. Monthly essential material usage records can be used to define the chemical consumption for B Plant and tank farms during the 1965 to 1982 period (McDonald 1968, McDonald and Hogan 1977, Hogan 1992). Data from these essential material use records are summarized by year in Table 5.23-5. The TBP, D2EHPA, and diluent ($C_{12}H_{26}$) use in 1968 through 1979 was used as the solvent for strontium recovery. Solvent losses and the associated TOC reported to the waste tanks. Table 5.23-5 shows that the total estimated TOC use by B Plant and tank farms processing in the period of 1965 through 1981 is 3,050 MT TOC.

Table 5.23-5. Total Organic Carbon Use in B Plant from 1965 through 1981. (3 Sheets)

Year	Chemical Use, MT						
	DTPA-Na $N_5(CH_2)_4(COONa)_5$	NTA acid $N(CH_2COOH)_3$	Hydroxy-acetic acid $HOCH_2COOH$	HEDTA-Na $N_2C_2H_4(C_2H_2O_2Na)_4(C_2H_4OH)$	EDTA-Na $N_2C_2H_4(C_2H_2O_2Na)_4$	Sodium gluconate $(CHOH)_6C-O_2HCH_2ONa$	
1965 ^a	7.8		64				
1966 ^a	7.1		55				
1967 ^a			8				
1968 ^b		9.9	174	82			
1969 ^b			166	88			
1970 ^b		4.5	112	188			
1971 ^b		0.7	150	115	63		
1972 ^b			124	218	148		
1973 ^b			85	539	372		
1974 ^b			109	757	375		
1975 ^b			87	522	343	16.6	
1976 ^b			95	613	363	18.9	
1977 ^b			129	492	323	18.5	
1978 ^c			55	180	118	8.5	
1979 ^c					4	0.1	
1980 ^c							
1981 ^c				2			
Total	15	15	1,412	3,798	2,109	63	
TOC, MT	4	6	446	1,386	758	21	
Subtotal TOC, MT							2,620 ^f

Table 5.23-5. Total Organic Carbon Use in B Plant from 1965 through 1981. (3 Sheets)

Year	Chemical Use, MT						
	Acetic acid CH ₃ CO ₂ H	Citric acid C ₆ H ₈ O ₇ (COOH) ₃	Tartaric acid (CO ₂ H) ₂ (CHOH) ₂	TBP (C ₄ H ₉ O) ₂ PO	D2EHPA (C ₄ H ₉ O) ₂ POOH	Diluent C ₁₂ H ₂₆	
1965 ^a		1			1.7		
1966 ^a	2.3	6			0.6		
1967 ^a							
1968 ^b		175	64.4	6.0	10.0		
1969 ^b		145	28.6	5.5	5.4		
1970 ^b		49	18.7	4.7	8.3		
1971 ^b		100	19.6	3.2	5.5		
1972 ^b		131		7.4	9.7		
1973 ^b		2		3.2	6.6		
1974 ^b		1		2.6	6.3		
1975 ^b		86		2.1	2.3		
1976 ^b		69		5.7	6.3		
1977 ^b		65		1.8	6.8		
1978 ^c		14			4.0		
1979 ^c							
1980 ^c		2					
1981 ^c		3					
Total	2	849	131	43	73	31 ^d	
TOC, MT	1	318	42	23	44	5 ^e	
Subtotal TOC, MT	430 ^f						
Total TOC, MT	3,050						

Table 5.23-5. Total Organic Carbon Use in B Plant from 1965 through 1981. (3 Sheets)

Year	Chemical Use, MT				
	Acetic acid CH ₃ CO ₂ H	Citric acid C ₆ H ₈ O ₇ (COOH) ₃	Tartaric acid (CO ₂ H) ₂ (CHOH) ₂	TBP (C ₄ H ₉ O) ₃ PO	D2EHPA (C ₄ H ₉ O) ₂ POOH

D2EHPA = Di(2-ethylhexyl)phosphoric acid

DTPA = Diethylene-triamine penta acetate

EDTA = Ethylenediamine tetra acetate

HEDTA = N(2-hydroxyethyl)ethylenediamine tetra acetate

NTA = Nitritriacetic

TBP = Tri-butyl phosphate

TOC = Total Organic Carbon

^aMcDonald (1968)

^bMcDonald and Hogan (1977), used at Strontium Semiworks

^cHogan (1992)

^dKlem (1996)

^eAssumes 80 percent evaporation of diluent from tank farm operations

^fSubtotal B Plant TOC to tank farms from 1965 through 1981; total B Plant TOC to tank farms from 1965 through 1981 = 3,050 MT TOC.

5.23.3 Best-Basis Estimate for Total Organic Carbon Inventory

Table 5.23-6 provides the results from this evaluation for TOC, together with comparable estimates from other sources. The global best-basis estimate for Hanford Site tank waste is about 4,000 MT of TOC. This estimate is 200 to 300 percent higher than HDW model estimate of 1,880 MT TOC (Agnew et al. 1996) and the current value of 1,142 MT in the TWRS reference chemical inventory (Shelton 1996). The current TWRS reference inventory is based on 1986 EIS estimates for tank wastes, corrected for known additions to the waste tanks.

Table 5.23-6. Estimated Amount of Total Organic Carbon in the Hanford Site Tank Wastes, MT.

Process	Present estimate	HDW model (Agnew et al. 1996)	TWRS inventory (Shelton 1996)
BiPO ₄ wastes in SST/DSTs	95	22	NA
UR wastes in SST/DSTs	25	1	NA
REDOX	30	0	NA
PUREX	830	514	NA
B Plant	3,050	1,344	NA
Total	4,000	1,880	1,144

- DST = Double-shell tank
- HDW = Hanford defined waste
- NA = Not attributed to individual waste streams
- PUREX = Plutonium-uranium extraction
- REDOX = Reduction oxidation
- SST = Single-shell tank
- TWRS = Tank Waste Remediation System
- UR = Uranium recovery.

The current Hanford Site tank waste inventory estimate of 4,000 MT TOC has an uncertainty in the residual UR, PUREX, and B Plant solvent extraction diluents in the wastes following evaporation of diluents during tank farm operations. The assumed 80 percent volatilization of solvent extraction diluents represents a residual 220 MT TOC or 5 percent of the current estimate. The actual volatilization of diluent from UR, PUREX, and B Plant may approach the 100 percent assumed in the HDW model.

Also, the estimated total of 4,000 MT TOC does not include decontamination agent use by processing facilities, tank farm operations, and for equipment reclamation by T Plant. These decontamination wastes and laboratory wastes are routed to tank farms. The expected total TOC content in decontamination chemicals and laboratory wastes is expected to be less than 100 MT TOC.

The 4,000 MT TOC is conservatively high because no allowance for organic degradation in the tanks was considered. Single carbon atom degradation products would be converted to carbon dioxide, methane, or methanol and be absorbed as inorganic carbonate or vented as volatile carbon monoxide, methane, or methanol. No estimate of the extent of TOC removal by in-tank degradation is available at this time.

5.24 ZIRCONIUM

Zirconium was used in the scavenger precipitation step of the bismuth phosphate process and as cladding for reactor fuels. Three zirconium-clad fuels were processed at the Hanford Site: (1) N Reactor fuel, (2) PRTR fuel, and (3) Shippingport fuel. The zirconium used in the bismuth phosphate process contributed a relatively small fraction to the total zirconium in the waste. Most of the zirconium came from N Reactor fuel.

Zirconium is also a reactor fission product. The amount of zirconium introduced as a fission product is small relative to the other sources of zirconium.

5.24.1 Identification of Zirconium Values

Estimates of zirconium to date have primarily focused on discharge records and process flowsheets of the chemical processes. Table 5.24-1 compares some of the reported values.

Table 5.24-1. Reported Zirconium Inventory Values.

Reference	Inventory (MT)	Comment
TWRS Baseline (Shelton 1996)	687	
HDW model (Agnew et al. 1996)	232	
<i>Hanford Defense Waste Final Environmental Impact Statement</i> (DOE 1987)	277	Pre 1983 inventory only
TRAC (Jungfleisch 1984)	391	Pre 1981 inventory only
Allen (1976)	246	Pre 1973 inventory only

5.24.2 Zirconium Inventory Evaluation

Schneider (1951) states that the bismuth phosphate process used a single cerium-zirconium scavenger precipitation step prior to October 1, 1951. Subsequent to that date, processing of 600 MWd uranium after 40 day cooling led to high radioactivity in the Concentration Building final solution. Double cerium-zirconium scavenger precipitation was initiated to improve product decontamination. Using the fuel process history in Appendix B and the chemical addition rates discussed in Schneider (1951) results in an estimate of 1.9 MT zirconium used by the bismuth phosphate process before October 1, 1951, and 2.4 MT zirconium used after that date.

The reconstructed fuel process history in Appendix B shows zirconium-clad fuel was processed as follows: the REDOX Plant processed approximately 245 MTU between 1963 and 1966, whereas the PUREX Plant processed 1,572 MTU between 1967 and 1972, and 3,893 MTU between 1983 and 1989.

The zirconium-clad fuel processed at the REDOX Plant consisted of 12.4 MTU of Shippingport fuel, 4.5 MTU of PRTR fuel and 228 MTU of N Reactor fuel. Shippingport fuel contained 415 kg zirconium/MTU (Boldt and Ritter 1969). The PRTR fuel had 276 kg zirconium/MTU (Hanson 1965). The 0.947 percent enriched N Reactor fuel is clad with 70.35 kg zirconium/MTU whereas the 1.25-0.947 percent enriched "spike" fuel is clad with 85.65 kg zirconium/MTU (RHO 1980). (Details of the N Reactor fuel fabrication are found in Section 4.2 and in Section 7.0.)

The zirconium-clad fuel processed at the PUREX Plant consisted of 3.9 MTU of PRTR fuel, 5,460 MTU of N Reactor fuel, and 1.1 MTU of Pu-Al alloy fuel. The zirconium in PRTR fuel processed at the PUREX Plant was determined from flowsheets and loading lists (Matheison and Nicholson 1969, and Watrous 1969a, 1969b). The zirconium cladding on the Pu-Al alloy fuel was determined from the process flowsheet and loading list (Matheison and Nicholson 1972a and 1972b). Zirconium in N Reactor fuel was based on 70.35 kg zirconium/MTU of 0.947 percent enriched fuel and 85.65 kg zirconium/MTU of 1.25-0.947 percent spike fuel (RHO 1980). The high enrichment co-product driver elements were based on 87 kg zirconium/MTU (Austin and Nicholson 1967). The zirconium impurity allowance for N Reactor fuel of 65 $\mu\text{g/g}$ was used and is included in the estimates.

Section 7.2 indicates that 9,990 kg of total Zr were introduced into the tanks by fuel fission.

5.24.3 Best-Basis Estimate for Zirconium Inventory Value

Table 5.24-2 summarizes the zirconium sources identified in this section. The evaluation in Section 5.24.2 results in an estimate of 440 MT zirconium. This estimate is based on the processing of 5,710 MT of zirconium-clad fuel at the REDOX and PUREX Plants. It is assumed that all of the zirconium is in the tanks and none of the zirconium went to the cribs.

The HDW model (Agnew et al. 1996) estimates 232 MT of zirconium in the tanks. This estimate assumes that 16 MT of zirconium went to soil by the cribs and leaks. Discharge to zirconium-bearing waste to cribs would have occurred during bismuth phosphate processing, but was not the practice with the REDOX and PUREX processes. The HDW model reports 16.3 and 22.8 MT of zirconium in the bismuth phosphate process 1C1 and 1C2 waste types. These values are more than an order of magnitude larger than the respective values shown in Table 5.24-2. The root cause appears to be an erroneous zirconium concentration in the HDW model. The amount of zirconium that went to cribs should be a fraction of the 4.3 MT of zirconium discharged by the bismuth phosphate process.

Table 5.24-2. Summary of Zirconium Sources.

Zirconium Source	Zirconium Inventory (kg)
Bismuth phosphate process - pre October 1, 1951	1,930
Bismuth phosphate process - post October 1, 1951	2,360
REDOX process, PRTR	1,230
REDOX process, Shippingport	5,160
REDOX process, N Reactor	16,700
PUREX process, PRTR	1,120
PUREX process, Pu-Al Alloy	240
PUREX process 1966-1972, N Reactor	89,470
PUREX process 1966-1972, Coproduct Driver	31,280
PUREX process 1983-1989, N Reactor	283,180
Fission Product Zirconium	9,990
Total Zirconium	442,660

PRTR = Plutonium Recycle Test Reactor
PUREX = Plutonium-uranium extraction
REDOX = Reduction and oxidation.

The HDW model does not calculate zirconium correctly due to inaccuracies in the fuel reprocessing history. The HDW model does not indicate any zirconium-clad fuel being processed in the REDOX Plant, and only 886 MT of zirconium-clad fuel is shown as being processed by the PUREX Plant between 1968 and 1972. These values are well below the quantities of zirconium-clad fuel reported in Appendix B. However, the HDW model does correctly show 3,900 MTU of zirconium-clad fuel being processed between 1983 and 1989.

In addition, the HDW model appears to underestimate the amount of zirconium per unit of fuel. When the zirconium concentration and volumes of the CWZr1 and CWZr2 streams are multiplied to calculate a zirconium mass, and then divided by the mass of fuel, values of 64 and 49 kg Zr/MTU fuel result, respectively. The evaluation in Section 5.24.2, however, identifies 70.35 kg/MTU for 0.947 percent enriched fuel and 85.65 kg/MTU for spike fuel as the proper cladding values. The 70.35 and 85.65 kg Zr/MTU reconcile with the N Reactor fuel description found in the PUREX Technical Manual (Rockwell 1980). The root cause of the low zirconium-to-uranium ratio derived from the HDW model was not determined.

The HDW model does not indicate any zirconium in the P2, P2, and P3 streams. Schofield (1991) concluded that 2 to 6 percent of the zirconium cladding carries over from metathesis into the NCAW as both dissolved and suspended solids in the form of fines.

The TWRS baseline zirconium inventory is compiled from the Hanford Defense Waste Environmental Impact Statement inventory for SSTs, and sample data for DSTs. The Hanford Defined Waste Environmental Impact Statement inventory is considered to be an update of an estimate by Allen (1976). The Allen/Hanford Defined Waste Environmental Impact Statement inventories appear to overestimate the zirconium from the 1966-1972 PUREX Plant processing by 100-130 MT, relative to the independent evaluation. The Allen report uses a Zirflex process cladding waste composition containing 0.25 M zirconium that appears to reconcile with the charging data shown on the flowsheet (ARHCO 1972). That leaves waste volume as a possible source of error. It is not clear if the cladding waste volume associated with processing of coproduct drivers was tabulated as Zirflex decladding waste or as a separate decladding waste type. The zirconium concentration in cladding waste generated during processing of coproduct drivers is much smaller than zirconium concentration during processing of the standard N Reactor fuel. Assuming that coproduct decladding waste had the same zirconium concentration as the standard 0.947 percent enriched N Reactor fuel may have overestimated the zirconium from the 1966 to 1972 PUREX processing by about 150 MT of zirconium.

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Best-Basis Value for Zirconium. The best-basis value for the zirconium inventory is the value developed by the independent evaluation, 440 MT. The reasons for using the independent evaluation value are as follows:

- Knowledge of the zirconium-clad fuel processing history has recently been updated and is not yet reflected in the HDW model.
- The zirconium content of the PUREX Plant cladding waste stream as modeled by the HDW spreadsheet cannot be reconciled with amount of zirconium cladding known to be used in N Reactor fuel.
- The TWRS baseline inventory has an uncertain basis. Several potential sources of error were identified that could not be resolved.

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6.0 RADIONUCLIDES

Determination of the radionuclide inventories involves consideration of the date the radionuclides were generated in the reactor, the particular process through which they were subsequently processed, and the decay rate and decay chain specific to each isotope. Section 6.1 provides the basis used to develop inventory estimates of the 46 key radionuclides. Most of these radionuclides were considered waste and were directly discharged to the various process waste streams. However, some of the isotopes were considered to have beneficial use. Most notable in this respect were the isotopes of Pu, U, ^{237}Np , ^{137}Cs , and ^{90}Sr . Due to the unique aspects of recovering these materials, a separate discussion of ^{137}Cs and ^{90}Sr disposition is provided in Section 6.2, and uranium is discussed in Section 6.3.

6.1 ESTABLISH BEST INVENTORY FOR RADIONUCLIDES

Historically, global inventory values for Hanford Site waste tanks have been generated by reactor fuel activity codes with supplemental separations plant waste analysis data for certain radionuclides. To date, analysis of waste samples taken from the tanks has not been relied on to predict total quantities since not enough tanks have been sampled and comprehensively analyzed. Past calculational methods have been as follows:

- RIBD-II Code (Gumprecht 1968) for fission products plus hand calculations for activation products
- The TRAC model, which used factors derived from the RIBD code
- The RADNUC code (Schwarz 1995), which uses tables of Ci/MTU factors generated by ORIGEN2
- A simplified ORIGEN2 code calculation (Wittekind 1989) that scales annual radionuclide production proportional to annual fuel exposure (megawatt days).

Results from the RIBD Code and TRAC model methods were published as Engineering Support Data (RHO 1985a) for the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) and are also reported to the national Integrated Database Report (DOE 1994). Unfortunately, an adequate documentation trail for these global inventory data sets no longer exists, and the primary codes, RIBD and TRAC, are no longer operational on Hanford Site computers.

The RADNUC code has been successfully used to predict radionuclide inventories in fuel and PUREX process waste associated with N Reactor operations from 1972 through 1987. However, significant code modifications would be required before RADNUC could be used to predict contributions to the global inventory from pre-1972 operations.

The simplified ORIGEN2 method has been recently used to estimate global inventories (i.e., for all reactors, 1944 to present) and to predict radionuclide compositions for LLW (Schmittroth 1995). Results generated by the ORIGEN2 model should be relatively easy to document since the ORIGEN2 code, itself, is well documented and accepted.

However, for the task of generating tank-by-tank inventory estimates for radionuclides, neither the RADNUC code nor the simplified ORIGEN2 code alone can easily provide the month-by-month definition of waste stream activities that is needed. Even the task of modelling global radionuclide inventories requires complex modelling because global inventory values must agree with the sum of tank-by-tank inventory values and to make this agreement possible much of the month-by-month data describing separation plant production must be incorporated in the global calculation. Thus, the global calculation takes on the complexity of the tank-by-tank calculation.

The following sections describe the requirements for the overall modelling task and the model architecture that has been set up to carry out the global and tank-by-tank estimates. Tables of supporting data and assumptions are given in Appendix G of this report. Section 6.1.8 summarizes best-basis radionuclide inventory data for the tank system, listing values for the 46 key radionuclides.

6.1.1 Requirements for a Global Tank Radionuclide Inventory Model

The task of calculating global inventories for many radionuclides is more complicated than in the case of chemicals. Not only must radionuclide fill-data (additions to tanks) be determined but, in the case of radionuclides, the concurrent effects of decay and buildup also must be computed. The waste activity contribution of short-cooled, low-exposure fuel irradiated in the forties and early fifties is uniquely different from that of longer-cooled, higher-exposure fuel irradiated in the seventies. It is obvious that one or more computer codes or models is needed to support this type of calculation.

Specifically, the calculation must begin with a "fuel activity generation" model such as ORIGEN2 to calculate the curie levels of fission products, activation products and actinides in individual batches of fuel discharged from Hanford Site reactors. Modelling of curie levels in discharged reactor fuel needs to account for variables of: (1) production (tons of fuel sent to separations plants per month), (2) fuel type (uranium enrichment or ^{235}U concentration as well as cladding type), (3) impurity levels affecting the generation of activation products, and (4) fuel exposure (megawatt days per metric ton of fuel [MWd/MTU]).

The ORIGEN2 code or other code must account for the effects of radionuclide decay during variable fuel cooling periods in reactor storage basins and later during waste aging periods in storage tanks. The buildup of ^{241}Am in the fuel and waste is particularly sensitive to effects of fuel exposure and reactor-to-separations cooling time.

Next, the chemical separation of extractable elements (Tc, U, Th, Np, and Pu) and volatile elements (^{14}C , Kr, I, and Xe) must be modelled so as to predict the quantities of these radionuclides entering the system of 177 waste tanks.

Finally, a waste chemistry model must be employed to account for the effect of radionuclide solubilities and the loss of certain radionuclides from the tank system during the transfer of waste supernatants to cribs during the mid-fifties.

6.1.2 Requirements for a Tank-by-Tank Radionuclide Inventory Model

In addition to the global requirements, the overall model must calculate tank-fill data (separation plant output) for each of several waste streams and over relatively short-time intervals (monthly or shorter) so that the quantities of radionuclides entering specific receiving tanks can be properly accounted for. The model must also account for solubility effects on the transfer of certain radionuclides from tank-to-tank during the subsequent history of waste transfers.

6.1.3 Radionuclides to be Tracked

Forty-six key radionuclides have been identified for tracking in development of a best-basis inventory for Hanford Site tank wastes (see Table 6.1-1 and Appendix A of this report). Of these 46 radionuclides, five (^{14}C , ^{59}Ni , ^{63}Ni , ^{60}Co , and ^{125}Sb) are generated totally or in part by neutron activation of minor alloy or impurity elements present in the array of Hanford reactor fuels. The calculation of inventory values for these five radionuclides is therefore more uncertain due to the necessary approximations made in setting concentration parameters for these precursor elements in the fuels.

Another subgroup of key radionuclides (^{226}Ra , ^{227}Ac , ^{228}Ra , ^{229}Th , and ^{231}Pa) deserves mention. These are alpha decay chain daughters of parent radionuclides such as ^{232}Th , ^{233}U , ^{234}U , and ^{235}U . With the exception of ^{228}Ra , these daughter nuclides are expected to slowly buildup in Hanford tank wastes, not reaching peak activity levels until thousands of years in the future. (The ^{228}Ra will likely peak much sooner). Data users must recognize that the grossly approximate estimates for these five nuclides presented, herein, are of little value in the long term waste migration scenarios for which they are of concern. Further decay calculations derived from inventory values of the parent nuclides will be required to support long term release scenario evaluations. For this reason, no attempt has been made to refine the estimate of near-term activity for these five nuclides.

Table 6.1-1. Key Radionuclides to be Added to the Hanford Defined Waste Model.

^3H	^{129}I	^{233}U
^{14}C	^{134}Cs	^{234}U
^{59}Ni	^{137}Cs	^{235}U
^{60}Co	$^{137\text{m}}\text{Ba}$	^{236}U
^{63}Ni	^{151}Sm	^{237}Np
^{79}Se	^{152}Eu	^{238}Pu
^{90}Sr	^{154}Eu	^{238}U
^{90}Y	^{155}Eu	^{239}Pu
^{93}Zr	^{226}Ra	^{240}Pu
$^{93\text{m}}\text{Nb}$	^{227}Ac	^{241}Am
^{99}Tc	^{228}Ra	^{241}Pu
^{106}Ru	^{229}Th	^{242}Pu
$^{113\text{m}}\text{Cd}$	^{231}Pa	^{242}Cm
^{125}Sb	^{232}Th	^{243}Am
^{126}Sn	^{232}U	^{243}Cm
		^{244}Cm

6.1.4 Requirements for Output Reporting

Output from the overall model should provide both global and tank-by-tank inventories for the 46 key radionuclides with activity values decayed to January 1, 1994. In future versions the total elemental mass of these radionuclides also should be tracked and reported.

6.1.5 Model Architecture

One relatively straightforward way of generating global and tank-by-tank inventory values for these key radionuclides was to modify the Hanford Defined Waste (HDW) model to carry data fields for these 46 radionuclides. This existing model provides the calculational framework to identify receiver tanks for the various waste streams exiting the separations plants and to model subsequent tank-to-tank and tank-to-crib waste transfers and solubility effects.

Providing the HDW model with the necessary radionuclide input data is a Fortran code named DKPRO plus a number of spreadsheets, and existing databases linked together as illustrated by the data flow schematics of Figures 6.1-1 and 6.1-2, and described in Section 6.1.5.1.

An alternative way of generating global inventory values is also depicted in Figure 6.1-2. Here, the same fuel activity output file generated by the DKPRO code is coupled to a Hanford spreadsheet (named "SPLIT" for discussion purposes, here) which duplicates the role of the HDW model in accounting for the removal of certain radionuclides during fuel separations processing and waste fractionation operations. The spreadsheet SPLIT applies waste loss factors to individual fuel batches and calculates total inventory values accumulated in tank waste. Both of these model architectures are discussed, below, and compared.

6.1.5.1 ORIGEN2 and DKPRO Codes. At the center of Figure 6.1-1 is a box representing the newly created fortran code named DKPRO. The objective of the DKPRO code is to generate an output file of "fuel activity records"--curie values for 46 key radionuclides contained in batches of reactor fuel input monthly (or for shorter periods) to individual separations plants. The code, DKPRO, has the general capability to estimate radionuclide activity values in batches of reactor fuel (or fuel separations plant waste), using output from the ORIGEN2 code. However, to avoid an excessive number of ORIGEN2 calculations a "table interpolation technique" is used. Output from 15 ORIGEN2 runs, spanning various reactor fuel types and exposure levels, are used as a data library in DKPRO. The job of DKPRO is to read a second data file, the "Unified Separations Production File," and for each of approximately 1,300 fuel batch records (e.g., data on fuel type, fuel tonnage, exposure, separations date, and cooling time) calculate radionuclide activities in that batch of fuel by interpolating the ORIGEN2 output data library as a function of fuel type and exposure (MWd/MTU). (See Watrous and Wootan [1997] for further details of the ORIGEN2 and DKPRO code setup and for a discussion of how the Unified Separations Production File was generated.)

For the purpose of this radionuclide modelling task, Hanford Site fuels have been classified in six fuel types, as defined in Table 6.1-2.

Figure 6.1-1. Data Flow Schematic--Generation of Fuel Activity Records from ORIGEN2.

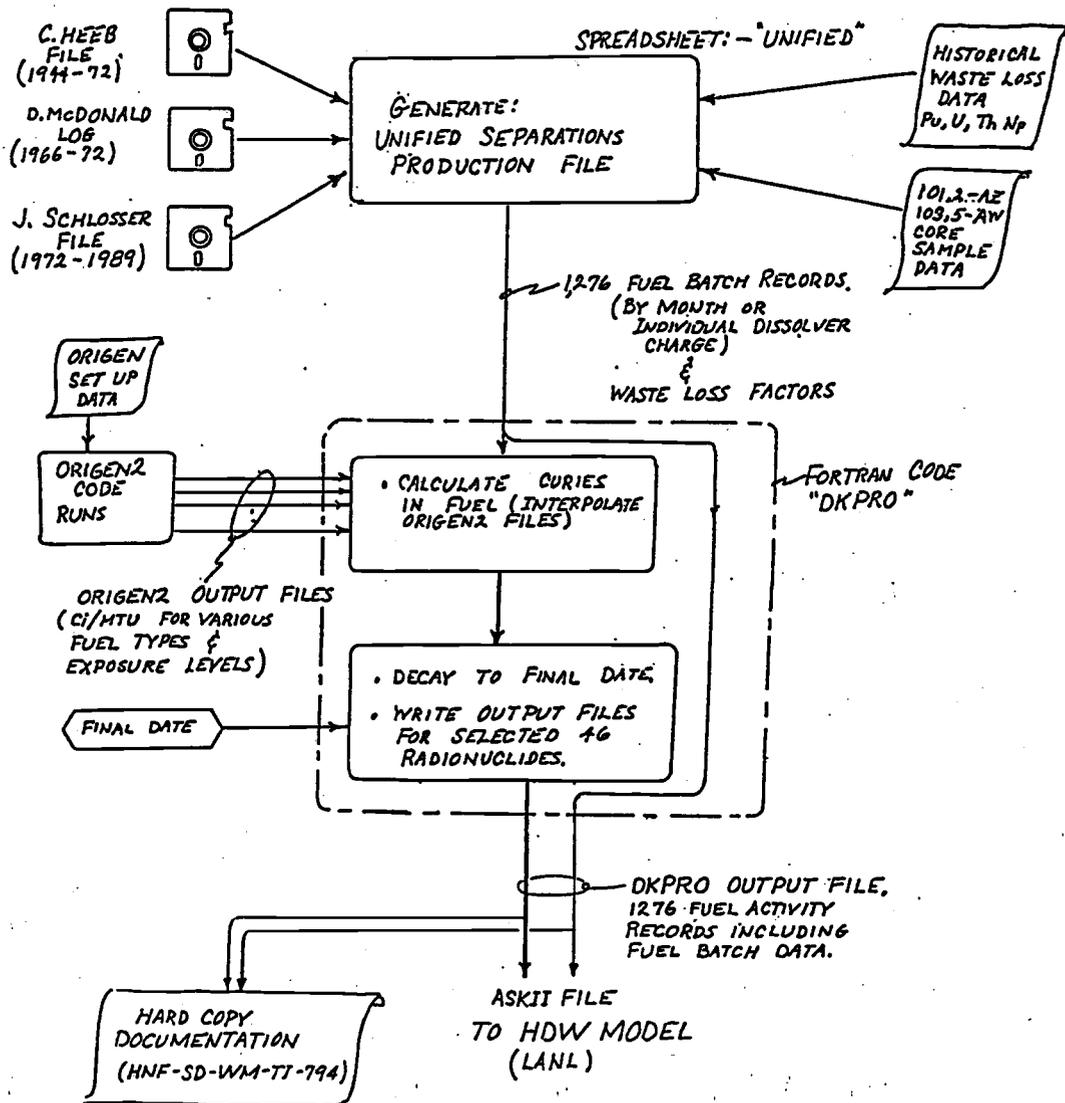


Figure 6.1-2. Data Flow Schematic--Generation of Tank Radionuclide Inventories.

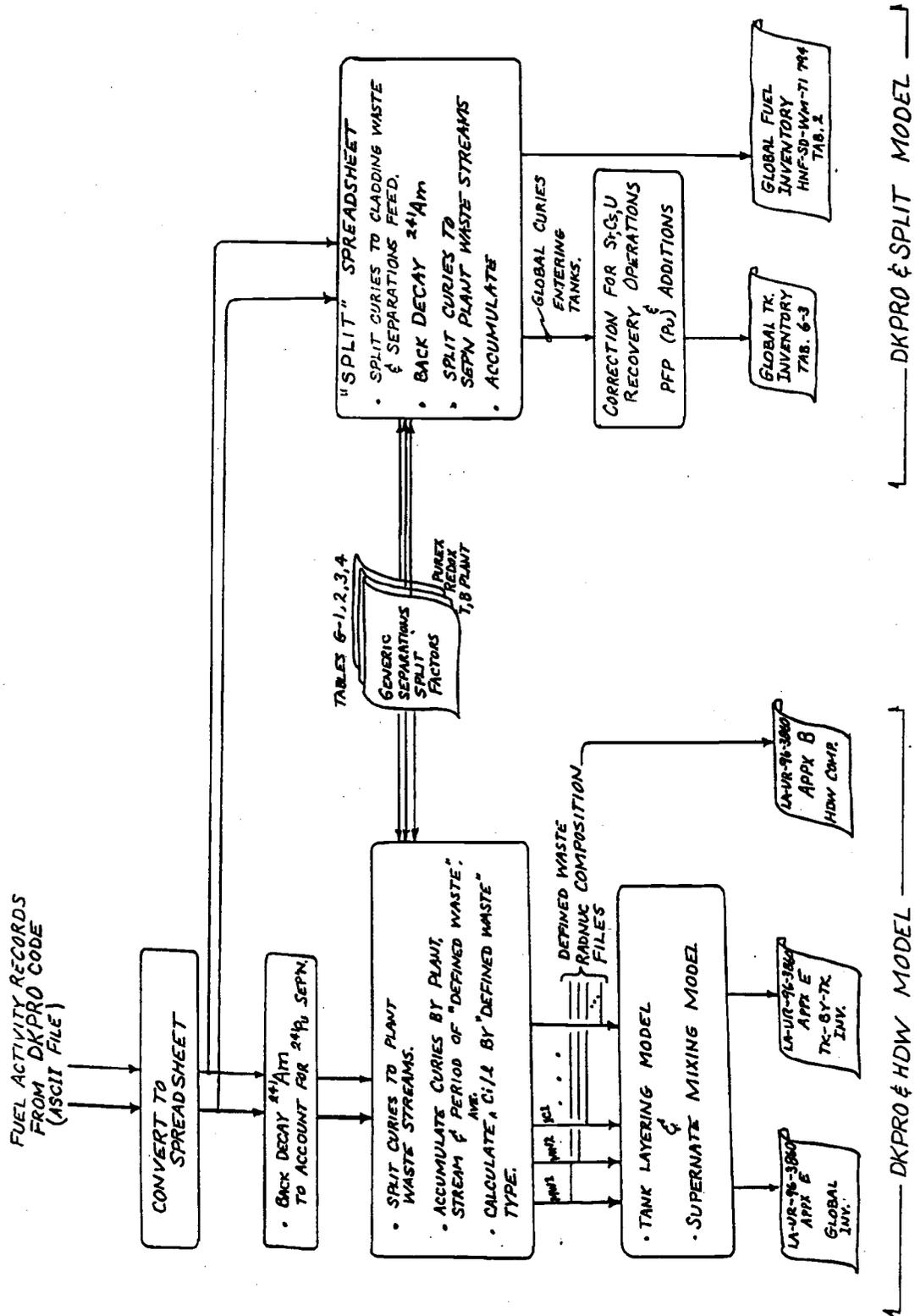


Table 6.1-2. Definition of Fuel Types and Fuel Codes for Radionuclide Inventory Modelling.

Code	Fuel definition
Al-0.71U	Aluminum clad, natural enrichment uranium (0.71 percent ^{235}U) -- either solid core or I&E. Single pass reactor fuel.
Al-0.94U	Aluminum clad, enriched uranium (0.94 percent ^{235}U). Single pass reactor fuel.
Zr-0.94U	Zircaloy®-clad, enriched uranium (0.94 percent ^{235}U). N Reactor fuel.
Zr-1.1U	Zircaloy®-clad, spike uranium (0.94 percent ^{235}U inner fuel tube and 1.25 percent ^{235}U outer fuel tube, together averaging 1.1 percent ^{235}U). N Reactor fuel.
Zr-2.1U	Zircaloy®-clad, co-product driver uranium (2.1 percent ^{235}U). N Reactor fuel.
Al-0.0Th	Aluminum-clad, thorium oxide target elements. Single pass reactors.

In this current version of the DKPRO code/HDW model interface, the DKPRO code calculates radionuclide decay from the date of fuel discharge to the "standard inventory reference decay date," January 1, 1994, accounting for the unique timing of each fuel batch. This "pre-decay" feature avoids the complexity of adding decay functions for all radionuclides in the LANL portion of the overall model. In general DKPRO also has the capability to account for the separation (i.e., the partial removal) of various radionuclides during fuel processing operations. In this current application, however, while DKPRO accounts for the total decay of radionuclides in fuel, it does not account for the chemical separation of routinely extracted elements: Tc (partial extraction), Th, U, Np, and Pu. Also, it does not account for the removal (to gaseous effluents, silver reactors, or process condensates to cribs) of volatile elements, including ^3H , ^{14}C , and ^{129}I . In effect, the "fuel activity file" output by this version of DKPRO represents the activity of fuels as if they were held from the date of reactor discharge to January 1, 1994, but without being processed through separations.

Thus, while fuel activity records for non-separated radionuclides can be used directly as values input to tanks, further data correction must be done (i.e., in the HDW or SPLIT models) to account for the separated radionuclides mentioned, above. A special correction is also required in the case of ^{241}Am to account for the removal of its parent, ^{241}Pu , at the fuel separation date. Note that this correction for ^{241}Am remaining in tank waste on January 1, 1994, is very sensitive to the fuel cooling time, the fraction of ^{241}Pu remaining in waste, and the waste aging period (separation date to January 1, 1994).

Output from the DKPRO code is an electronic file of 1,276 waste activity records of pre-decayed radionuclide values, each record representing the curies associated with a batch of fuel processed by month (or a shorter period) through individual separation plants. Within each record DKPRO generates two sub-records: (1) curies in the fuel cladding and (2) total curies in fuel cores to be fed to extraction operations. The calculation splitting activation products between cladding and fuel cores is based on target element concentrations in fuel cladding and cores. These data are presented in Appendix G of this report as Table G-5. The DKPRO output file also carries data fields giving batch-specific waste loss factors for extractable elements (U, Th, Np, and Pu) for use later in the HDW and SPLIT models.

Format of the DKPRO output file is explained in Watrous and Wootan (1997).

6.1.5.2 Hanford Defined Waste Model. During FY 1996, the HDW model (then under development at LANL) was modified to carry data fields for the 46 key radionuclides in addition to data fields for inert chemicals. Then, as indicated in Figure 6.1-2, a separations partitioning model was, in effect, added to the overall LANL model to account for the partial removal of U, Th, Np, and Pu during fuel separations processing, and to generate "defined waste compositions" (in terms of radionuclides) for various separations process waste streams. Waste stream split factors and separations loss factors for U, Th, Np, and Pu as used in the HDW model are described in Agnew et al. (1997a).

In FY 1996, modifications to the then existing HDW model were made to account for the solubility behavior of radionuclides. Many of the 46 key radionuclides are elements whose solubility was already modelled (e.g., Sr, Cs, U, Pu, and Am). Others of the key radionuclides are elements that are new to the HDW model (e.g., Se, Co, or Ru) and for which only a grossly approximate definition of solubility behavior has been programmed (i.e., the assumption of a 50/50 split of the radionuclide between sludges and supernatants).

The HDW model also accounts for the removal from tank inventory of all soluble radionuclides that were routed to cribs during tank volume recovery operations in the 1950's. Likewise, the loss of tritium (^3H) to evaporator and boiling tank condensate is accounted for in the model as is the separation of Sr-Y and Cs-Ba during waste fractionization operations in B Plant.

Results from the HDW model are given in Agnew et al. (1997a), in terms of tank by tank curie inventories and global inventories (see Appendix E of Agnew et al. 1997a). The global inventory results were also copied into Table 6.1-3 (under the heading DKPRO/HDW model), where they can be compared to other inventory data sources.

Table 6.1-3. Global Radionuclide Inventory Comparisons.
(Curies remaining in 177 tanks - decayed to January 1, 1994) (2 Sheets)

Nuclides	Calculated by DKPRO/SPLIT model	Calculated by DKPRO/HDW model	From IDB Rev. 12 ^a	From TWRS FEIS ^b	From nuclear material accountability and samples ^c
³ H	3.40 E+04	7.16 E+04			
¹⁴ C	3.28 E+03	4.81 E+03	4.57 E+03	5.34 E+03	
⁵⁹ Ni	1.18 E+03	9.34 E+02			
⁶⁰ Co	1.33 E+04	1.23 E+04			
⁶³ Ni	1.17 E+05	9.20 E+04			
⁷⁹ Se	1.00 E+03	7.73 E+02			
⁹⁰ Sr	7.16 E+07	6.16 E+07	6.25 E+07	6.19 E+07	
⁹⁰ Y	7.16 E+07	6.16 E+07	6.25 E+07	6.19 E+07	
⁹³ Zr	4.74 E+03	3.63 E+03			
^{93m} Nb	3.48 E+03	2.69 E+03			
⁹⁹ Tc	2.47 E+04	3.26 E+04	3.21 E+04	3.21 E+04	
¹⁰⁶ Ru	1.14 E+05	1.04 E+05			
^{113m} Cd	2.24 E+04	1.69 E+04			
¹²⁵ Sb	2.19 E+05	2.08 E+05			
¹²⁶ Sn	1.53 E+03	1.19 E+03			
¹²⁹ I	4.57 E+01	6.30 E+01	2.98 E-01	3.83 E+01	
¹³⁴ Cs	8.89 E+04	8.82 E+04			
¹³⁷ Cs	4.64 E+07	4.71 E+07	3.95 E+07	4.01 E+07	
^{137m} Ba	4.39 E+07	4.46 E+07	3.74 E+07	3.79 E+07	
¹⁵¹ Sm	3.54 E+06	2.75 E+06	1.07 E+06		
¹⁵² Eu	1.89 E+03	1.48 E+03			
¹⁵⁴ Eu	1.74 E+05	1.47 E+05			
¹⁵⁵ Eu	1.60 E+05	1.36 E+05			
²²⁶ Ra	7.72 E-02	6.31 E-02			
²²⁷ Ac	1.08 E+02	8.76 E+01			
²²⁸ Ra	6.44 E+01	7.71 E+01			
²²⁹ Th	2.92 E+01	1.81 E+00			

Table 6.1-3. Global Radionuclide Inventory Comparisons.
(Curies remaining in 177 tanks - decayed to January 1, 1994) (2 Sheets)

Nuclides	Calculated by DKPRO/SPLIT model	Calculated by DKPRO/HDW model	From IDB Rev. 12 ^a	From TWRS FEIS ^b	From nuclear material accountability and samples ^c
²³¹ Pa	1.91 E+02	1.56 E+02			
²³² Th	2.11 E+00	4.36 E+00			
²³² U	1.19 E+02	2.59 E+02			1.23 E+02
²³³ U	4.60 E+02	9.94 E+02			4.76 E+02
²³⁴ U	2.78 E+02	8.50 E+02			3.46 E+02
²³⁵ U	1.17 E+01	3.66 E+01			1.45 E+01
²³⁶ U	7.69 E+00	1.61 E+01			9.57 E+00
²³⁷ Np	1.43 E+02	1.41 E+02			
²³⁸ Pu	2.77 E+03	2.08 E+03			1.45 E+03
²³⁸ U	2.59 E+02	9.06 E+02			3.22 E+02
²³⁹ Pu	3.91 E+04	4.70 E+04	2.64 E+04	2.64 E+04	3.59 E+04
²⁴⁰ Pu	8.93 E+03	9.87 E+03	6.69 E+03	6.70 E+03	8.97 E+03
²⁴¹ Am	8.07 E+04	6.99 E+04	1.04 E+05	1.04 E+05	
²⁴¹ Pu	2.29 E+05	1.67 E+05	1.03 E+05	1.00 E+05	1.47 E+05
²⁴² Pu	1.16 E+00	7.90 E-01	2.80 E-01		1.06 E+00
²⁴² Cm	1.10 E+02	7.70 E+01			
²⁴³ Am	4.19 E+01	9.34 E+00			
²⁴³ Cm	1.37 E+01	1.00 E+01			
²⁴⁴ Cm	3.14 E+02	2.42 E+02			

FEIS = Final Environmental Impact Statement

HDW = Hanford Defined Waste

IDB = Integrated Database

TWRS = Tank Waste Remediation System

^aIDB Data from Rev. 12 (DOE 1996a), Table 2.11, corrected to January 1, 1994

^bTWRS Final Environmental Impact Statement (DOE 1996b), Appendix A, corrected to January 1, 1994

^cAccountability data for period 1944-1971 from (ERDA 1977). Sample Data for 1972-1989 Production, from Tank Characterization Reports (DiCenso and Simpson 1994, Hodgson 1995a and b, and Ryan 1995). (No decay date adjustment due to long half lives.)

6.1.5.3 DKPRO/SPLIT Model. The combination of the DKPRO output file (1,276 fuel activity records) and the SPLIT spreadsheet serves as an alternate method for estimating global radionuclide inventories (but not tank-by-tank inventories). In its use of separations processing split factors for ^{14}C , ^{99}Tc , ^{129}I , the isotopes of U, Th, Np, and Pu, and material balance data for encapsulated ^{90}Sr , ^{137}Cs , recovered U and added Pu (from PFP operations), the DKPRO/SPLIT methodology is independent of HDW model assumptions. This methodology uses an alternate set of separations waste loss factors that are carried on the DKPRO output file. Likewise, an alternate set of separations waste split factors, given in Appendix G, are used to account for separations plant removal of ^{14}C , ^{99}Tc , and ^3H . These split factors are organized in three tables, one for each separation process (BiPO₄ [B and T Plants], REDOX, and PUREX), as shown in Tables G-2, G-3, and G-4. The separations partitioning model uses these defined factors to calculate the partitioning of fuel activity into the several waste streams discharged to tanks from the separations plants.

Like the modified HDW model, the SPLIT spreadsheet also calculates a decay correction for ^{241}Am to account for the timing of ^{241}Pu separation from each fuel batch and its effect on ^{241}Am buildup in the waste.

Unlike the HDW model, the DKPRO/SPLIT model does not account for losses of soluble radionuclides in tank waste leaks or supernate transfers to cribs in the early years. However, these effects are estimated to be minor. Global Inventory results from the DKPRO/SPLIT method are summarized in Table 6.1-3.

6.1.6 Comparison of Global Inventory Model Results

Global Inventory results from the above two alternative models are listed in Table 6.1-3 along with two previously published tank inventory data bases--the Integrated Data Base report, Rev. 12 (DOE 1996a) and the TWRS Final EIS (DOE 1996b).

For uranium and plutonium an additional source of global tank inventory information exists in a combination of accountability records and tank sample data. Table 6.1-4 illustrates the generation of global inventory values for uranium isotopes. Here, accountability data (based on waste stream concentrations measured at the separations plants) are used to represent the waste losses associated with production from 1944 through 1971, while tank sample data (for tanks 241-AZ-101, 241-AZ-102, 241-AW-103, and 241-AW-105) give a truer representation of waste losses associated with post 1971 production (zirconium clad fuel processing operations).

The estimate for plutonium is similar to that for uranium (waste stream measurements for the 1944 through 1971 period plus tank sample analyses for the post 1971 production period). However, the "accountability/sample" value for Pu also accounts for the addition of 142 kg of Pu in waste from Plutonium Finishing Plant (PFP) operations, added in the period 1981 and later.

Table 6.1-4. Calculation of Global Uranium Isotopic Inventory from Accountability Records and Tank Samples.

1 Nuclide	2 Ci/g	3 Curies by DKPRO/ SPLIT ^a	4 Curies (44-71) from ERDA 77-44	5 Curies (72-89) from TCRs	6 Calculated Activity*			8 Total curies ^b	9 Mass U ^h (g)
					Curies (44-71)	Curies (72-89)	Curies (72-89)		
²³² U	21.4	1.1918 E+02	-	-	1.2338 E+02 ^d	-	1.2338 E+02	5.7652 E+00	
²³³ U	0.009639	4.5980 E+02	476 ^b	-	476 ^d	-	4.7600 E+02	4.9383 E+04	
²³⁴ U	0.006249	2.7832 E+02	-	-	3.2250 E+02 ^e	2.3880 E+01 ^f	3.4638 E+02	5.5430 E+04	
²³⁵ U	2.2 E-06	1.1689 E+01	13 ^b	-	1.3544 E+01 ^e	1.0029 E+00 ^f	1.4547 E+01	6.7286 E+06	
²³⁶ U	6.5 E-05	7.6861 E+00	-	-	8.9062 E+00 ^e	6.5946 E-01 ^f	9.5656 E+00	1.4785 E+05	
²³⁸ U	3.4 E-07	2.5890 E+02	300 ^b	-	3.0000 E+02 ^e	2.2213 E+01 ^f	3.2221 E+02	9.5840 E+08	
Total U mass (g):		7.7566 E+08	-	6.655 E+07 ^e				9.6538 E+08	

^aUranium isotopic estimate from DKPRO/SPLIT model, Table 6.1-3.

^bAvailable curie data from accountability records for the waste accumulation period (1944 through 1971). Value for ²³³U (400 Ci) from ERDA 77-44 has been corrected to 476 Ci to match original measured waste stream losses of 49.4 kg ²³³U.

^cTank sample data for 1972 to 1989 production period is summed from Tank Characterization Reports (TCR) (DiCenso and Simpson 1994, Hodgson 1995a and b, and Ryan 1995).

^dCurie value for ²³²U is estimated from ²³³U (Column 4), in ratio to Column 3 curies.

^eCurie values for ²³⁴U, ²³⁵U, and ²³⁶U are estimated from ²³⁸U (Column 4), in ratio to Column 3 curies.

^fCurie values for ²³⁴U, ²³⁵U, ²³⁶U, and ²³⁸U are estimated from U mass (Column 5), in ratio to Column 3 curies. Specific activity constants from Column 2 are first used to calculate the equivalent mass for Column 3 curies (7.7566 E+08 g U).

Example for ²³⁴U:

$$2.388 \text{ E}+01 \text{ Ci} = (2.7832 \text{ E}+02 \text{ Ci})(6.655 \text{ E}+07 \text{ g U})/(7.7566 \text{ E}+08 \text{ g U})$$

^gTotal curies are the sum of Columns 6 and 7.

^hIsotopic mass and total mass are converted from Column 8, using specific activity constants from Column 2.

*Values filled by Ratio to DKPRO Curies in column 3.

The Table 6.1-3 comparison of DKPRO/HDW model results to DKPRO/SPLIT model results shows minor discrepancies for most radionuclides (differences of about 20 percent or less) and somewhat larger discrepancies for those elements that have experienced extraction or volatilization during fuel separations operations or waste fractionation processing. This model-to-model comparison is also illustrated by Figure 6.1-3, which shows an X-Y scatter plot of DKPRO/HDW versus DKPRO/SPLIT model results. Here, it can be seen that (relative to the DKPRO/SPLIT model) the DKPRO/HDW model predicts significantly higher values for ^3H , ^{129}I , ^{232}Th , and all U isotopes. These differences are largely a result of the DKPRO/HDW model's not accounting for volatilization losses or using conservatively high waste loss factors. For example, in the case of ^{233}U , detailed accountability records for the 1966 and 1970 Thoria fuel processing campaigns (Isochem 1967, Jackson and Walser 1977) indicate that a global total of 49.4 kg of ^{233}U (476 Ci) was lost to waste tanks. This value compares well with ^{233}U results of the DKPRO/SPLIT model (460 Ci). In contrast, the DKPRO/HDW global estimate for ^{233}U (994 Ci) exceeds the accountability value by a factor of 2.1.

Comparison of both model results to previous inventories (i.e., to IDB and TWRS FEIS reports) shows close agreement between the DKPRO/HDW result for ^{90}Sr and previous inventories, whereas the DKPRO/SPLIT model predicts 15 percent more. Comparison for ^{137}Cs shows that DKPRO/HDW results agree closely with DKPRO/SPLIT results and both exceed previous inventories by about 13 percent to 17 percent.

For most radionuclides the discrepancies between both models and previous inventory lists are so minor (generally less than 20 percent) as to make it difficult to judge which model may be closer to reality. However, the relatively larger discrepancies for tritium, ^{129}I , Th, U, and Pu isotopes invite a closer analysis.

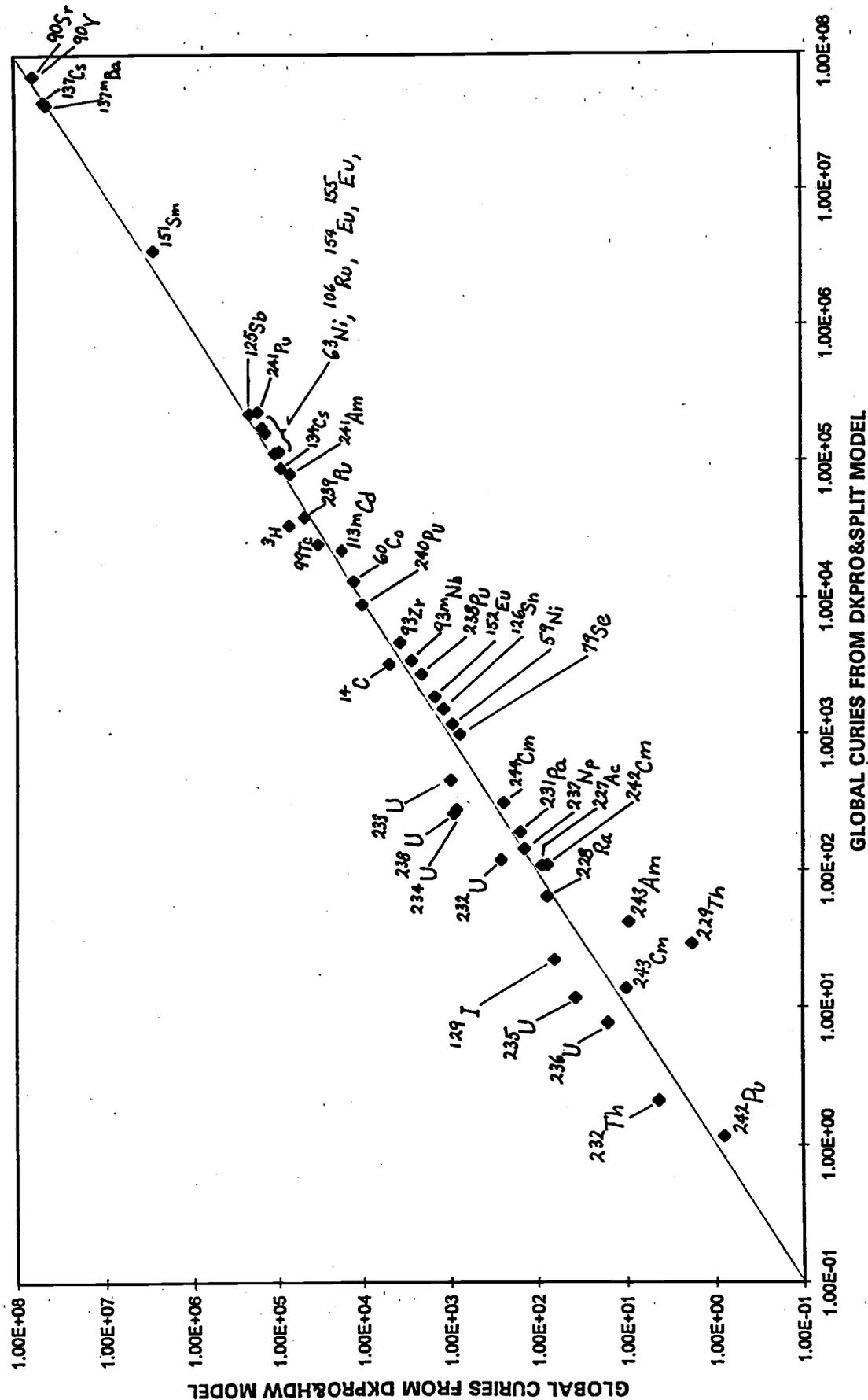
- Tritium values estimated by the DKPRO/SPLIT model attempt to account for losses of ^3H to separations process condensates and atmospheric emissions, whereas the DKPRO/HDW model assumes that 100 percent of ^3H in fuel is routed to tank waste. Thus, it is reasonable that DKPRO/SPLIT tritium values should be lower (and a more realistic prediction) than DKPRO/HDW results.
- The ^{99}Tc inventory value from the DKPRO/SPLIT model is 76 percent of the value predicted by the DKPRO/HDW model. This difference occurs because the DKPRO/SPLIT model accounts for an estimated 24 percent of technetium being extracted and routed to the uranium product in PUREX operations and for 37 percent being extracted in REDOX operations. The DKPRO/HDW model, on the other hand, assumes no loss of Tc during fuel separations processing. Due to the uncertainties in the Tc extraction factors used in the DKPRO/SPLIT model, the ^{99}Tc inventory predicted by the DKPRO/HDW model (32,600 curies) is judged to be the more reliable and conservative bounding value.
- Iodine values estimated by the DKPRO/SPLIT model attempt to account for losses of ^{129}I to atmospheric emissions, solid waste burials (spent silver reactors)

and silver reactor flushes that were routed to cribs. The model assumes split factors for iodine (fraction routed to tank waste) of 0.4, 0.4, 0.4, and 0.8 for T, B, REDOX, and PUREX plants, respectively. In effect an overall split factor of 0.71 results for the combined four plant throughput. In contrast, the DKPRO/HDW model makes the more conservative and simplifying assumption that essentially all of the ^{129}I in reactor fuel was routed to waste tanks (a split factor of 1.0). Due to the uncertainties in factors used in the DKPRO/SPLIT model, the ^{129}I inventory predicted by the DKPRO/HDW model (63 curies) is judged to be the more reliable and conservative bounding value.

- Although the DKPRO/HDW model's prediction for ^{232}Th at first appears to be the higher and more conservative value, the DKPRO/SPLIT model's value of 2.11 curies (roughly equivalent to 25 metric tons elemental thorium) is judged to be the more realistic, being based on accountability measurements made during the two thorium production campaigns in 1966 and 1970.
- Comparison of model results for uranium isotopes (from Table 6.1-3) to accountability/sample data (from Table 6.1-4) shows an interesting spread of values: In comparison to the accountability/sample curie values for ^{238}U (equivalent to a global inventory of 960 MTU), the DKPRO/SPLIT model predicts a value that is slightly lower (80 percent), while the DKPRO/HDW model predicts a value that is significantly higher by a factor of 2.8. The HDW result for ^{238}U , being a factor of 2.8 greater than previous estimates, has no basis in sample evidence to suggest that a correction of this magnitude could be real. (The apparent overprediction of the HDW model is thought to be due to the model's use of a conservative factor for fraction of metal waste not recovered.)
- For the plutonium isotopes, ^{239}Pu , ^{240}Pu and ^{242}Pu , DKPRO/SPLIT model results and accountability/sample derived values compare rather closely (a difference of 9 percent or less). This remarkably good agreement tends to support the validity of DKPRO/SPLIT model results for major Pu isotopes, and implies an equivalent Pu mass inventory range of 620 to 670 kg. DKPRO/HDW model results, being 20 to 30 percent higher, are likely less accurate due to relatively simplistic assumptions used in modelling. [Note that both DKPRO/SPLIT and accountability/sample inventory results include an estimated contribution from Plutonium Finishing Plant (PFP) waste that is equivalent to 142 kg of Pu.]

For the shorter half-lived Pu isotopes, ^{238}Pu and ^{241}Pu , the ratio comparison of DKPRO/SPLIT results over accountability/sample derived values is not as good, being factors of 1.9 and 1.6, respectively. Based on the understanding that the reported accountability values for ^{238}Pu were not actually measured but were calculated by ratio from earlier fuel activity codes, it seems reasonable to conclude that DKPRO/SPLIT values are likely the more accurate in regard to these minor isotopes. Therefore, DKPRO/SPLIT values are judged to be the best basis for a global Pu inventory.

Figure 6.1-3. Model Match--DKPRO and Hanford Defined Waste Versus DKPRO and Split.



6.1.7 Validation of Model Results from Tank Heat Measurements

Accuracy of DKPRO/HDW model predictions for total (global) ^{90}Sr and ^{137}Cs was tested by comparison of calculated equivalent decay heat to a combination of measured and independently estimated decay heat values for all tanks. "Measured" decay heat for 162 tanks (derived from a previous 1995 study that analyzed tank temperature data) (Kummerer 1995) was combined with engineering estimates for 15 other tanks (not measured in the 1995 study) to give a total tank decay heat value of 690 kilowatts (as of ca. 1994). Heat balance calculation results are summarized in Table 6.1-5.

Table 6.1-5. Comparison of Measured Tank Decay Heat to Model Predicted Heat.

Calculation of measured/estimated tank decay heat (as of ca. 1994)	
Measured Decay Heat in 162 Tanks (Watts) ^a	5.05 E+05
Calculated Decay Heat in AZ-101,2 (Watts) ^b	1.26 E+05
Estimated Decay Heat in 6 unmeasured DSTs (Watts) ^c	3.96 E+04
Estimated Decay Heat in 7 unmeasured SSTs (Watts) ^d	1.60 E+04
Total Decay Heat in 177 Tanks (Watts)	6.87 E+05
Model based tank decay heat (as of 1/1/1994)	
Cs/Sr Decay Heat Predicted from DKPRO/SPLIT Model (Watts)	6.87 E+05
Cs/Sr Decay Heat Predicted from DKPRO/HDW Model (Watts)	6.35 E+05

^aData from (Kummerer 1995).

^bBased on ^{137}Cs and ^{90}Sr predicted by the DKPRO/SPLIT model for 241-AZ-101, 241-AZ-102 tanks (PUREX throughput, 1983-1989).

^cEstimated by ratio to measured heat in 15 tanks in AN, AP, and AW farms. (Data from Kummerer 1995)

^dEstimated by ratio to measured heat in 33 tanks in TX, SX, and U farms. (Data from Kummerer 1995).

In comparison to this measured/estimated decay heat total of 687 kilowatts, the DKPRO/HDW model predicts a total decay heat of 635 kilowatts--a difference of only 8 percent, while the DKPRO/SPLIT model predicts a total decay heat of 687 kilowatts. (In this latter comparison, the perfect agreement is likely coincidental and should probably not be viewed as evidence that the DKPRO/SPLIT model is more valid than the DKPRO/HDW model.) This close comparison provides additional confirmation to the accuracy of the HDW model (for global values of non-extracted radionuclides). The close agreement also tends to validate precursor codes and databases such as the ORIGEN2 code (as set to model fuel activity for Hanford's single-pass reactors and N Reactor) and our historical Separations Production data file.

6.1.8 Selection of Best-Basis Global Radionuclide Inventory

From the comparisons made in sections 6.1.6 and 6.1.7, above, it is judged that DKPRO/HDW model global results are sufficiently accurate to serve as best basis values for most radionuclides (i.e., non-extracted elements). However, for extractable or volatile radionuclides (^3H , including iodine, and ^{232}Th) it is judged that results of the DKPRO/SPLIT model are generally a more realistic estimate of global tank inventories. For isotopes of uranium it is judged that the accountability/sample inventory as generated in Table 6.1-4 should be considered more valid than predictions of the DKPRO/SPLIT model and more correct than the unsupported high loss predictions of the DKPRO/HDW model. Likewise, accountability/sample inventory values for Pu given in Table 6.1-3 are considered to be the more valid.

Accordingly, the best-basis global radionuclide inventory given in Table 6.1-6 has been composed from DKPRO/SPLIT results (for ^3H , ^{232}Th), accountability/sample results from (for isotopes of U and Pu) plus DKPRO/HDW model results (for all other key radionuclides).

Table 6.1-6. Best-Basis Global Radionuclide Inventory. (2 Sheets)

Nuclide	Curies remaining in 177 tanks	Data source
	(Decayed to January 1, 1994)	(From DKPRO/HDW model unless stated otherwise)
^3H	3.40 E+04	Based on DKPRO/SPLIT model
^{14}C	4.81 E+03	
^{59}Ni	9.34 E+02	
^{60}Co	1.23 E+04	
^{63}Ni	9.20 E+04	
^{79}Se	7.73 E+02	
^{90}Sr	7.16 E+07	Based on DKPRO/SPLIT model
^{90}Y	7.16 E+07	Based on DKPRO/SPLIT model
^{93}Zr	3.63 E+03	
$^{93\text{m}}\text{Nb}$	2.69 E+03	
^{99}Tc	3.26 E+04	
^{106}Ru	1.04 E+05	
$^{113\text{m}}\text{Cd}$	1.69 E+04	
^{125}Sb	2.08 E+05	
^{126}Sn	1.19 E+03	
^{129}I	6.30 E+01	
^{134}Cs	8.89 E+04	Based on DKPRO/SPLIT model
^{137}Cs	4.64 E+07	Based on DKPRO/SPLIT model

Table 6.1-6. Best-Basis Global Radionuclide Inventory. (2 Sheets)

Nuclide	Curies remaining in 177 tanks	Data source
	(Decayed to January 1, 1994)	(From DKPRO/HDW model unless stated otherwise)
^{137m} Ba	4.39 E+07	Based on DKPRO/SPLIT model
¹⁵¹ Sm	2.75 E+06	
¹⁵² Eu	1.48 E+03	
¹⁵⁴ Eu	1.47 E+05	
¹⁵⁵ Eu	1.36 E+05	
²²⁶ Ra	6.31 E-02	
²²⁷ Ac	8.76 E+01	
²²⁸ Ra	7.71 E+01	
²²⁹ Th	1.81 E+00	
²³¹ Pa	1.56 E+02	
²³² Th	2.11 E+00	Based on DKPRO Waste Loss Factor.
²³² U	1.23 E+02	Based on Accountability/Sample Data.
²³³ U	4.76 E+02	Based on Accountability/Sample Data.
²³⁴ U	3.46 E+02	Based on Accountability/Sample Data.
²³⁵ U	1.45 E+01	Based on Accountability/Sample Data.
²³⁶ U	9.57 E+00	Based on Accountability/Sample Data.
²³⁸ U	3.22 E+02 (or 958 MT as defined in Figure 6.3-1)	Based on Accountability/Sample Data.
²³⁷ Np	1.41 E+02	
²³⁸ Pu	2.77 E+03	Based on DKPRO/SPLIT Model.
²³⁹ Pu	3.91 E+04	Based on DKPRO/SPLIT Model.
²⁴⁰ Pu	8.93 E+03	Based on DKPRO/SPLIT Model.
²⁴¹ Pu	2.29 E+05	Based on DKPRO/SPLIT Model.
²⁴² Pu	1.16 E+00	Based on DKPRO/SPLIT Model.
²⁴¹ Am	6.99 E+04	
²⁴² Cm	7.70 E+01	
²⁴³ Am	9.34 E+00	
²⁴³ Cm	1.00 E+01	
²⁴⁴ Cm	2.42 E+02	

HDW = Hanford Defined Waste.

Note that for ^3H , ^{129}I , Th, and U isotopes, global values as estimated in Table 6.1-6 may not necessarily match the sum of the tank-by-tank values given in Agnew et al. (1997a), Appendix E or the sum of best-basis values for all 177 tanks. These expected discrepancies are to be reconciled in future work.

6.1.9 Summary of Errata and Accuracy Issues for Table 6.1-6

Comparisons made in the sections, above, show that best-basis global inventory results (as given in Table 6.1-6) are reasonably accurate for major radionuclides such as ^{90}Sr , ^{137}Cs , ^{233}U , and plutonium--those radionuclides that can be compared to some independent measurement. The basic assumption being made in this methodology is that if the ORIGEN2 code and historical Separations Production data file can, together, accurately predict inventory values for certain major radionuclides, then values for all other (unmeasured) radionuclides can be predicted with equal accuracy. Although this assumption is generally true, there are a few radionuclides (activation products and extractable elements) for which model predictions will be more uncertain.

In the current version of the DKPRO/HDW model, several simplifying assumptions have been made that affect the accuracy of global and tank-by-tank inventory estimates for the following radionuclides:

- Losses of Tritium to process condensate and to atmosphere at the four separations plants can vary markedly, depending on the degree of waste stream evaporation/concentration. The global inventory value given on Table 6.1-6 is based on tritium split factors of 1.0, 1.0, 0.78 and 0.014 for T, B, REDOX, and PUREX plants, respectively (fraction of fuel activity routed to tank waste) as derived from process flowsheets. The reader needs to recognize possible inaccuracies in the tritium inventory associated with the use of these assumed split factors.
- ORIGEN2 code values for ^{14}C (Ci/MTU) are based on an assumed constant level of nitrogen impurity in uranium fuel cores of 20 ppm. In reality, the nitrogen level varied with time, ranging from highs of 90 ppm in the late 1940's to lows of about 8 ppm during the 1970's. A second simplifying assumption is made regarding losses of ^{14}C (to atmosphere) during fuel dissolution: it is assumed that 100 percent of ^{14}C in fuel is routed to waste tanks. (In actuality, 30 to 40 percent of the ^{14}C was measured leaving the plant in the dissolver offgas during PUREX campaigns from 1983 through 1987.) For lack of any historical operating data, the same 100 percent assumption was also applied to fuel processed through T, B, and REDOX separations plants.

Consequently, global inventories for ^{14}C given in Table 6.1-6 may be conservatively high.

- Activation products, ^{59}Ni and ^{63}Ni , are based on an assumed constant 800 ppm of Ni in aluminum cladding--an average value for all aluminum clad fuel types. (Beginning in 1959, fuel cladding (aluminum alloys) contained nickel levels of 9,000 to 13,000 ppm.) Thus, while HDW model results for global $^{59}\text{Ni}/^{63}\text{Ni}$ may be fairly representative of our total tank inventory, individual tank model results may be significantly distorted, being high in tanks filled before 1959 and low in tanks filled after 1959.
- The ORIGEN2 estimates for activation product, ^{60}Co , assume a cobalt impurity level of 10 ppm in cladding and 0 ppm in core uranium. Since actual levels of cobalt in uranium are unknown but probably greater than zero, it is predictable that the DKPRO output results for ^{60}Co are low.
- The ORIGEN2 code's prediction of ^{237}Np is based on an assumed constant level of 400 ppm ^{236}U in initial 0.947 percent enriched uranium input to the Hanford reactors. In actuality, the ^{236}U level varied from nearly zero to over 800 ppm in enriched uranium irradiated during the 1980's. This buildup of ^{236}U over time was the result of a closed fuel cycle, which in effect exposed the same uranium to multiple irradiation cycles. The ^{237}Np production can be expected to vary somewhat with the ^{236}U level, but a direct proportionality is not expected (due to the multiple neutron capture paths involved in ^{237}Np production). In addition, the efficiency of Np recovery in the REDOX and PUREX processes has been variable over the years and incompletely documented. Consequently, the global and tank-by-tank estimates for ^{237}Np contain a higher degree of uncertainty than do most other radionuclides.

One problem internal to the ORIGEN2 code is now known which affects the accuracy of curie values listed here for ^{79}Se :

- Curie values for ^{79}Se calculated by ORIGEN2 are suspected of being high by a factor of about 8 due to the use of an erroneous half-life value in the ORIGEN2 data library.

6.1.10 Tank-By-Tank Accuracy Issues

Radionuclide inventory values for individual tanks are predicted by the DKPRO/HDW model. Section 6.1.9, above, discusses some of the accuracy issues that are associated with the use of the DKPRO/HDW model for estimating global radionuclide inventory values. When using the DKPRO/HDW model for individual tank inventory values, another set of model assumptions, the solubility factors, come into play to add additional uncertainty to the inventory results. The degree of error between model results and actual tank contents can only be evaluated by comparison to actual core sample results.

To illustrate the degree of model error for individual tanks, scatter plots have been prepared in which available sample-based tank inventory values (for selected radionuclides) are plotted versus DKPRO/HDW model results for the same set of tanks. Inventory values for an additional set of approximately 20 tanks (derived from best basis engineering estimate methods) are also compared on the same plots. Each plot also contains a 1:1 correspondence line on which the scatter points should lie if there is no error in the model and no error in the measured value.

Comparisons for ^{90}Sr , ^{137}Cs , ^{239}Pu , and ^{241}Am are shown in Figures 6.1-4 through 6.1-7. Plots for ^{90}Sr and ^{137}Cs (Figures 6.1-4, and 6.1-5) are similar in that DKPRO/HDW model values greater than about 100,000 Ci per tank show a moderate correlation with measured or estimated values. The somewhat even distribution of points on either side of the 1:1 correspondence line also indicates that there is no major bias in either the model or measured values. However, for tanks containing less than about 100,000 Ci the scatter becomes much worse, indicating relative errors of several orders of magnitude are possible.

The comparison for ^{239}Pu inventory values (Figure 6.1-6) indicates a similarly weak to moderate correlation where errors of up to a factor of 40 may generally be possible with rare tanks being in error by several orders of magnitude.

The comparison for ^{241}Am (Figure 6.1-7) reveals a weak correlation and a strong indication that the analytical procedure for ^{241}Am may be biased high. It is unfortunate that model or sample accuracy for an important alpha emitter such as ^{241}Am cannot be characterized better than that indicated by Figure 6.1-7. Future work should be aimed at better understanding any shortcomings in the ^{241}Am analytical methods being used.

Scatter plot comparisons for ^{60}Co , ^{99}Tc , and ^{155}Eu have been analyzed but are not shown for reason of too few data points. The comparison for ^{60}Co , however, does show a strong indication that the DKPRO/HDW model is predicting low. This situation may be due to the fact that ORIGEN2 model runs lacked realistic setup values for Co impurities present in Hanford uranium fuel cores.

Figure 6.1-4. Model/Sample/Estimate Correlation for Strontium-90.

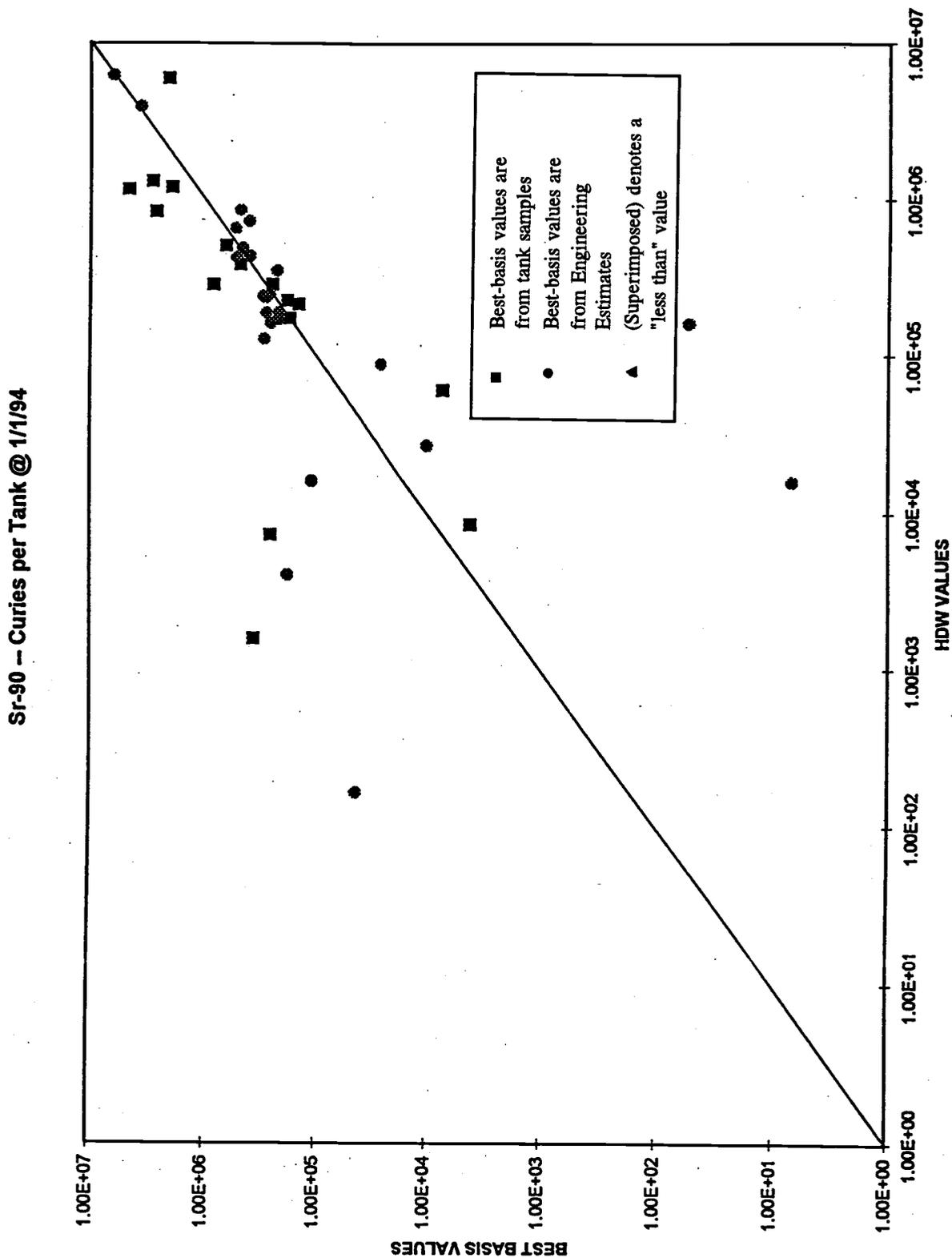


Figure 6.1-5. Model/Sample/Estimate Correlation for Cesium-137.

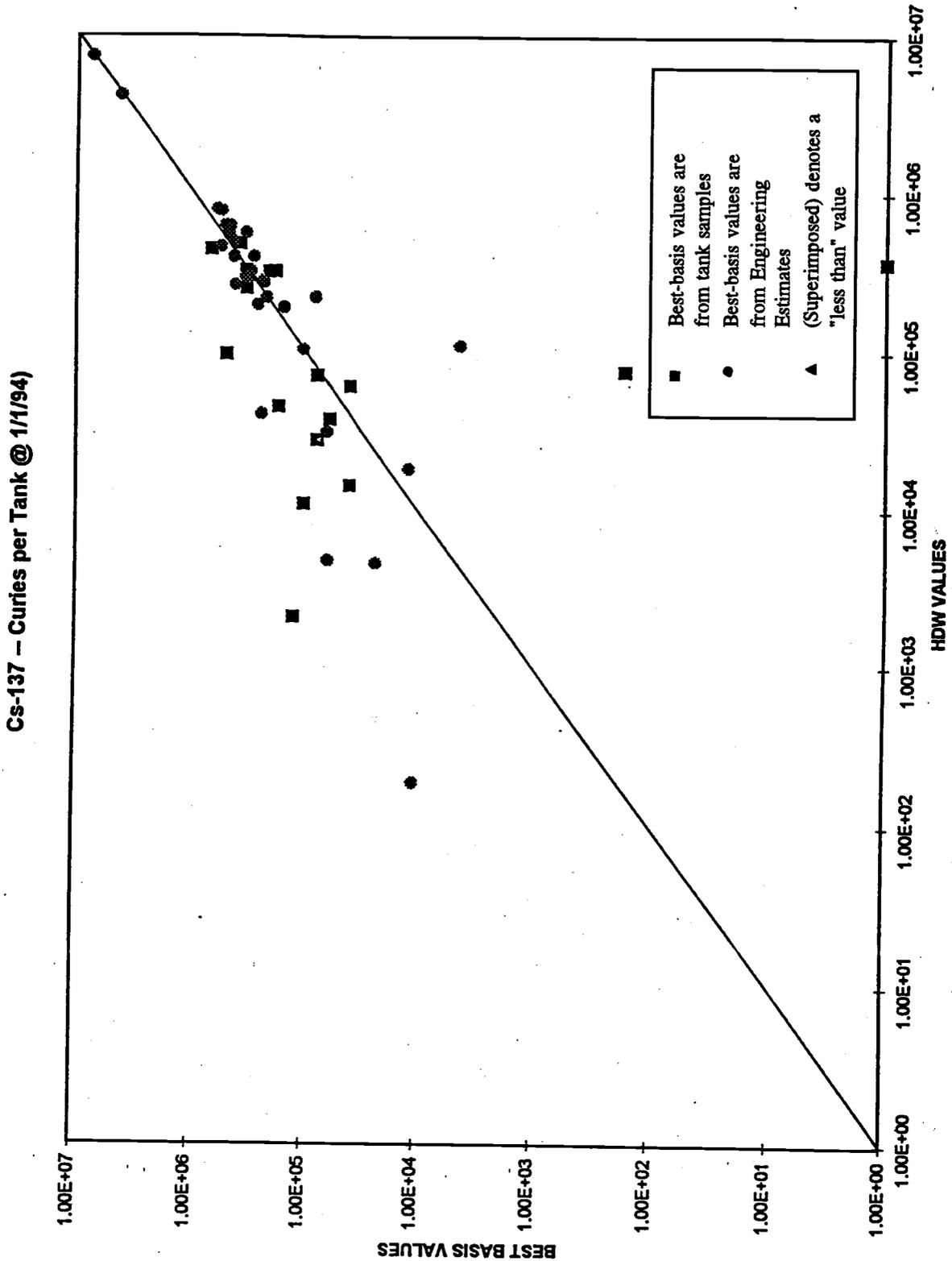


Figure 6.1-6. Model/Sample/Estimate Correlation for Plutonium-239.

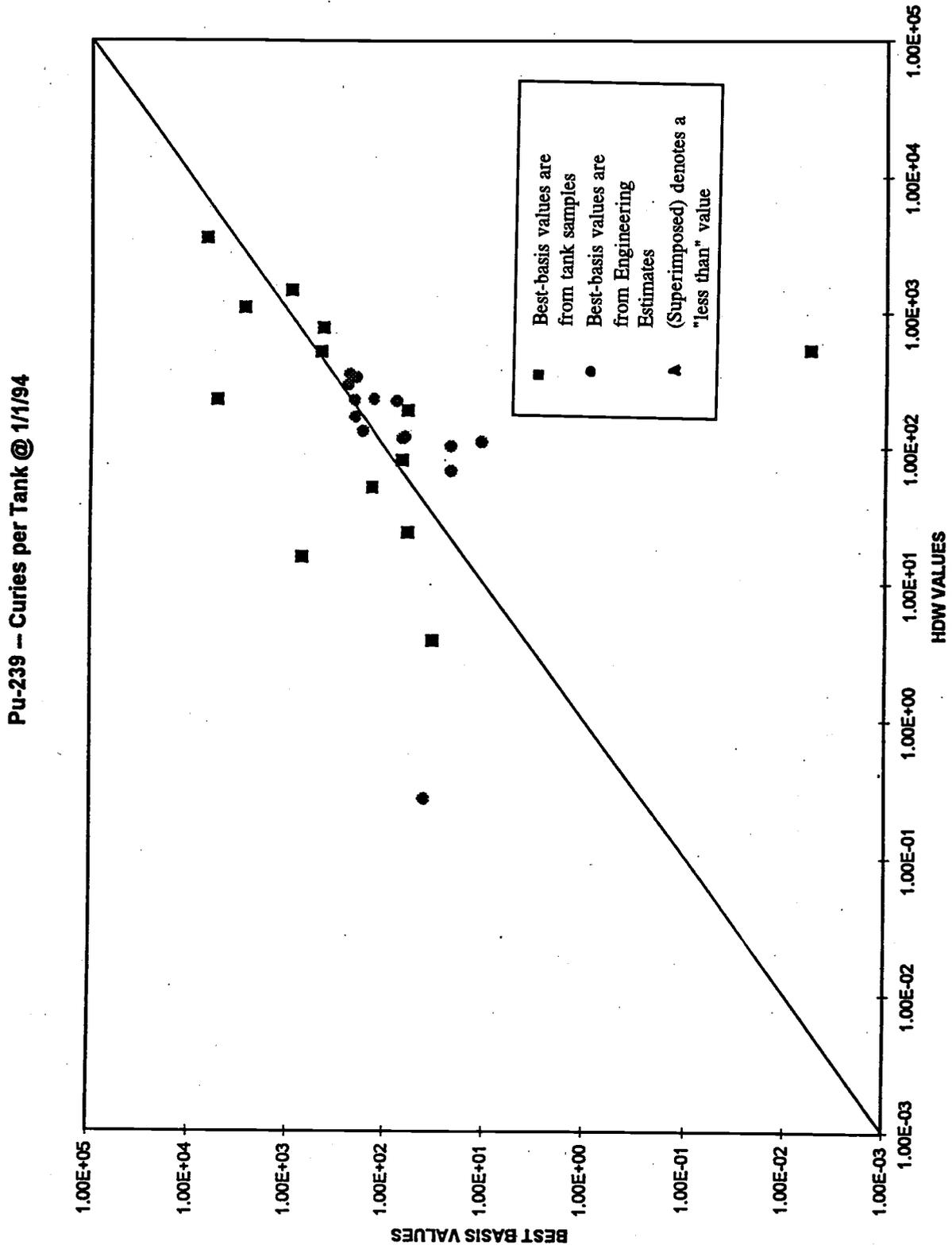
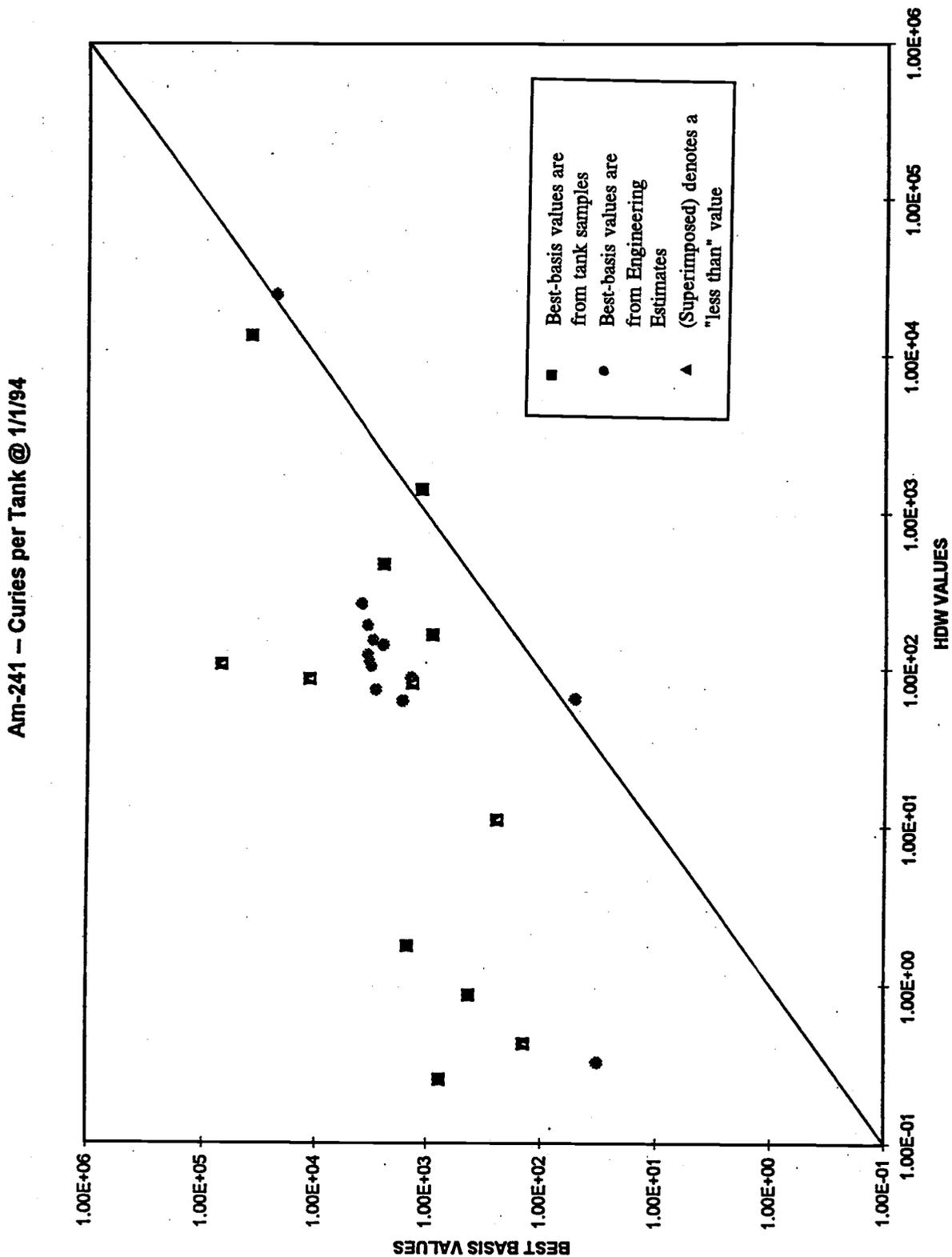


Figure 6.1-7. Model/Sample/Estimate Correlation for Americium-241.



6.2 CESIUM AND STRONTIUM

The two radionuclides, ^{137}Cs and ^{90}Sr , along with certain others, e.g., ^{99}Tc , ^{79}Se , and the actinide elements, are the most important radioactive constituents of Hanford Site tank wastes. Because of their approximate 30-year half lives, ^{137}Cs and ^{90}Sr contribute not only almost all the decay heat associated with the tank wastes, but also most of the radioactivity (curie) content of the tank wastes. Systems for disposing of retrieved Hanford Site tank wastes all involve removal of the bulk of the ^{137}Cs in such wastes. Removal of ^{90}Sr from some retrieved wastes may also be necessary or desirable to obtain LLW that can be disposed of in near-surface facilities. For economic reasons, i.e., costs involved in designing, constructing, and operating a ^{137}Cs removal facility, it is particularly important to establish a high-quality best estimate of the global inventory of ^{137}Cs in the 177 underground tanks at the Hanford Site.

The principal objective of the analyses and evaluations reported here is to estimate the global inventory of ^{137}Cs and ^{90}Sr currently in Hanford Site tanks. However, information was also obtained relating to the range of inventories of ^{137}Cs and ^{90}Sr that might eventually be transferred to the tanks as the result of future decontamination and decommissioning activities in the Hanford Site B Plant.

6.2.1 Identification and Comparison of Cesium-137 and Strontium-90 Inventory Values

Currently reported global inventories of ^{137}Cs and ^{90}Sr (from two data sources) in Hanford Site tanks are listed in Table 6.2-1. For comparison purposes, it is convenient to consider separately the amounts of these two radionuclides in DSTs and SSTs. A significant amount of analytical data for wastes in all the individual DSTs allows a reasonable estimate of the global inventory of ^{137}Cs and ^{90}Sr in the DSTs. Preliminary comparisons indicate that the DST global inventories of ^{137}Cs and ^{90}Sr established from analytical data differ by about 25 percent from inventories predicted by the HDW (Rev. 3) model. However, the SST inventory of ^{137}Cs predicted by the HDW (Rev. 3) model is over twice that reported in the present TWRS inventory. Also, the HDW model predicts the ^{90}Sr inventory of the SSTs to be about 25 percent less than that shown in the TWRS inventory. The following subsections describe new material balance calculations to establish SST inventories of ^{137}Cs and ^{90}Sr . Best-basis estimates of the global inventories of ^{137}Cs and ^{90}Sr are provided in Section 6.2.3.

Table 6.2-1. Currently Reported Inventory Values for Cesium-137 and Strontium-90 in Hanford Site Tanks.

Tanks	¹³⁷ Cs, MCi ^a		⁹⁰ Sr, MCi ^a	
	TWRS data	HDW (Rev. 3) model data ^b	TWRS data	HDW (Rev. 3) model data ^b
Double-shell tanks	25.4 ^c	20.2	11.4 ^c	16.3
Single-shell tanks	9.5 ^d	20.5	41.2 ^d	33.4
All	34.9	40.7	52.6	49.7

HDW = Hanford Defined Waste

TWRS = Tank Waste Remediation System

^aDecayed to January 1, 2000.

^bAgnew (1995), Agnew et al. (1996).

^cShelton (1995a) describes the sources and bases for the TWRS data for double-shell tanks and single-shell tanks.

^dShelton (1995a and b), Boldt et al. (1996).

6.2.2 Cesium-137 and Strontium-90 Inventories Evaluation

Historically, not all the ¹³⁷Cs and ⁹⁰Sr originally introduced into the Hanford Site tanks remained there. A large fraction of both radionuclides was removed from 1968 to 1978 and eventually encapsulated as ¹³⁷CsCl and ⁹⁰SrF₂, respectively. Other fractions were discharged to soil columns, sent to other DOE (then Atomic Energy Commission) facilities, or disposed of as solid waste. Still other fractions were removed from the underground tanks and then later reintroduced into the tanks. For all these material transactions, there is some degree of uncertainty concerning the amounts of ¹³⁷Cs and ⁹⁰Sr transferred. Propagation of these uncertainties complicates estimation of the current tank inventories of ¹³⁷Cs and ⁹⁰Sr.

6.2.2.1 Material Balance Considerations. One method for estimating the inventory of ¹³⁷Cs and ⁹⁰Sr in the SSTs is through material balance calculations. Such calculations thus allow estimation of the global inventory of radiostromium and radiocesium in all the tanks. Two key assumptions underlie the material balance approach to estimating the ¹³⁷Cs and ⁹⁰Sr content of the SSTs.

- The total amounts of ¹³⁷Cs and ⁹⁰Sr generated in Hanford Site reactors are known from computer code calculations.
- The ¹³⁷Cs and ⁹⁰Sr contents of the DSTs are known from analytical data.

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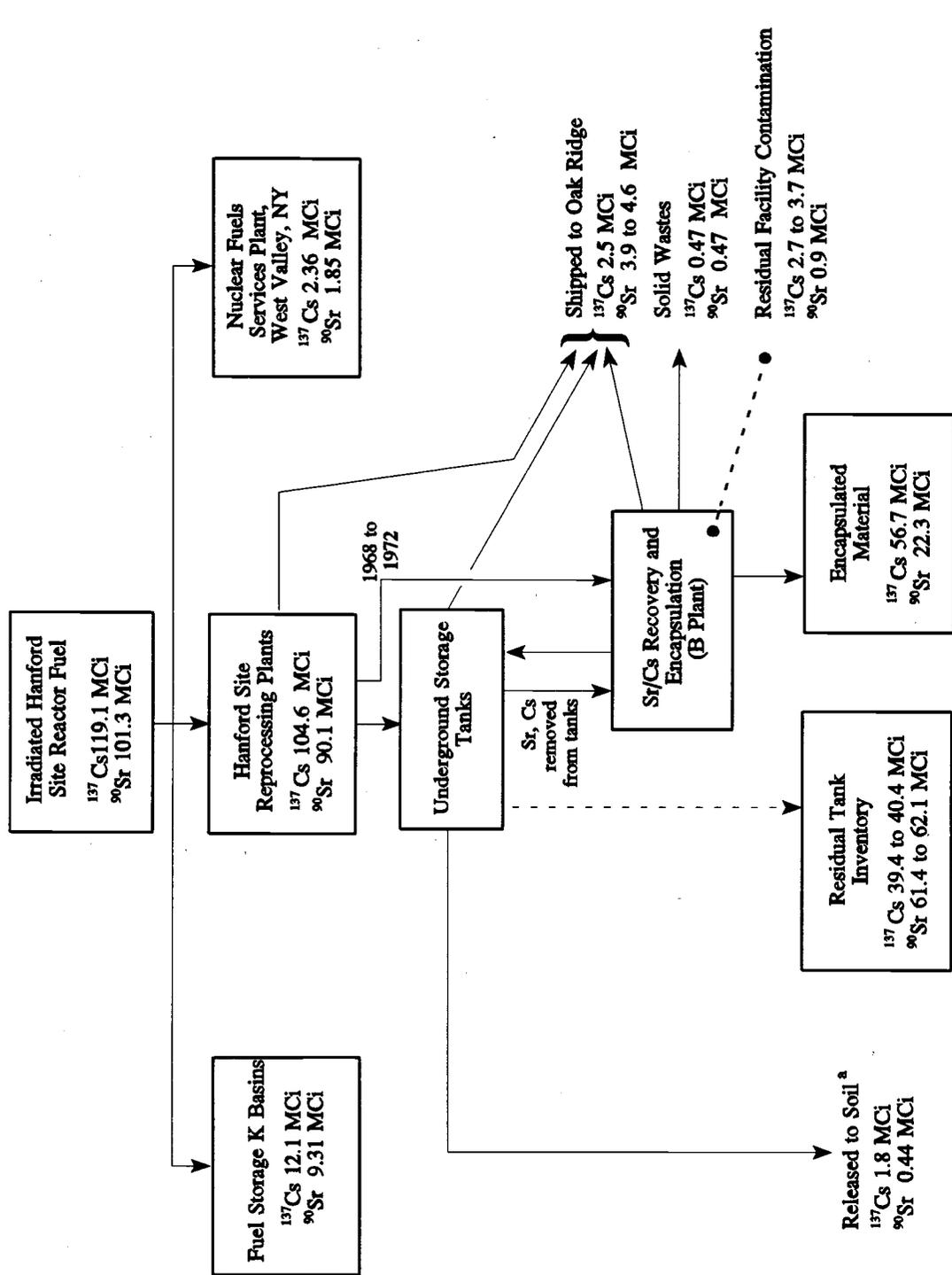
Figure 6.2-1 illustrates the various components of the Hanford Site ^{137}Cs and ^{90}Sr material balances. Table 6.2-2 summarizes best available values for certain of the material balance components cited in Figure 6.2-1. The accompanying Tables 6.2-3, 6.2-4, and 6.2-5 (and text) support information shown in Table 6.2-2 and also indicate sources of data. The validity and utility of global SST inventories of ^{137}Cs and ^{90}Sr derived from the material balance calculations presented in Table 6.2-2 are examined in a later section.

Estimates for the amounts of ^{137}Cs and ^{90}Sr in all fuel processed through the BiPO_4 , REDOX, and PUREX Plants from 1944 through 1989 have recently been generated by the DKPRO code (Schmittroth 1996), and alternatively by the RADNUC code in combination with the RIBD code. Schmittroth's results were obtained using the newly-developed computer code DKPRO (Version 1) which uses Ci/MTU values from the ORIGEN2 code. Outputs of the RIBD and RADNUC codes were combined to calculate the amounts of ^{137}Cs and ^{90}Sr in fuel reprocessed in the Hanford Site separations plants. The RIBD code is preferred to calculate production of ^{137}Cs and ^{90}Sr from 1944 to 1971 while the RADNUC code is preferred for the best estimate of the amounts of ^{137}Cs and ^{90}Sr generated from 1972 through 1989. Watrous and Wootan (1997) data are generated from a later version of the DKPRO code using data for single pass reactors specifically modelled by ORIGEN2. Results (activity in all fuel decayed to January 1, 2000) obtained by these codes are as follows:

^{137}Cs :	DKPRO (Version 1)	= 103.5 MCi
	RIBD/RADNUC	= 102 MCi
	DKPRO (1997)	= 104.6 MCi
^{90}Sr :	DKPRO (Version 1)	= 90.3 MCi
	RIBD/RADNUC	= 93.2 MCi
	DKPRO (1997)	= 90.1 MCi

These three sets of data agree within 3 percent. Agnew et al. (1996) notes that, from the slight differences in thermal fission yields and in half-lives, the inventory of ^{90}Sr after some 35 to 40 years of decay would be expected to be about 15 percent less than the inventory of ^{137}Cs . In the case of the DKPRO-1997 results, the ^{90}Sr inventory is 86 percent of the ^{137}Cs inventory. DKPRO-1997 results are selected for use in Figure 6.2-1 and Table 6.2-2. Data shown in Table 6.2-3 are used to compute the total quantities of ^{137}Cs and Sr^{90} listed in Figure 6.2-1 and Table 6.2-2.

Figure 6.2-1. Estimated Disposition of Cesium-137 and Strontium-90 Generated In Hanford Site Reactors.



All values are decayed to January 1, 2000

^a Includes amounts disposed of as solid waste and liquid waste as tank leaks and liquids sent to cribs and trenches.

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Previously, Agnew et al. (1996) estimated that 94 MCi of ^{137}Cs and 78.4 MCi of ^{90}Sr (decayed to January 1, 2000) were introduced into the Hanford Site tanks. Peterson (1996) recently reported that 97 MCi of ^{137}Cs and 82 MCi of ^{90}Sr were sent to the underground tanks. The DKPRO-1997 estimate of 90.1 MCi of ^{90}Sr is thus 8.1 to 11.7 MCi higher than the previous estimates. The DKPRO-1997 estimate of 104.6 MCi of ^{137}Cs is 7.6 to 10.6 MCi higher than the previous estimates. Because they are based on more accurate estimates of fuel irradiation history, ^{137}Cs and ^{90}Sr inventories derived by the DKPRO-1997 code are considered more reliable than prior computer code calculations.

Note that in the table (above), in Figure 6.2-1, and in all subsequent tables of this section (6.2), curie values for ^{90}Sr and ^{137}Cs are based on a decay date of January 1, 2000, which is different from the decay date basis (January 1, 1994) assumed in Tables 6.1-3 and 6.1-6. The January 1, 2000, decay date was chosen for Section 6.2 for ease of comparison to many existing data sources.

Table 6.2-2. Simplified Hanford Site Underground Tank Cesium-137 and Strontium-90 Material Balance.

Material balance component	MCi ^a	
	^{137}Cs	^{90}Sr
Generated in Hanford Site reactors	119.1	101.3
In fuel not reprocessed at the Hanford Site ^b	14.5	11.2
In fuel input to Hanford reprocessing plants	104.6	90.1
Output to capsules ^d	56.7	22.3
Output to other DOE facilities ^d	2.5	3.9 to 4.6
Output to solid waste disposal ^d	0.47	0.47
Output to facility contamination ^d	2.7 to 3.7	0.9
Output in releases to soil ^c	1.8	0.44
Total tank inventory remaining ^f	39.4 to 40.4	61.4 to 62.1
Inventory in double-shell tanks ^e	25.4	11.4
Inventory in single-shell tanks ^f	14.0 to 15.0	50.0 to 50.7

^aAs of January 1, 2000

^bSee Table 6.2-3

^cSee Table 6.2-4

^dSee Table 6.2-5

^eSee Table 6.2-1

^fCalculated value from material balance analysis.

6.2.2.2 Cesium-137 and Strontium-90 Not Reprocessed at the Hanford Site. Not all the fuel irradiated in Hanford Site reactors was reprocessed at the Hanford Site. As noted in Tables 6.2-2 and 6.2-3, part of the irradiated fuel was shipped to the Nuclear Fuels Services Company plant in West Valley, New York. Another part of the irradiated fuel is still stored in water basins at the K East and K West areas at the Hanford Site. The amounts of ^{137}Cs and ^{90}Sr , decayed to January 1, 2000, in the fuel not processed at the Hanford Site, are listed in Tables 6.2-2 and 6.2-3. These amounts of ^{137}Cs and ^{90}Sr were not introduced into the Hanford Site reprocessing plants.

Table 6.2-3. Inventory of Cesium-137 and Strontium-90 in Reactor Fuel Not Reprocessed at the Hanford Site.

Fuel disposition	Radionuclide content, MCi ^a	
	^{137}Cs	^{90}Sr
Shipped to Nuclear Fuel Services Co. Facility ^b	2.36	1.85
Stored in the Hanford Site K Reactor Basins ^c	12.1	9.31
Total	14.46 ^d	11.16 ^d

^aAs of January 1, 2000

^bBoldt et al. 1996, Myers 1995

^cBoldt et al. 1996, Willis and Praga 1995

^dRounded to 14.5 and 11.2, respectively, in Table 6.2-2.

Boldt et al. (1996) provides considerably more detail concerning fuel irradiated in Hanford Site reactors but not subsequently reprocessed at the Hanford Site. The ^{137}Cs and ^{90}Sr inventories shown in Table 6.2-3 are those cited by Boldt et al. (1996).

6.2.2.3 Cesium-137 and Strontium-90 Removed from Underground Tanks. Contrary to most other fission products, not all the ^{137}Cs and ^{90}Sr originally introduced into the underground Hanford tanks remained there. A major fraction (Table 6.2-4) of these two radioisotopes was transferred to the Hanford B Plant for recovery and subsequent conversion to $^{137}\text{CsCl}$ and $^{90}\text{SrF}_2$ and eventual encapsulation. During the period 1968 to 1972, acidic high-level wastes were transferred directly from the PUREX plant to B Plant. Other smaller fractions of ^{90}Sr and ^{137}Cs were separated directly from the PUREX Plant, the Strontium Semi-Works, and from tank farms.

Table 6.2-4. Inventory of Cesium-137 and Strontium-90 Removed from Tanks.

Transaction	MCI ^a	
	¹³⁷ Cs	⁹⁰ Sr
Released to soil	1.8	0.44
Sent to the Hanford Site B Plant	75.5	40.4
Total	77.3	40.8

^aAs of January 1, 2000.

Boldt et al. (1996) provides full details of the removal of ¹³⁷Cs and ⁹⁰Sr from the underground tanks to the underlying soil or to Oak Ridge National Laboratory. Data for the amounts of ¹³⁷Cs and ⁹⁰Sr (decayed to January 1, 2000) transferred to the Hanford Site B Plant are provided by a recent evaluation (Gehrke 1996).

B Plant ¹³⁷Cs and ⁹⁰Sr Material Balance Considerations. In one approach to calculating global inventories by material balance, the important data are the amounts of ¹³⁷Cs and ⁹⁰Sr transferred from tanks to B Plant and the amounts returned from B Plant to the underground tanks. Estimates for these input/output transfer values are given in Table 6.2-5.

Table 6.2-5. Material Balance for Cesium-137 and Strontium-90 in B Plant and Waste Encapsulation and Storage Facility^a.

Material balance component	MCI ^b	
	¹³⁷ Cs	⁹⁰ Sr
Input		
Transferred to B Plant	75.5	40.4
Output		
Encapsulated	56.7	22.3
Offsite Cask Shipments	2.5	3.9 to 4.6
Solid Waste Disposal	0.47	0.47
Returned to Tanks	1.8 to 11.0	0.9 to 5.5
Residual Facility Contamination	2.7 to 3.7	0.9
Total output	64.2 to 74.4 (85 to 99%) ^c	28.5 to 33.8 (70 to 84%) ^c

^aData from Gehrke (1996).

^bDecayed to January 1, 2000.

^cOf input to B Plant.

For completeness, Table 6.2-5 also lists the quantities of ^{137}Cs and ^{90}Sr (decayed to January 1, 2000) stored in an encapsulated form in the Hanford Waste Encapsulation and Storage Facility as well as material balance data for other fractions of radiocesium and radiostrontium transferred from and remaining in the B Plant. The information presented in Table 6.2-5 derives from a recent analysis by Gehrke (1995, 1996).

As indicated in Table 6.2-5, the exact amounts of ^{137}Cs and ^{90}Sr returned from B Plant to the underground tanks are very uncertain. Gehrke (1996) states, "The range is my estimate of 1 to 5 percent losses during 3 cycles of processing, based on the flowsheets used. The wide range reflects the appropriate uncertainty that exists for this stream. ...The waste loss estimates are probably no better than -50 to +200 percent."

Gehrke thinks that the uncertainty associated with other entries in Table 6.2-5 is less, e.g., +10 percent for encapsulated material, +20 percent for feed and cask shipments, and +50 percent for residual facility contamination. According to Gehrke, the information for the amounts of cesium and strontium in the feed to the B Plant came from a chart (author unknown) prepared in 1985 after shutdown of processing operations in B Plant. Note, also, that estimates for the sum of output streams from B Plant do not equate to the input stream estimates. Consequently, an estimate of ^{90}Sr and ^{137}Cs remaining in underground tanks that is calculated from estimates of "transfers to" and "returns from" the B Plant will contain significant uncertainty.

Fortunately, an alternative estimate of ^{90}Sr and ^{137}Cs remaining in underground tanks can be made by using estimates of other B Plant output streams (given in Table 6.2-5) in a material balance as itemized in Table 6.2-2 and illustrated in Figure 6.2-1. In this alternative, a system envelope is drawn around "Reprocessing plants," "Underground Storage Tanks," and "B Plant operations" (refer to Figure 6.2-1). This method allows the Residual Tank Inventory to be calculated by difference (using the more certain estimates for the "Encapsulated Inventory," and "Inventory Shipped to Oak Ridge") without having to account for the more uncertain "Removed from tanks" and "Returned to tanks" streams.

6.2.2.4 Comparison with Hanford Defined Waste Estimates. Table 6.2-6 shows a comparative accounting for ^{137}Cs and ^{90}Sr , as estimated by three models.

Table 6.2-6. Comparison of Cesium-137 and Strontium-90 Accounting by Model.

	¹³⁷ Cs decayed to January 1, 2000 (MCi) ^a		
	HDW model (Rev. 3) ^b	DKPRO/HDW model (Rev. 4) ^c	DKPRO/SPLIT model ^d (1997)
In reprocessed fuel	94	104.6	104.6
Released to soil	1.3	1.2	1.8
Shipped to other facilities ^e	0	2.5	2.5
Solid waste	0	0	0.5
Residual B Plant contamination	0	3.2	2.7-3.7
Encapsulated	53	57.5	56.7
Remaining in tanks	40.7	41.0	40.4-39.4 ^f
Site accounted	95.0	105.4	104.6
Site unaccounted	-1.0	-0.8	0
	⁹⁰ Sr Decayed to January 1, 2000 (MCi) ^a		
In reprocessed fuel	78	90.1	90.1
Released to soil	0.6	0.97	0.4
Shipped to other facilities ^e	0	4.3	3.9-4.6
Solid waste	0	0	0.5
Residual B Plant contamination	0	0.9	0.9
Encapsulated	23	22.5	22.3
Remaining in tanks	49.7	53.3	62.1-61.4 ^g
Site accounted	73.3	82.0	90.1
Site unaccounted	+4.7	+8.1	0

HDW = Hanford Defined Waste

^aAll decayed to January 1, 2000

^bAgnew et al. 1996. Agnew provided a table showing the ¹³⁷Cs and ⁹⁰Sr accounting used in Revision 3 of the HDW model.

^cAgnew et al. (1997a)

^dData taken from Tables 6.2-2 through 6.2-5 of this report

^eOak Ridge National Laboratory and Pacific Northwest National Laboratory.

^fCalculated by material balance (Fuel - releases and products). Range corresponds to "Residual B Plant" range.

^gCalculated by material balance (Fuel - releases and products). Range corresponds to "other facilities" range.

It is convenient to consider ^{137}Cs data in Table 6.2-6 separately from that for ^{90}Sr . The following differences between the three model predictions are noticeable.

- The HDW Rev. 3 estimate of the amount of ^{137}Cs in the waste discharged to the Hanford Site tanks is 10.6 MCi less than the value estimated by the DKPRO-1997 code. The HDW estimate apparently derives from data provided by the TRAC model (Jungfleish 1984) while the data used in the later models is from the DKPRO-1997 code (Watrous and Wootan 1997).
- The HDW Rev. 3 estimate of the amount of encapsulated ^{137}Cs (decayed to January 1, 2000) is approximately 4 MCi less than the values used in later models. The DKPRO/SPLIT model value derives from the recent memorandum report by Gehrke (1996).
- The HDW Rev. 3 model does not take into account ^{137}Cs transferred to other sites, solid waste, or residuals remaining in facilities. This omission coupled with a low input value (94 MCi in waste sent to tanks) results in the models prediction of 40.7 MCi ^{137}Cs remaining in tanks--a value which is coincidentally close to the prediction of the later models.
- Comparison of the later models (DKPRO/HDW (Rev. 4) versus DKPRO/SPLIT) indicates close agreement in nearly all transaction values. The DKPRO/HDW (Rev. 4) model does predict 0.6 MCi more ^{137}Cs as remaining in tanks than does the DKPRO/SPLIT model--a relative difference of only 1.5 percent.

For ^{90}Sr global inventories (see Table 6.2-6), the HDW Rev. 3 model predicts only 78 MCi of ^{90}Sr were added to the tanks as a result of fuel reprocessing, whereas the later DKPRO/HDW and DKPRO/SPLIT models predict 90.1 MCi. Other highlights for the ^{90}Sr global inventories are as follows:

- No account is taken in the HDW Rev. 3 model of ^{90}Sr known to have been shipped to other facilities, ^{90}Sr disposed in solid waste or that remaining in facility residues.
- Table 6.2-6 shows 4.7 MCi of ^{90}Sr are unaccounted for in the HDW Rev. 3 model predictions. All or most of the missing ^{90}Sr likely represent material sent to the Oak Ridge and the Pacific Northwest National Laboratories.
- Accounting values used in the later models are approximately the same, with the exception that the DKPRO/HDW (Rev. 4) model predicts about 8.8 MCi less ^{90}Sr remaining in tanks than does the DKPRO/SPLIT model (53.3 MCi versus 62.1 MCi). This difference also shows up as 8.1 MCi being unaccounted for in the DKPRO/HDW (Rev. 4) model's balance. It appears that this imbalance is related to the problem of "missing ^{90}Sr " discussed in Agnew et al. (1997a).

6.2.3 Best-Basis for Cesium-137 and Strontium-90 Global Inventory Values

Four different estimates of the total inventory of ^{137}Cs and ^{90}Sr in DSTs and SSTs are compared in Table 6.2-7.

Table 6.2-7. Comparison of Tank Inventory Estimates for Cesium-137 and Strontium-90.

	^{137}Cs decayed to January 1, 2000 (MCi)			
	TWRS data	HDW Rev. 3	DKPRO/HDW (Rev. 4)	DKPRO/SPLIT model
DST inventory	25.4	20.2	20.3	25.4
SST inventory	9.5	20.5	20.7	14.0 to 15.0
Total inventory	34.9	40.7	41.0	39.4 to 40.4
	^{90}Sr decayed to January 1, 2000 (MCi)			
DST inventory	11.4	16.3	15.2	11.4
SST inventory	41.2	33.4	38.1	50.0 to 50.7
Total inventory	52.6	49.7	53.3	61.4 to 62.1

DST = Double-shell tank
 HDW = Hanford Defined Waste
 SST = Single-shell tank
 TWRS = Tank Waste Remediation System.

The analysis in Table 6.2-6 indicates that about 8 MCi of ^{90}Sr are not accounted for in the HDW Rev 4 model's inventory balance. To correct this imbalance in Table 6.2-7, the model's estimate of 38.1 MCi ^{90}Sr in SSTs would need to be increased to 46.9 MCi to bring the all-tank total to the value of 62.1 MCi. Even then the ratio of ^{90}Sr between single shell and double shell tanks would be inconsistent with the sample-derived estimate of the DKPRO/SPLIT model. (Inconsistencies in the SST/DST ratio are presumably due to shortcomings of solubility functions within the HDW model.) For these reasons, the results of the DKPRO/SPLIT model are used as the best basis global inventory.

Table 6.2-8 summarizes current best-basis estimates of the inventories of ^{137}Cs and ^{90}Sr in the DSTs and SSTs as well as the global inventories of these radionuclides in all the Hanford Site tanks. The inventories, as of January 1, 2000, of ^{137}Cs and ^{90}Sr in the 28 DSTs are established, from analytical data, as 25.4 and 11.4 MCi, respectively. Future activities, i.e., more analytical data for ^{137}Cs and ^{90}Sr contents of the SSTs and validated modeling predictions, will likely result in some revision to the best-basis global inventory data shown in Table 6.2-8.

Table 6.2-8. Global Best-Basis Estimates of Cesium-137 and Strontium-90 Inventories in Hanford Underground Tanks.

Tanks	MCi ^a	
	¹³⁷ Cs	⁹⁰ Sr
Double-shell tanks ^b	25.4	11.4
Single-shell tanks	15.0	50.7
Total	40.4	62.1

^aDecayed to January 1, 2000.

^bAnalytical data.

The best-basis global inventory (Table 6.2-8) of ¹³⁷Cs in the Hanford Site SSTs is 15.0 MCi versus the 9.5 MCi value presently carried in the TWRS estimate. This additional 5.5 MCi of ¹³⁷Cs results as a consequence of an increase of 10.6 MCi in estimated fuel activity offset by a more complete accountability for ¹³⁷Cs losses or transfers to other facilities, solid waste and residual facility contamination.

The best-basis global inventory of ⁹⁰Sr is 62.1 MCi (decayed to January 1, 2000). Of this total, 11.4 MCi are in the DSTs and 50.7 MCi in the SSTs. The former value is identical to that in the present TWRS database and derived from analyses of wastes in each of the 28 DSTs. The best-basis global inventory, 50.7 MCi listed in Table 6.2-8 for the SSTs is 9.5 MCi more than the 41.2 MCi carried in the present TWRS database. The additional ⁹⁰Sr originally introduced into the Hanford Site tanks is discussed in this report (see Table 6.2-2).

Best-basis global inventory values given in Table 6.2-8 (40.4 MCi of ¹³⁷Cs and 62.1 MCi of ⁹⁰Sr at a decay date of January 1, 2000) are equivalent with values given in Table 6.1-6 (46.4 MCi and 71.6 MCi, respectively), allowing for the six year difference in decay date bases.

6.3 URANIUM

Another radionuclide present in tank waste is uranium--significant because of its long half-life and importance to the acceptability of near-surface waste disposal alternatives, and because it is a major component in total waste oxides. Waste core samples have not all been analyzed for uranium isotopic makeup or even for total uranium mass. Therefore, we need to calculate inventories for many of the tanks based on historical fuel throughput and separations plant operating data.

The modelling prediction of uranium inventories in tank wastes is somewhat more uncertain than for fission products since U is one of the several "extractable" fuel components--meaning that another piece of information, the U separation factor, is needed.

The following sections discuss (1) the fuel sources and separation plant history contributing to the tank waste inventory of uranium, and (2) a material balance for U inventory data.

6.3.1 Uranium Fuel Sources and Separation History

From 1944 through 1971 aluminum clad fuels were processed through three separations processes (BiPO₄, REDOX, and PUREX). In the BiPO₄ process (T and B Plant operations, 1944 through 1956), 100 percent of the uranium in fuels was routed to tank waste and later recovered in a sludge sluicing operation. Recovered sludge was then dissolved and processed through a TBP solvent extraction process for uranium separation and purification, with a low percentage loss (possibly 1 to 2 percent) of uranium being returned from the TBP process to the tanks. However, it is the somewhat larger fraction of uranium bearing sludge that was not recovered in sluicing operations (typically about 5 percent) that contributes to the bulk of the tank U inventory from BiPO₄ operations. Consequently, our knowledge of this portion of the global tank uranium inventory is uncertain to the degree that historical records accurately describe the actual quantities of sludge remaining in tanks and the uranium concentration of those sludges.

Aluminum clad fuels plus a small quantity of zirconium clad fuels were processed through the REDOX and PUREX plants from 1952 through 1972. For the aluminum clad fuel these separation processes recovered a high percentage of the uranium, sending typically less than one percent to the combined cladding waste and extraction waste streams. Our knowledge of the resulting tank uranium inventory contribution is based on process waste analyses as reported through nuclear material accountability records. The quality of these waste sample analyses is thought to be relatively good since (unlike the case of zirconium clad fuel) acidic extraction waste streams contained few solids (i.e., most of the uranium in the waste stream was in solution and thus was amenable to a representative sample and analysis). Fortunately, uranium in the alkaline cladding waste solutions, while not easily measured, was a small part of the total uranium waste loss.

It should be noted that during 1966 and again in 1970, Thoria target elements were also processed through the PUREX plant for the recovery of ²³³U. Waste streams from these two processing campaigns were routed to C-farm tanks and are responsible for contributions of ²³³U and ²³²U to the global uranium inventory.

Zirconium clad fuels processing in the PUREX plant during the post 1971 time period suffered greater uranium waste losses due to (1) the attack of decladding solutions on fuel core uranium (up to 6 percent of the uranium was observed to react), and (2) the inefficiency of plant centrifuges to recover uranium solids from the decladding streams before their

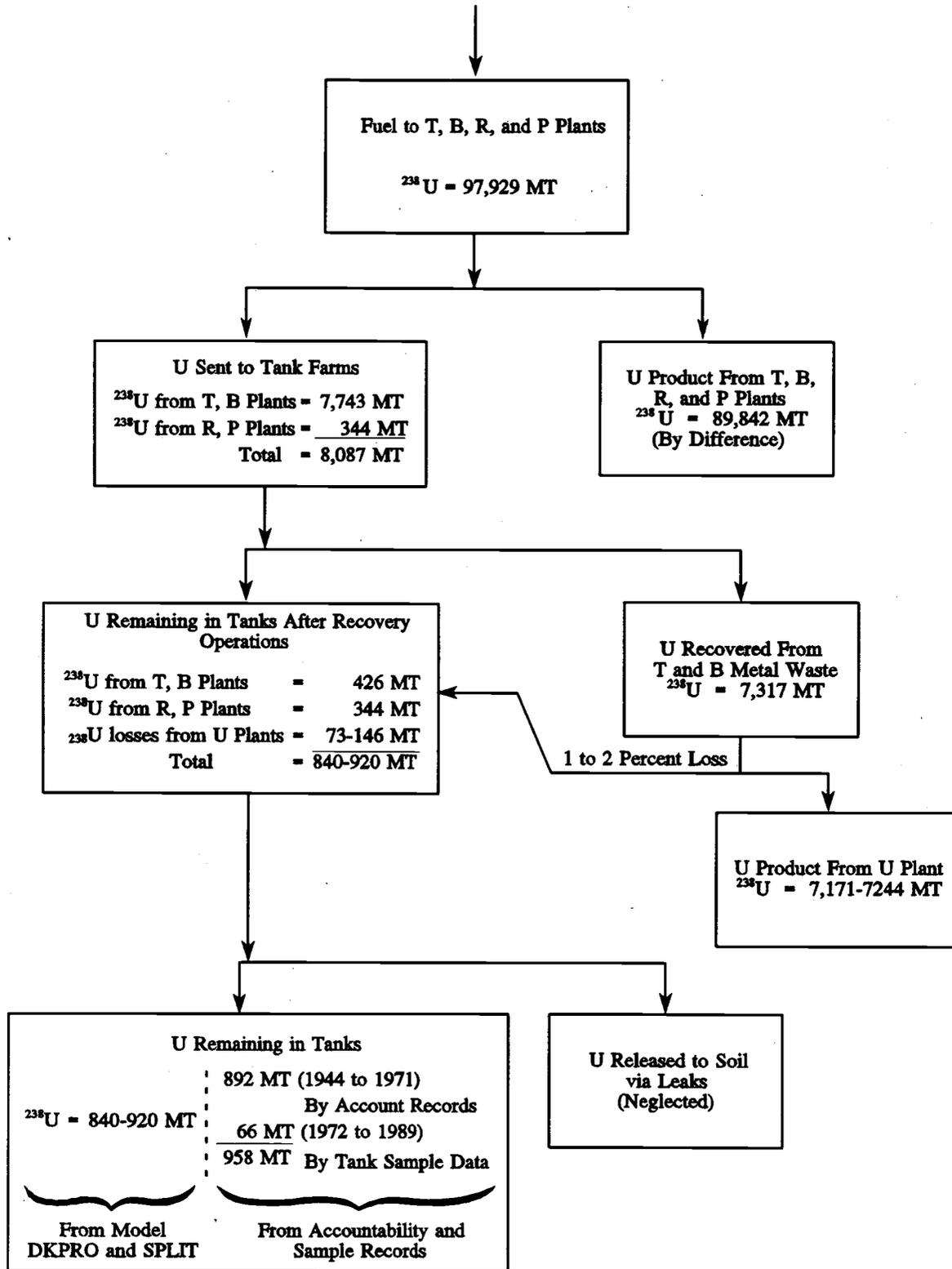
transfer to underground waste tanks. Measurement of U losses in Zirflex decladding streams was also uncertain due to the difficulty in getting a representative sample of the solid/liquid slurry. Likewise, the measurement of U in extraction wastes was also believed to be biased low due to the presence of U bearing solids in the waste. Uranium losses to post-1971 zirconium cladding waste streams and extraction waste streams, fortunately, have been contained in only four tanks (241-AW-103, 241-AW-105, 241-AZ-101, and 241-AZ-102). All four of these tanks have been core sampled with results indicating that, indeed, uranium inventory values are significantly greater than would be indicated by plant waste stream accountability records. Thus, for waste from zirconium clad fuel processing, an alternative and more representative inventory data source is available in core sample results for these four tanks.

6.3.2 Calculation of Global Uranium Inventory

The material balance for uranium, from fuels to tank residuals, is summarized by Figure 6.3-1 in units of metric tons of ^{238}U . Of 97,929 MT ^{238}U in fuels processed through T, B, REDOX, and PUREX plants (based on fuel activity records given in Watrous and Wootan [1997]), Figure 6.3-1 indicates that 7,743 MT ^{238}U were sent to tanks from T/B Plant operations while 344 MT ^{238}U were sent to tanks from REDOX/PUREX operations--this latter U loss value being based on REDOX/PUREX monthly average waste loss factors which are used in the DKPRO/SPLIT model. The material balance then accounts for an assumed 94.5 percent of the T/B uranium being recovered (factor derived from McDonald (1959), and used in the DKPRO/SPLIT model), resulting in an estimate of 7317 MT ^{238}U being sent to Uranium Recovery (UR) extraction operations at U Plant. Uranium losses from UR operations are estimated to have been 1 to 2 percent, which equates to 73 to 146 MT returned to tanks. Residual ^{238}U left in tanks from T/B operations is then estimated as $(7,743 - 7,317 = 426)$ MT. This combined with the UR waste losses (73 - 146 MT) plus the 344 MT sent to tanks from REDOX/PUREX operations sums to a total ^{238}U tank inventory of 840 to 920 MT for the assumptions used in the DKPRO/SPLIT model.

Figure 6.3-1 alternatively shows the ^{238}U tank inventory as derived from accountability records plus recent tank core samples (also see Table 6.1-4). Here, accountability records for operations 1944 through 1971 (as reported in ERDA 1977) indicate that 892 MT ^{238}U remain in all tanks after accounting for recovery of T/B uranium and measured waste losses from REDOX and PUREX. This alternative inventory method then estimates ^{238}U losses from PUREX operations (1983 through 1989) from core sample results for tanks 241-AZ-101, 241-AZ-102, 241-AW-103, and 241-AW-105 (66 MT ^{238}U), recognizing that core sample results are likely more representative than waste stream samples measured during plant operations. The alternative method then estimates the total ^{238}U tank inventory as the sum of these two sub-inventories $(892 + 66 = 958)$ MT ^{238}U .

Figure 6.3-1. Estimated Disposition of ^{238}U Processed Through Hanford Separations Plants.



revelmain/FIG6-3-1.WPG

6.3.3 Best-Basis for Uranium-238 Global Inventory Value

As discussed in Section 6.3.2, the 958 MT ^{238}U value derived from accountability and tank sample data sources is judged to be the preferred best-basis value. This value for ^{238}U is equivalent to a total uranium tank inventory of 965 MTU as derived in Table 6.1-4.

7.0 MINOR ELEMENTS AND CHEMICALS

Minor elements and chemicals were introduced into the tanks by several pathways. These pathways include, but are not limited to, the following:

- Fuel fabrication
- Reactor fission and activation products
- Chemical process operations
- Process chemical impurities
- Direct addition by tank farms.

7.1 MINOR ELEMENTS INTRODUCED BY FUEL FABRICATION

Approximately 99,000 MTU were processed by various separation facilities at Hanford. Details of the inventory are provided in Appendix B. This value includes the fuel processed by the PUREX Plant between 1983 and 1989. Fuel elements were clad in either aluminum or zirconium to protect the uranium from corrosion. Zirconium-clad fuel was used in N Reactor exclusively, whereas aluminum-clad fuel was used in all the older single-pass reactors.

Fuel fabrication methods, as well as fuel dimensions, changed over the years to save costs, and in response to increasingly severe reactor conditions. A discussion of the fuel fabrication methods, fuel types and quantities of the various types processed is provided in Section 4.2.

7.1.1 Aluminum-Clad Fuel

Table 7.1-1 summarizes the minor elements introduced into the process facility waste by aluminum-clad fuel fabrication. The estimates reflect the fuel quantities discussed in Section 4.2, i.e., 93,200 MTU of aluminum-clad fuel processed, of which 10,800 was triple dip and 82,400 was lead dip fuel.

Table 7.1-1. Elements Introduced by Fabrication of Aluminum-Clad Fuel.

Element	Uranium core (MT)	Intermetallic layer (MT)	Al-Si bond layer (MT)	Aluminum jacket (MT)	Total (MT)
B	--	--	≤0.01	≤0.03	≤0.04
Bi	--	--	≤0.1	≤0.25	≤0.4
Cd	--	--	≤0.03	≤0.08	≤0.1
Co	--	--	≤0.01	≤0.03	≤0.04
Cr	≤6.06	--	≤0.55	≤0.76	≤7.4
Cu	--	≤6.63	≤1.1	≤3.8	≤12
Fe	≤14.0	--	≤10.9	≤14.5	≤39
Li	--	--	≤0.08	≤0.2	≤0.3
Mg	≤2.33	--	≤0.1	≤0.25	≤2.7
Mn	≤2.33	--	≤0.29	≤0.76	≤3.4
Ni	≤9.3	--	≤0.33	≤20.1	≤30
Pb	--	≤213	≤1.94	≤0.25	≤220
Sn	--	≤14	≤0.1	≤0.25	≤14
Ti	--	--	≤0.22	≤1.3	≤1.5
Zn	--	--	≤0.29	≤0.76	≤1.1

7.1.1.1 Elements from Uranium Core Metal. Weakley (1958) provides maximum impurity specifications for uranium core metal. These specifications and an estimate based on fabrication and processing of 93,200 MTU of aluminum-clad fuel are shown in Table 7.1-2. The estimate is based on maximum impurity specifications and should be considered to be bounding.

Table 7.1-2. Elements Introduced by Uranium Metal Used in Aluminum-Clad Fuel.

Element	Maximum impurity ($\mu\text{g/g}$)	Maximum inventory (MT)
Cr	65	≤ 6.06
Fe	150	≤ 14.0
Mg	25	≤ 2.33
Mn	25	≤ 2.33
Ni	100	≤ 9.32
Si	75	Compiled in Section 5.19, Silicon

7.1.1.2 Elements in the Intermetallic Layer. The intermetallic layer is assumed to be either bronze or lead. The presence of intermetallic compounds of U-Pb, are not considered. The amount of intermetallic layer is calculated from the thickness of the layer and the surface area of the uranium cores. Elements attributable to the intermetallic layers are listed in Table 7.1-1.

The lead dip process left a layer of lead and U-Pb compounds between the uranium core and the Al-Si bond layer. The U-Pb compounds are UPb and UPb₃. The lead layer was about 0.0025-cm thick (Kahle and Bement 1958). Approximately 82,400 MT of lead dip aluminum-clad fuel was processed. The quantity of lead introduced by the lead dip process is estimated at 213 MT. If the reaction layer is dominated by UPb, the Pb inventory may be only 100 MT.

The exact thickness and composition on the bronze/tin interface layer between the uranium core and the aluminum cladding from the triple dip process is unknown. However if bronze/tin intermetallic layer is assumed to be the same thickness as lead dip intermetallic layer, then it is possible to make an estimate of copper and tin introduced into the tank waste. The maximum amount of copper introduced by this source would be 6.6 MT based on an intermetallic layer with the composition of the bronze dip. The tin inventory would range from 7.5 MT of tin, if the intermetallic layer was essentially bronze, to 14 MT tin, if the bronze layer was essentially displaced by the tin bath. The largest uncertainty in this estimate is the thickness of the intermetallic layer.

7.1.1.3 Elements in the Al-Si Bond Layer. The composition of the Al-Si bath varied with time due to dragout and makeup. Through use, the bath is enriched in aluminum due to erosion/corrosion of the aluminum cans. Virgin Al-Si, lathe turnings and silicon were all added to the bath as makeup material and to adjust the silicon content. The baths were sampled weekly for impurities. Initially strict impurity limits were relaxed to the point that

the baths were not changed unless an unusual contamination problem occurred. Impurities quickly built up to saturation values (Weakley 1958). Table 7.1-3 compiles an estimate of the impurities in the Al-Si braze layer based on saturation values, virgin Al-Si and virgin aluminum jacket stock.

Table 7.1-3. Elements Introduced by the Aluminum-Silicon Bond Layer.

Element	Saturation limit of impurity (wt%)	Maximum inventory (MT)
Fe	1.00	≤10.9
Pb	0.20	≤1.94
Cr	0.05	≤0.55
Ni	0.03	≤0.33
Ti	0.02	≤0.22
Virgin Al-Si Braze Alloy Impurity Specification		
Cu	0.10	≤1.1
Aluminum Jacket Alloy Impurity Specification		
Mn	0.03	≤0.29
Zn	0.03	≤0.29
Mg	0.01	≤0.10
Sn	0.01	≤0.10
Bi	0.01	≤0.1
Li	0.008	≤0.08
Cd	0.003	≤0.03
Co	0.001	≤0.01
B	0.001	≤0.01

7.1.1.4 Elements in the Aluminum Jacket. At least two types of aluminum alloy were used for the fuel element jacket: the 1245 (C-64) alloy was introduced in 1944 and the M-388 (X-8001) alloy was introduced in mid 1957 (Weakley 1958). Full scale use of the X-8001 alloy by Fuel Fabrication was in effect in December 1960 (DeNeal 1970). An estimate of the impurities from the aluminum jacket is provided in Table 7.1-4. The basis for the estimate is 93,200 MT of aluminum-clad fuel. With the exception of nickel, the estimate is based on the alloy with the larger impurity specification. However, since there is

a large difference in the nickel specification of the two alloys, it is assumed that the 1245 alloy was used on fuel processed through the end of 1958 and the M-388 alloy was used on fuel processed after that date. The aluminum in the Al-Si bond layer is, as previously noted, expected to have the same impurities as the jacket material since the aluminum content of the Al-Si bath needed to be routinely supplemented with silicon as the canning process tended to erode the aluminum jacket material.

Table 7.1-4. Minor Elements Introduced by the Aluminum Jacket Alloy.

Element	Aluminum alloy		Maximum inventory (MT)
	1245 (wt%)	M-388 (wt%)	
Ni	0.01	0.9-1.3	≤20.1
Fe	0.35 - 0.50	0.45 - 0.77	≤14.5
Si	0.17	0.17	Compiled in Section 5.19, Silicon
Ti	0.05	--	≤1.3
Cu	0.04	0.15	≤3.8
Mn	0.03	--	≤0.76
Cr	0.03	--	≤0.76
Zn	0.03	--	≤0.76
Mg	0.01	--	≤0.25
Sn	0.01	--	≤0.25
Pb	0.01	--	≤0.25
Bi	0.01	--	≤0.25
Li	0.008	0.008	≤0.20
Cd	0.003	0.003	≤0.08
Co	0.001	0.001	≤0.03
B	0.001	0.001	≤0.03

7.1.2 Zirconium-Clad Fuel

Fuel elements for N Reactor consisted of assemblies of two concentric uranium tubes. Each uranium element was co-extruded into Zircaloy-2[®] cladding. Each assembly was made up of an inner and outer element. The REDOX Plant processed 8.0 MT N Reactor fuel in 1965 and 220 MT in 1966. The REDOX Plant also processed 5.0 MT of PRTR fuel and 12.4 MT of Shippingport fuel (Jenkins and Foster 1978). The PUREX Plant processed 1,572 MT of N Reactor fuel in the 1967 to 1972 time period, and 3,890.4 MT in the 1983 to 1989 campaign. Table 7.1-5 summarizes the elements introduced from fabrication of N Reactor zirconium-clad fuel. Minor component specifications are not available for the Shippingport Reactor and PRTR fuel.

Table 7.1-5. Elements Introduced by Fabrication of Zirconium-Clad Fuel.

Element	Uranium alloy 601 (MT)	Zircaloy-2 [®] cladding (MT)	Braze filler (MT)	Total (MT)
Al	4.6	≤0.03	≤0.001	4.6
B	≤0.0014	≤0.0002	0.0	≤0.0016
Be	≤0.057	--	0.39	0.45
Cd	≤0.0014	≤0.0002	0.0	≤0.0016
Co	--	≤0.0042	≤0.0002	≤0.004
Cr	≤0.37	0.42	0.008	0.80
Cu	≤0.43	≤0.021	≤0.0005	≤0.45
Fe	2.0	0.57	0.001	2.6
Hf	--	≤0.084	≤0.0016	≤0.086
Mg	≤0.14	≤0.0084	≤0.0005	≤0.15
Mn	≤0.14	≤0.021	≤0.0004	≤0.16
Mo	--	≤0.021	≤0.0004	≤0.021
Na	--	≤0.0084	≤0.0	≤0.0084
Ni	≤0.57	0.23	≤0.014	≤0.81
Pb	--	≤0.042	≤0.001	≤0.04
Sn	--	6.1	≤0.11	6.2
Ti	--	≤0.021	≤0.0002	≤0.021
U	--	≤0.001	0.0	≤0.001
V	--	≤0.021	≤0.0004	≤0.021
W	--	≤0.021	≤0.0008	≤0.022
Zr	≤0.37	413	7.3	421

7.1.2.1 Elements in Uranium 601 Alloy. Rockwell (RHO 1980) provides impurity specifications for the uranium 601 alloy used in N Reactor fuel. These specifications and an estimate based on fabrication and processing of 5,690 MTU are shown in Table 7.1-6. The estimate is based on the mid-range or maximum impurity specifications as available.

Table 7.1-6. Elements Introduced by Uranium 601 Alloy.

Element	Specification ($\mu\text{g/g}$)	Inventory (MT)
Al	700-900	4.6
Be	10	≤ 0.057
Fe	300-400	2.0
Cr	65	≤ 0.37
Ni	100	≤ 0.57
C	365-735	3.1
Zr	65	≤ 0.37
B	0.25	≤ 0.0014
Cd	0.25	≤ 0.0014
Cu	75	≤ 0.43
Mg	25	≤ 0.14
Mn	25	≤ 0.14
N	75	≤ 0.43
Si	124	Compiled in Section 5.19, Silicon

7.1.2.2 Elements in the Zirconium Cladding. Zircaloy-2[®] alloy was used as cladding material for N Reactor fuel. Composition specifications for the Zircaloy-2[®] are provided in Rockwell (RHO 1980). These specifications and an estimate based on fabrication and processing of 5,690 MTU are shown in Table 7.1-7. The estimate is based on the mid-range or maximum impurity specifications as available. The weighted average of cladding to uranium in the various N Reactor fuel elements is 70.35 kg/MTU for 0.947 percent ²³⁵U fuel, 85.65 kg/MTU for spike fuel, and 87 kg/MTU for co-product drivers.

Table 7.1-7. Elements Introduced by Zircaloy-2® Cladding.

Element	Specification ($\mu\text{g/g}$)	Inventory (MT)
Al	75	≤ 0.031
Sn	12,000-17,000	6.1
Fe	700-2,000	0.57
Cr	500-1,500	0.42
Ni	300-800	0.23
C	275	≤ 0.12
U	2.5	≤ 0.001
Zr	Balance	413
B	0.5	≤ 0.0002
Cd	0.5	≤ 0.0002
Co	10	≤ 0.0042
Cu	50	≤ 0.021
Hf	200	≤ 0.084
Pb	100	≤ 0.042
Mg	20	≤ 0.0084
Mn	50	≤ 0.021
Mo	50	≤ 0.021
N	80	≤ 0.034
Si	100	Compiled in Section 5.19, Silicon
Na	20	≤ 0.0084
Ti	50	≤ 0.021
W	50	≤ 0.021
V	50	≤ 0.021

7.1.2.3 Elements in the Braze Filler. The end caps of the N Reactor fuel are brazed to the rest of the fuel element with a 95 percent zirconium, 5 percent beryllium braze. Composition specifications for the braze filler are provided in Rockwell (RHO 1980). These specifications and an estimate based on fabrication and processing of 5,690 MTU are shown in Table 7.1-8. The estimate is based on the mid-range or maximum impurity specifications as available.

Table 7.1-8. Elements from N Reactor Fuel Braze.

Element	Specification ($\mu\text{g/g}$)	Inventory (MT)
Al	145	≤ 0.001
Be	47,500-52,500	0.39
Sn	11,400-17,000	0.11
Fe	600-2,100	0.001
Cr	500-1,500	0.0078
Ni	2,800-800	0.014
C	500	≤ 0.0039
U	4	0.0
Zr	Balance	7.3
B	0.5	0.0
Cd	0.5	0.0
Co	20	≤ 0.0002
Cu	60	≤ 0.0005
Hf	200	≤ 0.0016
Pb	130	≤ 0.001
Mg	60	≤ 0.0005
Mn	60	≤ 0.0005
Mo	50	≤ 0.0005
N	200	≤ 0.0016
Si	250	Compiled in Section 5.19, Silicon
Ti	20	≤ 0.0002
W	100	≤ 0.0008
V	50	≤ 0.0004

7.2 MINOR ELEMENTS INTRODUCED BY FUEL FISSION AND ACTIVATION

Of all the fission and activation radionuclides, as indicated by ORIGEN2 code runs, only a few are present in chemically significant quantities. Table 7.2-1 summarizes the reactor generated elements that were determined to amount to more than 1.0 MT.

Table 7.2-1. Radionuclides with Chemically Significant Mass^a.

Fission product element	Mass (MT)
Rb	1.03
Sr ^b	1.23
Y	1.32
Zr ^b	9.99
Mo	7.98
Tc	1.61
Ru	4.26
Rh	1.26
Pd	1.30
Te	1.00
Cs	2.08
Ba	4.47
La ^b	2.98
Ce ^b	5.71
Pr	2.75
Nd	9.87
Sm	2.13

^aFission Product/Activation Product elements with mass greater than 1.0 MT are listed.

^bElements which are also evaluated in Section 5.0 and, therefore, are not considered further in this section.

Most of the radionuclides were discharged from the separation plants in the primary waste stream and were dispersed to the underground storage tanks. Some of these

radionuclides, however, were considered to have economic value and were recovered as products for beneficial use under defense and non-defense programs. The principal radionuclides targeted for recovery were plutonium, uranium, cesium, strontium, and neptunium. Cesium and strontium are discussed in Sections 6.2, and strontium is also discussed in Section 5.22. Zirconium is discussed in Section 5.24. Uranium is not listed in Table 7.2-1 as it is a major waste component, separately discussed in Section 6.3

7.3 MINOR ELEMENTS INTRODUCED BY CHEMICAL PROCESS OPERATIONS

A number of chemical processes were operated at the Hanford Site to recover plutonium, uranium, strontium, cesium, as well as other isotopes. The processes reviewed are as follows:

Bismuth Phosphate Process. All elements identified for this process are discussed in Section 5.0.

Tri-Butyl Phosphate/Uranium Recovery Process. All elements identified for this process are discussed in Section 5.0.

In-Farm Scavenging. All elements identified for this process are discussed in Section 5.0.

REDOX Process. Review of the REDOX facility indicated that silver nitrate was used to regenerate iodine reactors in the offgas system.

PUREX Process. Review of the PUREX facility and process indicated that silver nitrate was used to regenerate iodine reactors and cadmium nitrate was used as a neutron poison. PUREX was also used to separate ^{233}U from thorium.

Waste Fractionization at B Plant. Review of the waste fractionization processes indicated that tungsten was used in the Phosphotungstic Acid (PTA) Precipitation process for cesium recovery.

Plutonium Finishing Plant. All elements identified for this process are discussed in Section 5.0.

7.3.1 Cadmium

Cadmium was used as a neutron poison at PUREX during PRTR fuel processing and as a neutron poison during rework of the concentrated plutonium product. Cadmium was procured as 50 wt% cadmium nitrate solution for this use. Cadmium was also a trace impurity present in reactor fuel fabrication.

7.3.1.1 Identification of Cadmium Values. Allen (1976) reports that 19,000 gm-moles of $\text{Cd}(\text{NO}_3)_2$ (2,100 kg Cd) was used by the various Hanford Site separations facilities for the time period from 1944 to 1975. The HDW-EIS (DOE 1987) reports an inventory of 4,000 kg of Cd, all in the SSTs. The TWRS Baseline inventory (Shelton 1996) shows an inventory of 14,200 kg Cd, all in the DSTs. Neither the TRAC model nor the HDW model include values for cadmium.

7.3.1.2 Cadmium Inventory Evaluation. Cadmium is a neutron poison, and its presence in reactor fuel was tightly controlled during fuel fabrication. As is discussed in Section 7.1, "Minor Elements Introduced by Fuel Fabrication," a maximum of 0.1 MT cadmium could have come from this source. Nearly all of the cadmium would have been in aluminum clad fuel.

The first indication of cadmium usage by a reprocessing facility on the Hanford Site is from the monthly report for April 1968 (ARHCO 1968). The reports states that a process test using $\text{Cd}(\text{NO}_3)_2$ as a neutron poison during rework of plutonium at PUREX was completed that month.

The flowsheet for the first PRTR fuel processing campaign at the PUREX Plant states that cadmium nitrate was used as a soluble poison, added to the mixed oxide dissolvent as a secondary criticality prevention measure (Matheison and Nicholson 1969). The first PRTR processing campaign at PUREX was in February 1969. The flowsheet indicates that the cadmium was added as 50 percent $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, defined as 135 lb Cd in 80 gal solution. Previous flowsheets for processing of PRTR and Shippingport fuel at REDOX do not indicate the use of cadmium.

Material specifications for $\text{Cd}(\text{NO}_3)_2$ were not included in the 1958 or 1966 Essential Material Manuals (HAPO 1958, Isochem 1966). The 1978 Essential Material Manual includes a specification for 50 wt% $\text{Cd}(\text{NO}_3)_2$ (Palmer 1978).

The conclusion from review of these documents is that the use or introduction of cadmium into the waste tank system before 1968 is unlikely.

Essential material consumption records were obtained for 1965 through 1984. These records were used to compile the cadmium nitrate consumption record shown in Table 7.3-1. Essential material consumption records of cadmium usage after 1984 were not maintained. Plant records, that may have shown cadmium usage, were not located. It is possible that substantial additions of cadmium were made by the PUREX Plant to the waste tank system during the 1985-1995 time period.

Table 7.3-1. Fifty Percent Cadmium Nitrate Consumption.

Year	lb	kg
1965	No record of use	--
1966	No record of use	--
1967	No record of use	--
1968	400	181
1969	1,500	680
1970	6,000	2,722
1971	1,750	794
1972	4,767	2,162
1973	490	222
1974	0	0
1975	0	0
1976	0	0
1977	7,086	3,214
1978	0	0
1979	149	66
1980	0	0
1981	0	0
1982	0	0
1983	3,411	1,547
1984	0	0
Total 1965-1984	25,553	11,590
1985 to Present	No records located	

There is some question as to how much of the 3,200 kg of 50 percent cadmium nitrate transferred from the warehouse in 1977 was actually consumed by PUREX. It is known that some of the cadmium nitrate storage drums at the warehouse were leaking, and that at least the solution from those drums was transferred to a holding tank at PUREX for storage. At the time PUREX was not operating and would have had little demand for cadmium nitrate. This remaining material was used after the PUREX plant restarted in 1983.

Table 7.3-2 summarizes the cadmium inventory in the waste tanks indicated by the essential material consumption records. The notations used in the essential material consumption records are not clear as to the form of cadmium nitrate being recorded, 50 percent basis or 100 percent basis. However, by the conventions used elsewhere in the records, a 50 percent basis is assumed. Based on the PUREX PRTR flowsheets, 50 wt% $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ was assumed as the formulation for 1964 through 1973. For the post 1973 usage, 50 wt% $\text{Cd}(\text{NO}_3)_2$ was assumed as the formulation, based on the 1978 Essential Material Manual (Palmer 1978).

Table 7.3-2. Cadmium Inventory from Essential Material Consumption Records.

Year	Cadmium inventory (100% basis) (kg)
1968-1973	1,232
1974-1984	1,148
1985 to Present	No records located
Subtotal	2,380

The TWRS Baseline inventory for cadmium is primarily based on sample characterization results for the DSTs and a projection of future cadmium usage during facility decontamination. The sample database does not include data for all of the DSTs. However, cadmium is only expected to be found in aging waste tanks and the aging waste tanks are included in those tanks that have been sampled. Sample data show relatively large amounts of cadmium in each of the AZ tanks, and relatively small quantities in the AY tanks. Most of the cadmium in the aging waste would have been added after 1983. The basis for the TWRS inventory can be summarized as shown in Table 7.3-3. The cadmium inventories of the aging waste tanks are based on samples collected after PUREX ceased operation in 1989.

Table 7.3-3. Summary of Tank Waste Remediation System Baseline Cadmium Inventory.

Source	Cadmium (kg)
Single-shell tanks	No values reported
Aging waste tank 241-AZ-101	1,110
Aging waste tank 241-AZ-102	4,280
Other double-shell tanks	1,325
Future facility waste	7,460
Total	14,175

The cadmium estimate for future facility waste shown in Table 7.3-3 is based on a 1988 projection of cadmium usage during cleanout of the PUREX Plant. The basis for the cadmium estimate in the facility waste projection was reassessed for this evaluation. A report by Harty (1993) indicates that the miscellaneous waste from the PUREX Plant will contain on the order of 99 kg of cadmium. Recent waste profile data sheets for the PUREX Plant do not indicate the presence of cadmium.

7.3.1.3 Best-Basis Value for Cadmium. The various values for cadmium are in wide disagreement. In addition, the supporting data for each of the values are generally incomplete.

The 19,000 gm-moles of $\text{Cd}(\text{NO}_3)_2$ (2,100 kg Cd) reported by Allen (1976) to have been used in the various Hanford Site separations facilities between 1944 and 1975 was traced to the essential material consumption records through the authors notes. Because of the notation conventions in the records it is not clear that Allen was told or understood that the recorded values were for 50 wt% $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ rather than 100 percent $\text{Cd}(\text{NO}_3)_2$ which is what appears to have been assumed by the author. Adjustment of Allen's calculations to the 50 wt%, 4-hydrate basis, would reduce the cadmium usage estimate to 798 kg. This value is consistent with cadmium usage reported in the essential material consumption records (Table 7.3-1) for 1965-1971 and may indicate that the author did not have access to all of the records.

With the exception of the AX Farm, little cadmium should be expected to be found in the SSTs. Cadmium was not introduced into the waste until 1968. From 1968 until 1972, when PUREX shut down, PUREX waste was routed to SSTs in AX Farm. In 1976 AX Farm was sluiced for strontium recovery. After 1978, PUREX waste was routed to double shell tanks in AY Farm and in 1983 to double shell tanks in AZ Farm.

The best-basis cadmium inventory estimate uses the DST data from the TWRS Baseline Inventory and the pre-1976 essential materials data for the SSTs. The reevaluated future facility waste estimate is included. These sources are summarized in Table 7.3-4.

Table 7.3-4. Best-Basis Cadmium Inventory.

Source	Cadmium (kg)	Basis
Single-shell tanks	1,232	1965-1976 Essential Material Records
Aging waste tank 241-AZ-101	1,110	TWRS Baseline Inventory
Aging waste tank 241-AZ-102	4,280	TWRS Baseline Inventory
Other double-shell tanks	1,325	TWRS Baseline Inventory
PUREX Plant cleanout	99	Facility Waste Reassessment
Impurity from aluminum clad reactor fuel	< 100	Section 7.0
Total	8,145	

PUREX = Plutonium-uranium extraction
TWRS = Tank Waste Remediation System.

The best-basis value for cadmium is the value developed by the independent evaluation, 8,200 kg. The reasons for using the independent evaluation value are as follows:

- The HDW model does not consider cadmium.
- The TWRS Baseline Inventory overestimates cadmium in the future facility waste.
- Essential material records were not available after 1984 and sample data for the SSTs are lacking. The use of a large uncertainty value (± 25 percent) is recommended.

7.3.2 Silver

From 1950 through 1989, small, but significant (hundreds of kilograms), amount of silver were used in "silver reactors" in Hanford Site fuel reprocessing plants to trap and restrict emission of large amounts of radioisotopes of iodine, specifically ^{131}I . Some, perhaps a sizeable fraction, of the silver used in these reactors is believed to have eventually been introduced into one or more of the underground tanks.

Smaller amounts of silver were also introduced into the Hanford tanks as a result of pilot plant and plant scale use of silver nitrate to catalyze sodium peroxydisulfate oxidation of $^{144}\text{Ce(III)}$ to $^{144}\text{Ce(IV)}$ to facilitate its separation from associated trivalent rare earths. Laboratory wastes containing very small amounts of silver were also introduced, occasionally, into one or more tanks.

There is concern about establishing a global inventory of silver, not only for completeness, but also for two other reasons. There is some evidence to suggest silver present in sludges that are to be vitrified may be reduced to the metallic state, along with any platinum group metals, thereby interfering with operation of a Joule-heated melter. Secondly, air quality regulations limit the amount of silver that can be present in gases released to the environment.

7.3.2.1 Utilization of Silver in Hanford Plants.

7.3.2.1.1 Silver Reactors. So-called silver reactors were used in all Hanford Site fuel reprocessing plants after 1950 to restrict release of radioiodine, specifically the isotopes ^{129}I and ^{131}I to the environment. Although construction designs of the silver reactors changed over 40 years, the principle of operation of the silver reactors remained essentially the same. A solution of silver nitrate was passed over a bed of ceramic bodies, typically Berl saddles, to wet all the packing with AgNO_3 . The packing bed was then dried. Dissolver offgases were heated to temperatures in the range 230-260 °C and then passed through a cylindrical silver reactor. Molecular iodine was removed from the heated offgases by one or more of the following chemical reactions:

1. $6 \text{AgNO}_3 + 3 \text{I}_2 + 3\text{H}_2\text{O} = 5\text{AgI} + \text{AgIO}_3 + 6\text{HNO}_3$
2. $6 \text{AgNO}_3 + 3 \text{I}_2 = 5 \text{AgI} + \text{AgIO}_3 + 6\text{NO}_2 + 3/2 \text{O}_2$
3. $2 \text{AgNO}_3 + \text{I}_2 = 2 \text{AgI} + 2 \text{NO}_2 + \text{O}_2$
4. $2 \text{AgNO}_3 + \text{I}_2 + 2 \text{O}_2 = 2 \text{AgIO}_3 + 2 \text{NO}_2$

Data garnered over more than 40 years of plant-scale operation of silver reactors supplemented, in some cases, by laboratory-scale tests showed the following:

- Fresh silver nitrate reactors retained molecular iodine quite efficiently, i.e., decontamination factors of the order of 1000.
- Sorption of radioiodine in the first few centimeters depth of the silver reactor was quite significant.
- In some cases, silver reactors did not remove organic iodine compounds present in the offgases passed through them.
- Chlorine present in the offgas in some PUREX process operation was sorbed very efficiently by the silver reactor and significantly decreased capacity of the bed for removing radioiodine.

Historical technical manuals, e.g., GE (1951), RHO 1980, provide much information concerning design and operation of the silver reactors. Apparently, up to about 1956 or 1957 silver reactors in the BiPO₄ and REDOX processes were used on a once-through basis. When their efficiency for removing radioiodine decreased to a predetermined level they were removed from service and disposed of as a solid waste. Hence, AgNO₃, at least the bulk of it, consumed from 1950 through 1958 was not routed to the underground tanks.

Barnes (1958) described a new silver reactor design for use in the REDOX plant. This new design allowed for periodic flushing of the reactor contents and recharging with fresh AgNO₃ solution. Silver reactors used after 1958 in the REDOX and PUREX plants were all constructed to permit periodic flushing and regeneration. Walser (1994) reported complete details for flushing and regeneration of PUREX plant silver reactors from plant startup to plant shutdown in 1988.

What is not at all clear from published historical reports are details of the silver reactor flushing procedure. Apparently, the flush solution was water. But, no data have been found to indicate how much silver was removed in each flush. Certainly, any unreacted AgNO₃ left on the reactor would have dissolved in water; perhaps AgIO₃ on the reactor bed might also have been soluble in the flush water. But, what about AgI and AgCl? Were these water-insoluble silver compounds removed by mechanical action or did they remain on the reactor? Very likely, these two latter compounds split between the spent flush solution and the ceramic support material.

There appears to be no reason to doubt that the spent silver reactor flushes were routed to the underground waste tanks. Based on technical conversations with plant personnel, at least one flush solution was routed to tank 241-AW-102 in March 1986, and one or more failed silver reactors, presumably after water flushing, are (or were) stored in the PUREX plant equipment storage tunnel. As noted above, these failed beds likely contained at least some of the silver purchased at the Hanford site. A recent report by Dodd (1997) accounts for 740 kg of silver in used silver reactors stored in the PUREX Number 2 Storage Tunnel.

7.3.2.1.2 B Plant and Hot Semiworks. Very limited use was made in the 1960's in the Hot Semiworks and in B Plant of silver as a catalyst in sodium peroxydisulfate oxidation of ¹⁴⁴Ce(III) to ¹⁴⁴Ce(IV). In all cases the goal was to separate ¹⁴⁴Ce from associated trivalent rare earths. Once oxidized to the tetravalent state, Ce(IV) can be efficiently extracted with a solution of bis(2-ethylhexyl)phosphoric acid in a hydrocarbon diluent from highly acidic, i.e., >1M H⁺, aqueous solution. Under these conditions, little, if any, of the trivalent rare earths are extracted.

The Ag-catalyzed peroxydisulfate oxidation process for extraction and separation of ¹⁴⁴Ce was tested once on a pilot-plant scale in the Hot Semiworks. Subsequently, in the early days of B Plant operation it was also apparently used on a plant scale for a very short time (1967 through 1968). No written records were located that stated exact details of B Plant operation of the cerium oxidation procedure. Engineers who worked at B Plant do not recall routine or extensive application of the Ag-catalyzed cerium oxidation process. The

best and most positive evidence that AgNO_3 was used for a time in B Plant comes from historical Essential Material Consumption records maintained by McDonald (1965) and McDonald and Hogan (1969). These records indicate consumption of 4,175 lb of AgNO_3 , equivalent to 1,200 kg of Ag, used in the period 1966 through 1968. Presumably, all this silver was eventually routed to the underground storage tanks.

7.3.2.2 Sources of Silver in Waste Tanks.

7.3.2.2.1 Impurity in Fuel and Cladding. Information presented in Section 7.1 does not account for any silver impurity in either the uranium metal fuel or in the aluminum or Zircaloy® cladding.

7.3.2.2.2 Fission Process. Fission of ^{235}U produces both stable (^{107}Ag and ^{109}Ag) and radioactive ($^{110\text{m}}\text{Ag}$) isotopes of silver. The half-life of $^{110\text{m}}\text{Ag}$ is 235 d. Because many years have gone by since the last atoms of $^{110\text{m}}\text{Ag}$ were produced in Hanford reactors, essentially no $^{110\text{m}}\text{Ag}$ remains in the tank waste.

The fission yield of ^{107}Ag is quite small compared to that of ^{109}Ag ; hence, the contribution of fission-generated ^{107}Ag to the global tank silver inventory can be safely ignored.

The contribution of fission-generated ^{109}Ag to the global tank silver inventory is considered only significant for waste derived from processing of N Reactor fuel. (Neutron irradiation of N Reactor fuel was significantly higher, i.e., greater megawatt days/ton of uranium, than for fuel in other Hanford plutonium production reactors.) Allen (1982) estimates that 0.0013 kg of fission-product silver was present in each metric ton of N Reactor fuel containing 6 percent ^{240}Pu . For irradiated N Reactor fuel containing 12 percent ^{240}Pu , Allen estimates that 0.0052 kg of fission-product silver were present in each metric ton of fuel.

Data in Appendix B of this document state that N Reactor fuel containing 3,890 MTU were reprocessed in the Hanford PUREX plant from 1983 through 1990. Of this quantity, 3,554 MTU are estimated (see tank 241-AZ-102 in Kupfer et al. 1997) to have contained 6 percent ^{240}Pu and 335 MTU to have contained 12 percent ^{240}Pu . From Allen's estimates then, the 6 percent ^{240}Pu fuel contributed 4.6 kg of stable fission product silver to the global tank inventory while the 12 percent ^{240}Pu fuel contributed 1.8 kg of fission product silver to this latter inventory. Note that this analysis does not take into account the few metric tons of N Reactor fuel processed in both the REDOX and PUREX plants before 1983.

7.3.2.2.3 Essential Material Purchases/Consumption. Silver nitrate was designated one of the many Essential Materials at the Hanford Site. Records were kept, at least from 1956 on, of the amounts of AgNO_3 that were purchased and used in various Hanford facilities. No records were found for either purchase or usage of AgNO_3 before 1956. Such records were either not kept or, more likely, are not readily available. Available historical information on purchases and consumption are summarized as follows:

1956 through 1964: Allen (1976) states that over the period 1956 through 1964 23,250 lb of silver nitrate were purchased. This amount of AgNO_3 contained 6,780 kg of silver.

1965 through 1967: McDonald (1965) reports that 4,734 lb of AgNO_3 , containing 1,381 kg of silver, were consumed in process operations performed in the PUREX (2,313 lb), REDOX (91 lb), and B Plants (2,330 lb).

1968: For 1968, McDonald and Hogan (1969) records consumption of 587 lb of AgNO_3 in the PUREX plant and 1,845 lb of AgNO_3 in the B Plant. This total amount of AgNO_3 contained 709 kg of silver.

1969 through 1977: Data recorded in McDonald (1965) indicate no consumption of AgNO_3 in the period 1969 through 1977. The REDOX plant completed operation in 1966, and the PUREX plant ceased operation from 1971 to 1983.

1978 through 1984: Hogan (1992) reported that restart of the PUREX plant in 1983 involved consumption of 500 lb of AgNO_3 .

1985 through 1988: Historical records, if they exist, detailing purchase and consumption of AgNO_3 in the last years (1985 through 1988) of Hanford PUREX plant operation were not located. As indicated previously, however, Walser (1994) estimates that 2,050 lb of AgNO_3 (649 kg of silver) were consumed in the period 1983 through 1988 in regenerating silver reactors.

Available historical records thus indicate a grand total of 33,166 lb of AgNO_3 were purchased and used in the period 1956 through 1988 in operation of various fuel reprocessing and waste management facilities at the Hanford Site. This quantity of AgNO_3 contained 9,665 kg of silver.

7.3.2.3 Best-Basis Global Silver Inventory Estimate. The most straightforward approach to arrive at a best-basis global inventory for silver in the Hanford tanks is to sum:

	kg Ag
Recorded (1956-1988) AgNO_3 Purchases and Consumption	9,665
Fission product stable silver isotopes in N Reactor fuel	6.4
Used silver reactors stored in PUREX storage tunnel number 2	-740
Total	8,931

The total of 8,931 kg of silver, even though it does not account for AgNO_3 purchased and consumed before 1956, is thought to represent a very conservative maximum of silver that could be in the 177 Hanford tanks. As noted previously, silver purchased, as AgNO_3 , before 1956 is believed to have been disposed of as a solid waste in spent silver reactors used in the B, T, and REDOX plants. Of the total of 9,665 kg of silver purchased and consumed from 1956 through 1988, at least some is known to have been discarded as solid waste in spent silver reactors (Dodd 1997).

Although considered conservative, the value 8,931 kg is considered to be the best-basis global silver inventory.

The fact that the 8,931 kg value of silver is at least a reasonable best-basis global inventory estimate is supported by the data in Table 7.3-5. The results listed in Table 7.3-5 indicate a total of 6,360 kg of silver are contained in 65 tanks for which silver inventory data have been reported. The notation "No data" in Table 7.3-5 for 53 tanks does not mean that silver is not present, only that the available information did not permit an estimate of the silver inventory of the tank. Less-than values listed in Table 7.3-5 were taken as equal to the numerical value cited in computing the total of 6,360 kg of silver.

Of the 65 tanks (Table 7.3-5) for which a silver inventory datum is available, at the time this report was written, most contain less than 100 kg of silver. Tanks 241-C-102, 241-C-103, 241-C-104, 241-C-106, 241-AY-102, 241-SX-108, 241-T-111, 241-AZ-102, 241-AW-105, and 241-AW-103 stand out for their relatively high content of silver. Tank 241-C-106 received sludge residues which did not dissolve when PUREX process sludges in single-shell tanks were treated with HNO_3 to remove ^{90}Sr in B Plant operations in the 1960's and 1970's. It is also possible that the Ag content in 241-C Farm tanks is high since flushes of the silver reactors were likely routed to nonboiling waste, and until DSTs were available, the 241-C Farm tanks were the PUREX process nonboiling waste receivers.

Table 7.3-5. Tank-By-Tank Inventory of Silver. (2 Sheets)

Tank	Ag, kg	Tank	Ag, kg	Tank	Ag, kg
241-A-104	No data	241-BY-105	39.5	241-T-105	28
241-A-105	No data	241-BY-107	27	241-T-106	No data
241-AN-102	<0.787	241-BY-108	<49	241-T-107	No data
241-AX-101	No data	241-BY-109	36.4	241-T-108	<3
241-AX-102	No data	241-BY-110	37.2	241-T-109	No data
241-AX-103	87	241-BY-111	No data	241-T-110	No data
241-AW-101	No data	241-BY-112	No data	241-T-111	280
241-AW-102	No data	241-C-102	280	241-T-112	40
241-AW-103	925	241-C-103	137	241-T-201	1.7
241-AW-104	No data	241-C-104	1,200	241-T-202	0.8
241-AW-105	415	241-C-105	38	241-T-203	No data

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Table 7.3-5. Tank-By-Tank Inventory of Silver. (2 Sheets)

Tank	Ag, kg	Tank	Ag, kg	Tank	Ag, kg
241-AW-106	No data	241-C-106	1,390	241-TX-102	No data
241-AY-101	194	241-C-108	No data	241-TX-104	No data
241-AY-102	563	241-C-109	<0.5	241-TX-105	No data
241-AZ-101	78.2	241-C-110	1.1	241-TX-106	No data
241-AZ-102	242	241-C-111	No data	241-TX-107	No data
241-B-104	No data	241-C-112	No data	241-TX-109	14
241-B-105	No data	241-C-201	No data	241-TX-110	37
241-B-106	<21	241-C-203	No data	241-TX-111	29
241-B-108	<126	241-C-204	No data	241-TX-113	43
241-B-109	<154	241-S-101	29	241-TX-115	No data
241-B-110	58.5	241-S-102	53	241-TY-101	2.8
241-B-111	6.34	241-S-104	No data	241-TY-105	No data
241-B-112	0.81	241-S-105	No data	241-U-101	No data
241-B-201	1.7	241-S-106	43	241-U-104	No data
241-B-202	1.0	241-S-107	89.5	241-U-105	47.7
241-B-203	1.0	241-S-108	No data	241-U-106	23.7
241-B-204	No data	241-S-110	No data	241-U-107	33
241-BX-101	No data	241-S-111	No data	241-U-108	34
241-BX-103	No data	241-SX-101	No data	241-U-110	No data
241-BX-104	<33	241-SX-102	46.2	241-U-111	No data
241-BX-105	No data	241-SX-103	57.0	241-U-202	No data
241-BX-106	No data	241-SX-104	52.9	241-U-204	No data
241-BX-108	No data	241-SX-105	61		
241-BX-109	No data	241-SX-106	43		
241-BX-110	4.68	241-SX-108	525		
241-BX-111	19.1	241-SX-109	4.58		
241-BX-112	No data	241-SX-110	6.21		
241-BY-101	<32.7	241-SX-111	7.92		
241-BY-102	<319	241-SX-112	7.01		
241-BY-103	41.9	241-SX-114	9.0		
241-BY-104	29.1	241-SX-115	0.76		

No data = Available information did not permit an estimate of the silver inventory in this tank. It does not mean that silver is not present in the tank.

7.3.3 Thorium

The PUREX Plant was used to recover ^{233}U and Th from irradiated thoria (ThO_2) targets. A plant process test of the thoria flowsheet was carried out in the PUREX Plant in January-February 1965 to test systems capabilities (Duckworth and Ward 1965). The first thoria campaign was run in 1966 (Isochem 1967). The second, final thoria campaign occurred in 1970 (Jackson and Walser 1977).

The campaign reports include material balance data for thorium charged to the dissolvers, thorium loaded out in the product, and thorium in the waste streams. Table 7.3-6 summarizes the thorium losses to the waste tanks.

Table 7.3-6. Thorium Losses to the Waste Tanks (MT).

	Process Test	1966 Campaign	1970 Campaign	Total
Dissolver Receipts minus Plant Loadout	0.83	5.6	19.2	25.6
Material Accounted for in Waste Streams	0.83	3.5	10.4	14.7
Material Unaccounted For (MUF)	--	2.1	8.8	10.9

The process losses determined by sampling and laboratory analysis of the waste stream are substantially less than the losses determined by comparison of the dissolver receipts and plant loadout records. Thorium was loaded out from PUREX as $\text{Th}(\text{NO}_3)_4$ solution and the loadout inventory was calculated from solution volumes and concentrations.

The difference, after subtraction of the thorium loaded out and the known losses from the dissolver receipts is the material unaccounted for (MUF). MUF represents the uncertainties in the thorium losses and the amount of thorium shipped. This difference is made up of both random and systematic analytical errors. For thorium, the total MUF amounts to 2 percent of the thorium shipped.

The thorium inside the reactor target elements was either in the form thoria powder or thoria wafers. As such, it is probable that some of the thorium would have been lost from the PUREX process as fines. Particulate losses in the coating waste stream from the dissolvers is a known phenomena. Analytical tests only determine the amount of thorium in solution. Laboratory procedures for sample preparation are, in general, less than 100 percent efficient at dissolving solids. Thus it is probable that the thorium MUF is material that was lost to the tanks rather than uncertainty in material loaded out. Although

the verifiable thorium in the tanks is only 14.7 MT, the best-basis estimate for thorium in the tanks includes the MUF and is 25.6 MT.

7.3.4 Tungsten

Tungsten was used at B Plant to recover cesium by the PTA precipitation process.

7.3.4.1 Identification of Tungsten Values. Allen (1976) reports that 3,600 gm-moles of $P_2O_5 \cdot 24WO_3 \cdot 44H_2O$ was used in the PTA precipitation process. The weight percent tungsten in PTA is approximately 68 percent, 3,600 gm-moles of PTA equates to 15,900 kg W. Neither The TWRS Baseline inventory (Shelton 1996), the TRAC model nor the HDW model include values for tungsten.

7.3.4.2 Tungsten Inventory Evaluation. PTA was purchased for use at B Plant as 92 wt% $P_2O_5 \cdot 24WO_3 \cdot 44H_2O$. Trace amounts of tungsten were also introduced as an impurity in the Zircaloy® cladding used for N Reactor fuel. A maximum of 22 kg tungsten could have come from this source.

Material specifications for $P_2O_5 \cdot 24WO_3 \cdot 44H_2O$ were not included in the 1958, 1966, or 1978 Essential Material Manuals (Isochem 1966, HAPO 1958, and Palmer 1978). Allen (1976) shows PTA usage between 1969 and 1973.

Essential material consumption records extracted by Allen (1976) are shown in Table 7.3-7. All numbers except for the 1973 entry were verified from original records. Essential material consumption records for 1973 indicate zero PTA usage. The total is equivalent to 23,255 kg or 3,580 gm-moles PTA at 100 percent basis. In this case, the PTA tungsten source term is 15,800 kg of tungsten.

Table 7.3-7. 92 percent Phosphotungstic Acid Consumption.

Year	lb	kg
1968	0	0
1969	21,250	9,639
1970	7,153	3,245
1971	16,353	7,418
1972	10,345	4,692
1973	625	284
Total 1969-1973	55,726	25,277

7.3.4.3 Best-Basis Value for Tungsten. The 3,600 gm-moles of $P_2O_5 \cdot 24WO_3 \cdot 44H_2O$ (PTA) reported by Allen (1976) appears to be the result of a modest typographical error or rounding assumption in the final report.

The best-basis tungsten inventory estimate is summarized in Table 7.3-8.

Table 7.3-8. Best-Basis Tungsten Inventory.

Source	Tungsten (kg)	Basis
PTA precipitation process	15,900	Essential Material Records
Impurity from Zircaloy®-clad reactor fuel	<22	Section 7.0
Total	15,900	

PTA = Phosphotungstic acid.

The best-basis value for tungsten is the value developed by this evaluation, 15,900 kg \pm 10 percent (1,590 kg). The reasons for using this evaluation are as follows:

The HDW model does not include tungsten.

The TWRS Baseline Inventory does not include tungsten.

The estimate is based on essential material records. Furthermore, no alternate source terms/or tungsten have been identified.

7.4 MINOR ELEMENTS INTRODUCED BY CHEMICAL IMPURITIES

Essential Material Specifications for the various chemical processes will be reviewed for the presence of minor elements. Section 5.20 identifies 54,200 MT of sodium in the tanks and an additional 23,000 MT sodium routed to the cribs. Assuming all sodium came from 50 wt% caustic soda, the sodium inventory corresponds to 262,600 MT 50 wt% NaOH. Based on this assumption the industry specifications for commercial grade NaOH, an estimate of the impurities introduced into the waste via that route can be made (see Table 7.4-1).

Table 7.4-1. Potential Impurities Introduced by Sodium Hydroxide.

Impurity	Impurity specification for commercial grade 50 wt% NaOH (wt%)	Element	Maximum impurity (MT)
NaCl	1.0	Cl	≤1,590
Fe	0.0004	Fe	≤1.1
Hg	0.00005	Hg	≤0.1
Na ₂ SO ₄	0.15	S	≤89
SiO ₂	0.04	Si	≤49
Al ₂ O ₃	0.004	Al	≤5.6
CaO	0.006	Ca	≤11
MgO	0.001	Mg	≤1.6
Mn	0.0001	Mn	≤0.3
Ni	0.0001	Ni	≤0.3
Cu	0.0002	Cu	≤0.5

The two other major chemicals purchased for use in the separation processes were nitric acid and aluminum nitrate. The essential material manuals provide little information on the impurities found in nitric acid. In addition the inventory of virgin nitric acid used has not been established. Section 5.1 provides an estimate of the amount of ANN consumed and the essential material manuals provide impurity limits. Table 7.4-2 provides a bounding estimate of trace chemicals due to the ANN consumption.

Table 7.4-2. Trace Chemicals Introduced by 50 Wt% ANN Consumption.

Impurity	(Isochem 1966) (wt%)	Palmer (1978) (wt%)	Element	Inventory (kg)
Insolubles	0.2	0.2	Insolubles	<244
Ammonia	0.02	0.02	NH ₄	<24
Chloride	0.005	0.003	Cl	<6
Alkali and alkali earths as sodium	0.2	0.2	As Na	<244
Heavy metals as Pb	0.01	0.01	As Pb	<12
Sulfate	0.02	0.02	S	<9.8
Silica	0.01	0.01	Si	<5.6
Iron	0.03	0.02	Fe	<36
Nitrate	0.001	0.001	NO ₃	<1
Calcium	0.0072	0.005	Ca	<8.8
Strontium	0.0014	0.0002	Sr	<1.7

7.4.1 Minor Elements Introduced by Tank Farm Actions

In 1966, tank farm operations added 57,000 kg of Portland cement to tank 241-BY-105 (Krieg et al. 1990). A composition of Portland cement is provided by Kirk-Othmer (1979). During 1970 to 1972, a total of 354,000 kg of diatomaceous earth was added to tanks 241-BX-102, 241-SX-113, 241-TX-116, 241-TX-117, 241-TY-106, and 241-U-104 (Krieg et al. 1990). A chemical analysis for typical diatomaceous earth was reported by Buckingham and Metz (1974). The moisture content of the diatomaceous earth that was used is unknown. Tables 7.4-3 and 7.4-4 summarize the inventories added due to these sources.

Table 7.4-3. Portland Cement Inventory.

Composition (Type II)	(Wt%)	Element	Inventory (kg)
SiO ₂	21.7	Si	5,780
Al ₂ O ₃	4.7	Al	1,420
Fe ₂ O ₃	3.6	Fe	1,440
CaO	63.6	Ca	24,600
MgO	2.9	Mg	1,000
SO ₃	2.4	S	550
Other	0.4	Other	228
Loss on Ignition	0.8	Loss	456

Table 7.4-4. Diatomaceous Earth Inventory.

Composition	(Wt%)	Element	Inventory (kg)
SiO ₂	92.3	Si	152,700
Al ₂ O ₃	1.1	Al	2,060
Fe ₂ O ₃	2.0	Fe	4,950
CaO	0.9	Ca	2,280
MgO	0.4	Mg	854
Other	0.2	Other	708
Loss on Ignition	3.1	Loss on Ignition	11,000

Zeolite ion exchange media from the evaporator operations were also disposed of in the tanks. The contribution of zeolite ion exchange media to the inventory is discussed in Section 5.19 Silicon.

No other significant additions due to tank farms activities, not already discussed in Section 5.0, were identified.

7.5 SUMMARY OF BEST-BASIS INVENTORY VALUES FOR MINOR COMPONENTS

Results of the minor components assessment are reported in Table 7.5-1.

Table 7.5-1. Best-Basis Inventory of Minor Components (2 Sheets).

Element	Fuel fabrication (MT)	Fission and activation products (MT)	Chemical process operations (MT)	Process chemical impurities (MT)	Tank farm additions (MT)	Total (MT)
Ag		(a)	8.93			8.93
B	≤0.04					≤0.04
Ba		4.5				4.5
Be	0.45					0.45
Cd	≤0.1	(a)	8.05			8.15
Co	≤0.044	(a)				≤0.044
Cs		2.1				2.1
Cu	≤12.5			≤0.5		≤13.0
Hf	≤0.086					≤0.086
Li	≤0.3					≤0.3
Mg	≤2.9			1.6	1.85	6.35
Mo		8.0				8.0
Nd		9.9				9.9
Pd		1.3				1.3
Pr		2.8				2.8
Rb		1.0				1.0
Rh		1.3				1.3
Ru		4.3				4.3
Sm		2.1				2.13
Sn	≤20.2	(a)				≤20.2
Sr		1.2	29.1	<1		31.3
Tc		1.6				1.61
Te		1.0				1.0

Table 7.5-1. Best-Basis Inventory of Minor Components (2 Sheets).

Element	Fuel fabrication (MT)	Fission and activation products (MT)	Chemical process operations (MT)	Process chemical impurities (MT)	Tank farm additions (MT)	Total (MT)
Th						25.6
Ti	≤1.5	(a)				≤1.5
V	≤0.021					≤0.021
W	≤0.022		15.9			15.9
Y		1.3				1.3
Zn	≤1.1					≤1.1

^aPresent in quantities less than 1 MT.

^bMeasured extraction and decladding loss from PUREX plant campaigns, 1966 and 1970.

8.0 METHODOLOGY FOR ESTIMATING INDIVIDUAL TANK WASTE INVENTORIES

The evaluation to establish a best-basis or standard inventory for waste in each single-shell tank (SST) or double-shell tank (DST) is included in all new Tank Characterization Reports (TCRs) beginning in fiscal year 1997. Previously issued TCRs will also be revised to include a best-basis inventory. Preliminary TCRs will be issued to establish the best-basis inventories when TCRs are not already available, or are in progress.

The first section of the best-basis inventory in the TCR summarizes all of the information sources that can be used to estimate the chemical and radionuclide inventories for the tanks. The information sources typically consist of historical records and documents as well as tank waste sample analyses if available. Typically useful information includes the following:

- Waste transaction records and waste volume estimates such as reported in the Waste Status and Transaction Record Summary (WSTRS) (Agnew et al. 1997b); the Tank Layer Model (TLM) and Supernate Mixing Model (SMM) documented in the HDW report (Agnew et al. 1997a); waste transfer records reported in Anderson (1990); the SORWT Model (Hill et al. 1995), and the Waste Tank Summary Reports (e.g., Hanlon [1997]). Many other historical documents and operating records pertaining to tank farm and waste processing facilities (e.g., technical manuals, process memos, evaporator run reports) are also useful.
- Tank waste sample analyses if available (e.g., from core samples, grab samples, and augers). The data can be from the tank in question or from other tanks known to have a similar process history and/or contain the same waste types.
- Tank waste inventory estimates derived by the HDW model (Agnew et al. 1997a)
- Process flowsheets for the waste entering the tank.

Once the information sources have been collected, a detailed evaluation of the information sources is performed to ultimately arrive at a best-basis inventory estimate for the tank. Appendix J of this report summarizes all the major steps in a procedural fashion for performing a best-basis inventory evaluation. This section (8.0) discusses, in additional detail, a key part of the best-basis evaluation guidelines described in Appendix J. The TCR refers to this part of the best-basis inventory section as the "Component Inventory Evaluation."

The Component Inventory Evaluation (typically Appendix D, Section D3.0, in the TCR) consists of two major parts that utilize the information sources noted earlier:

1. Establish historical basis for the process origin of the waste presently in the tank. This activity encompasses identification of likely contributing waste types and volumes.
2. Perform an engineering assessment that uses information independent of sample data and HDW derived values to predict the chemical and radionuclide inventories. The results of this assessment is then compared to and reconciled with the following:
 - Sample-based inventory (if available) for the tank
 - HDW model inventory prediction.

Elements of these key inputs to the inventory estimation procedure are discussed in the following text.

8.1 CONTRIBUTING WASTE TYPES AND WASTE VOLUMES

There are several historical-based and sample-based sources of information needed to formulate a basis for the most likely waste type(s) in a tank(s), and associated volume of each waste type. The waste type refers to the process origin or the specific flowsheet that produced the waste, e.g., BiPO_4 process, REDOX process, PUREX process, evaporator bottoms (salt cake) from 242-B evaporator, etc. It is important to utilize available data and information to establish, as clearly as possible, the type(s) and amount(s) of waste now in each tank. Historical operating records and contemporary surveillance information need to be compared with sample analytical data (when data are available) to arrive at an understanding of the waste types and quantities now in each tank. Some of the more important historical information requirements are discussed below in Section 8.1.1. Section 8.1.2 discusses the use of sampling information to help establish waste types and volumes.

8.1.1 Process History

A detailed knowledge of the waste transactions for each of the 177 tanks is essential in determining potential waste types in the tanks.

Detailed information about each waste transaction for a tank should include the following:

- Date of each transaction--year and quarter of year, i.e., first, second, etc.
- Type of transaction, i.e., into the tank or out of the tank

- Type and source of material transferred, e.g., process high-level waste, process cladding waste, water, supernatant liquid from another tank, etc.
- Quantity of material transferred, i.e., total volume and volume of any associated solids, if known
- Physically measured volume of solids and liquids in tank as a function of time.

Four references provide much of the details on waste types, waste volumes, and waste transactions. Anderson (1990) provides much transaction information, although in somewhat abbreviated form, up through 1980 for all 177 tanks. Hanlon (1997) frequently updates the status of each of the 177 tanks and provides a data base for establishing the amounts of different wastes in each tank. The Sort on Radioactive Waste Type Model (SORWT) (Hill et al. 1995) prioritizes the major waste types transferred through the tanks. However, no distinct volumes are assigned to the predicted waste types; only a qualitative assessment of the relative contribution of the waste type to the total waste volume is made.

The most complete and authoritative compilation of Hanford waste tank transactions is the WSTRS assembled at the Los Alamos National Laboratory (Agnew et al. 1997b). The WSTRS report builds on the information in Anderson (1990), but lists many more waste transactions for each tank. For each waste transaction, WSTRS systematically lists in condensed format the essential details of each transaction.

The WSTRS report should be used with appropriate caution, in performing an assessment of the major waste types (layers) and associated volumes for each of the 177 tanks. As noted by Agnew et al. (1997b), details of some historical transactions, particularly those involving transfer of wastes to and from evaporators, are known to be incomplete. Beyond this failing, some internal discrepancies and uncertainties are evident in the measured and/or estimated amount of solids in each waste slurry transferred, and in the type and volume of solids now reported to be present in each tank assessment.

The waste transaction information from Agnew et al. (1997b) is used to provide an estimate of waste types and associated volume of each solid waste type presently in the tank. This model, known as the TLM, is derived from WSTRS and is documented as part of the HDW model (Agnew et al. 1997a). The estimate of waste types and volumes from the TLM is generally used as a basis for comparison to other information sources that can be used for estimating tank waste types. The transaction information in WSTRS also provides a basis for the Supernatant Mixing Model (SMM). The SMM calculates the composition of supernatant concentrates within each tank. The SMM is also documented as part of the HDW model.

In the process of evaluating WSTRS, differences in assumptions and interpretation may lead to a conclusion regarding the tank contents that differs from the waste layers estimated by the TLM, or supernatants estimated by the SMM. This is generally the result of the waste transaction uncertainties and discrepancies in the WSTRS noted earlier. In one typical example, records show REDOX process cladding waste entering the tank from the REDOX

facility. However, solids level measurement records and waste type description records are either missing, misleading, or inaccurate. The TLM in many cases must make judgment of this incomplete or inaccurate historical information and, in this example, may not predict any cladding waste in the tank.

Other historical process records and documents should be used whenever possible to supplement interpretation of waste transaction records, and ultimately determine what waste types are in a tank. As an example, Rodenhizer (1987) provides evidence that metal waste (MW) in several tanks was essentially completely sluiced from a tank for processing to recover uranium. The WSTRS (and TLM) often credit little or no removal of MW by sluicing operations.

Sometimes the process campaign histories for some facilities such as the PUREX plant are known well enough to estimate quite accurately the waste types and volumes (and associated chemical masses) that entered specific tanks. For example, the process history combined with the flowsheet basis for high-level Zirflex process neutralized current acid waste from the PUREX facility is known well enough to predict the masses and volumes for this waste type in tanks 241-AZ-101 and 241-AZ-102. Similar assessments can be made to predict waste type and volume estimates for PUREX process cladding waste in tank 241-AW-103 and 241-AW-105 and for BiPO_4 process 224 waste in the 241-B-200 series tanks. Tank volume measurements are of course useful for confirming/enhancing the process history knowledge for these tanks and determining relevant changes in volume over time.

8.1.2 Waste Sample Analytical Information

The waste in many of the 177 tanks has been sampled and analyzed. Various sampling techniques have been used: core samples, auger samples, grab samples, etc., depending on the physical condition of the wastes and the type of data needed. The completeness and quality of the samples as well as the analytical data ranges from excellent to poor. Tank samples are often highly useful for estimating the tank waste chemical and radionuclide inventory (see Section 8.2), and also in establishing/confirming the waste types and volumes estimated by other process history sources (e.g., the TLM, Hanlon 1997, and Hill et al. 1995). Samples that provide a composition profile of the waste (e.g., core samples with segment level analysis, or grab samples taken at different depths) provide the most useful information. Additionally, samples taken from more than one position in the tank (different risers) provide a basis for estimating spatial variability, thus enhancing the statistical basis.

The best-basis inventory for tank 241-B-108 provides an example that utilized sample information to verify the waste type in the tank and to supplement and complement the historical basis. Core sample segment data for tank 241-B-108 were used to (1) verify if the process waste types predicted by (Agnew et al. 1997a) are present and (2) estimate the volume of these waste types versus the TLM estimate. The core samples for this tank not only provided a supplemental basis for predicting the waste types and volumes, but more importantly, also established strong evidence regarding the presence of cladding waste solids

at the tank waste surface. The TLM in Agnew et al. (1997a) does not predict this waste type in the tank, although the transaction records (Agnew et al. 1997b) indicate transfers of cladding waste into the tank. Core sample segment level photographs in combination with the segment level analyses for the two core samples enabled an estimate of the depth (and volume) of the cladding waste layers.

Other knowledge of process history can often be combined with tank sample analytical information to help establish and verify waste types and volumes. In particular the flowsheet basis for the process that generated the waste should be reasonably well known. For example, comparison of the analytes found in the sample with the "fingerprint" analytes for a particular flowsheet (e.g., particular analytes within a specific concentration range, such as Bi from the BiPO_4 process or high Al concentrations in cladding waste) is necessary to confirm the basis for the waste types. Flowsheet data are important not only as the basis for establishing the waste types in a tank, but also for predicting analyte inventories. This latter use is addressed in Section 8.2.

8.1.3 Surveillance Information

Tank waste level measurements are an important supplement to the process history evaluation and tank samples for estimating waste types and volumes. Tank waste level measurements are taken in the Hanford tanks to monitor for leaks and intrusions. Recently, these measurements have also been used to estimate retained gas volume. Four devices are used widely in these tanks. Three of these measure the level of the waste surface: they are the Food Instrument Company (FIC), manual tape (MT), and ENRAF (ENRAF is not an acronym, but the capitalized name of the manufacturer) devices. The fourth device measures from within a well embedded in the waste, thereby monitoring the liquid level even if the liquid level is below a dry waste crust. This measurement is referred to as Neutron ILL (for neutron count-based interstitial liquid level). Typically, only one of the three surface level instruments is used in a tank for any extended time period. The FIC and ENRAF level measurement devices are the most accurate of the four. The current deployment of these devices in the Hanford tank farms is described for each tank in Hanlon (1997). The information in Hanlon (1997) serves as an important basis for comparing with process history (Section 8.1.1) and sample data (Section 8.1.2) to estimate waste volumes for the inventory evaluation process.

8.2 APPROACH FOR ESTIMATING WASTE INVENTORIES

Once the basis for the waste types and volumes have been developed (Section 8.1), the next step is to estimate the best-basis chemical and radionuclide inventory for the tanks based on an evaluation of all information sources.

Two key sources of information for calculating tank waste inventories are: (1) analytical data from samples taken from the tanks, and (2) the HDW model (Agnew et al. 1997). The HDW model estimates the inventory for tanks using primarily process history rather than sample data as the basis. These information sources are reviewed as part of the evaluation process.

An engineering assessment is also performed, however, to predict the waste inventory. The engineering assessment is an "independent" method for predicting the inventory in a tank based on: (1) process history (e.g., process flowsheets and process throughput) and/or (2) tank sample analyses from other tanks known to contain the same waste type as the tank being evaluated. The process history information assessed as part of the independent engineering evaluation may use different input assumptions than those used by the HDW model (e.g., flowsheet assumptions, etc.).

To determine the best-basis inventory, the engineering assessment-based inventory prediction is compared to and reconciled with the sample-based inventory (if available) for the tank being assessed and it is also compared to and reconciled with the HDW model estimates for the tank. Reasons for the discrepancies are identified and discussed, and a decision is made regarding the most reasonable set of data and assumptions leading to a best-basis. The logic for this process is shown in Figure 8.2-1. A further discussion of the reconciliation process is described in Section 8.2.5.

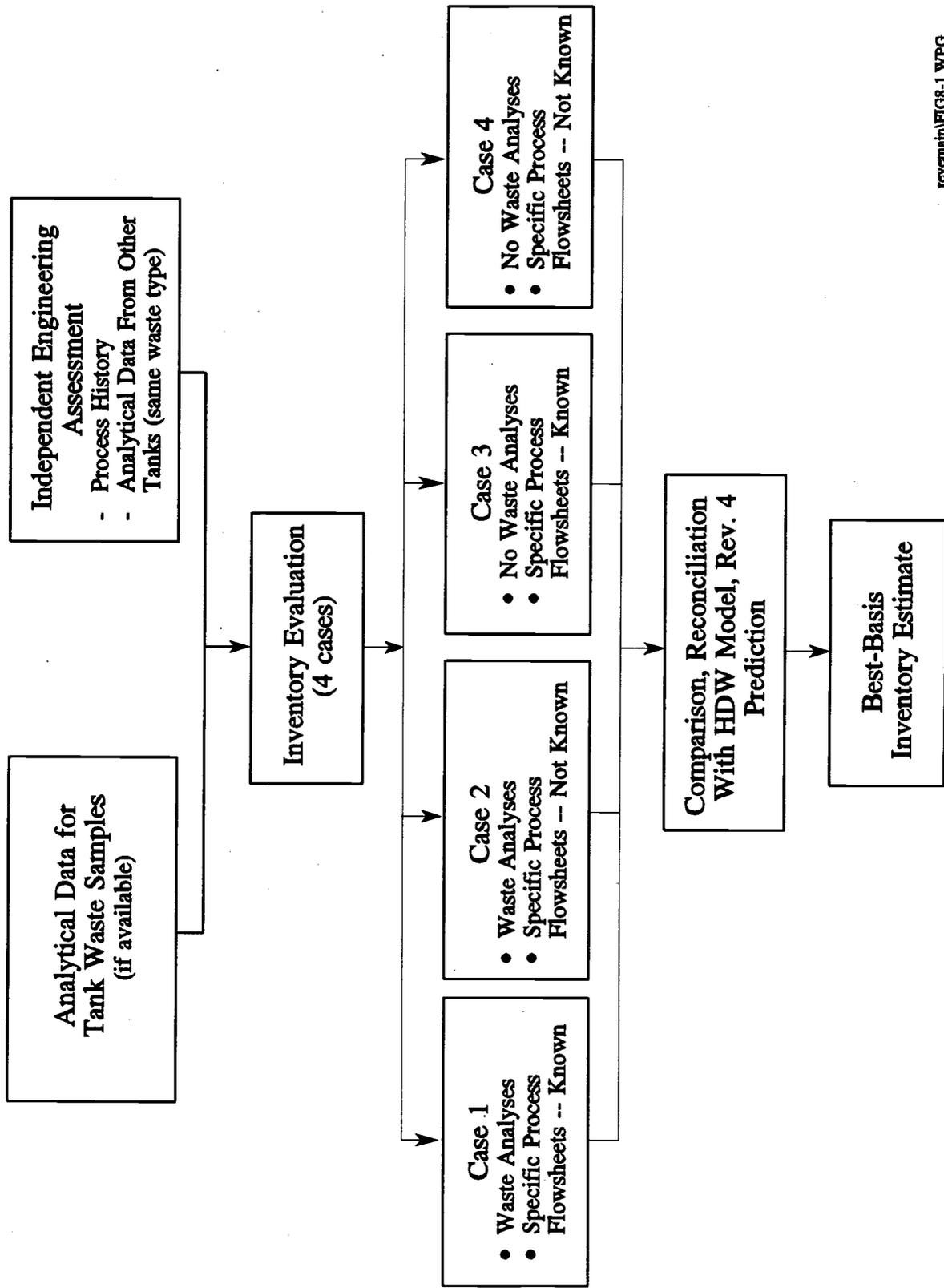
Depending on the nature and amount of the input data available for use (specifically process history and tank waste analyses) the method for deriving waste inventory estimates follows (corresponds to) one of four cases, as indicated in Figure 8.2-1.

8.2.1 Case 1: Waste Analyses Available and Process Flowsheets Known

This case is the most favorable for reliably estimating analyte inventories. The most optimistic scenario would include both high quality analytical data (e.g., multiple core samples), good recovery and complete analyses with low Relative Standard Deviation (RSD) of mean for the tank being evaluated; and also high quality analytical data for other tanks known to contain the same waste type(s).

If the chemical (flowsheet) composition used by the processing facility is known (e.g., which REDOX process flowsheet was used), this flowsheet information can be used to determine whether the sample data are consistent with the flowsheet basis. If there is consistency, this provides high confidence that the sample-based and/or engineering assessment-based inventory can be used as a best basis for the tank. The waste composition should reflect the flowsheet basis that generated the waste; i.e., the "fingerprint" analyses from a waste stream that was transferred from the processing plant should be seen in the sample. In particular, the presence of these analytes at certain concentrations should reflect the process flowsheet composition; e.g., iron, bismuth, manganese, etc. that are believed to have completely precipitated when acidic high-level wastes were made alkaline.

Figure 8.2-1. Waste Inventory Evaluation.



revcmain\FIG8-1.WPG

If the sample data are consistent with the flowsheet, the composition of the waste or waste layers in these tanks can often be used as "basis" tanks for a waste type. The composition of "basis" tanks can be used to estimate the waste composition in other tanks that have no samples, but are expected to contain the same waste type as the basis tank.

Tank samples that form a good basis for a waste type are best obtained from tanks with uncomplicated process histories (e.g., BiPO₄ process 224 waste and UR waste are the only waste types found in tanks 241-B-201 and 241-TY-105, respectively) or from tanks with core samples that show clearly distinguishable defined layers based on segment level analyses. Tanks 241-T-104 and 241-T-107 represent "basis" tanks for BiPO₄ process 1C waste since the process history shows these tanks (which were both the first tank in a three tank cascade) to receive only 1C waste. Substantial core sample information is available that agrees closely with the solids composition to the known flowsheet used.

This methodology is applicable to waste types resulting from separations processes with a known flowsheet basis. It is not applicable to salt cake waste and some supernatants since those waste types do not have a specific flowsheet basis. These wastes typically result from evaporation of supernatants from mixed waste types with variable compositions. The method generally used for deriving inventories for these waste types is described in Section 8.2.2.

8.2.1.1 Concentration Factor Approach for Determining Consistency of Samples With Flowsheet. A semi-quantitative approach can be used to determine to what degree a sample represents the flowsheet basis. A method that is often used is the "concentration factor approach." Concentration factors (CFs) are a mean of reconciling process flowsheet information and sample-based information for particular waste types. The CF is derived by dividing the concentration of a component found in the tank samples by the concentration of the same component in the neutralized process waste stream (i.e., flowsheet concentrations).

Appendix C of this report and Schneider (1951) shows the flowsheet concentrations for the BiPO₄ process 1C waste. The relative concentrations of components expected to completely (i.e., 100 percent) precipitate, (e.g., iron, bismuth, manganese, zirconium, etc.) should be approximately proportional to the respective flowsheet concentrations for those components (i.e., those components should have nearly the same CF). When this latter condition is true, the general conclusion is that sample data are consistent with results calculated from published chemical flowsheets and thus can be considered to correctly represent the tank contents for that waste type. Since the CFs for a particular waste type are often consistent from tank to tank whenever the waste type is present, inventories of components such as iron, bismuth, etc., in tanks which have not been sampled can be estimated from CF values if it is known that its defined waste type is present and that the volume is known or can be estimated.

The following example was used to calculate the CF for Bi in tank 241-BX-112 1C/CW waste. The analytical-based inventory for Bi is 14,200 kg (Winkelman 1996) which corresponds to a Bi concentration in the as settled and sampled solids of 0.110M. The flowsheet concentration for Bi is 0.0115M in the 1C/CW slurry (Appendix C). The CF_{Bi} is:

$$\frac{0.110 \text{ moles Bi/L}}{0.0115 \text{ moles Bi/L}} = 9.5$$

Calculated CFs for components in 1C waste for tanks 241-BX-112, 241-T-104, and 241-T-107 that are expected to fully precipitate, range from 9 to 12 (Bi, Si, Zr, Ce, and Cr). For the most part, these CFs are considered quite consistent which indicates that the samples likely represent the 1C flowsheet (Appendix C) basis for the wastes. The average concentrations of these components were used to calculate the inventories for the 1C waste component in tank 241-BX-110 which did not have a sample.

Once the CFs for fully precipitated components for a waste type are determined, the sample analysis can be used to establish how other components such as SO_4 and PO_4 partition between solids and supernatants. Concentration factors for components not expected to precipitate 100 percent (e.g., PO_4) can be ratioed to components that fully precipitate such as bismuth to obtain the partitioning factors (PFs) for those components. The PF for any component N, thus defined as CF_N/CF_{Bi} , is the fraction of N partitioned to the sludge.

$$\text{Thus the PF for } PO_4(241-BX-112) = \frac{CF_{PO_4}}{CF_{Bi}} = \frac{3.2}{9.5} = 0.34$$

i.e., $0.34 (100) = 34$ percent of the PO_4 partitions to the solids.

The calculated PFs for the basis tanks should provide significant confidence that the analytical data for the tanks are quite representative of the tank contents and could be used as a basis for component inventories in other tanks known to contain the same or similar waste types. The PFs should indicate reasonable partitioning of components based on experience and knowledge of the typical chemical behavior of the components in alkaline media. For example, the PFs for some components such as PO_4 can differ for different waste types. However, a valid explanation for this behavior should be noted. For example, the presence of fluoride as well as PO_4 in high concentration sodium salt solutions can result in crystallization of the hydrated sodium fluoride diphosphate salt and a much higher PF than if F was absent.

Once the tank samples have been shown to be consistent with the known process flowsheet for a waste type, the sample-based or engineering assessment-based inventory calculation procedure is as follows:

1. Calculate the mass of waste in a particular tank from the assumed volume (see Section 8.1) and the measured density values.

2. Calculate the tank inventory of various analytes by multiplying analyte concentrations, either $\mu\text{g/g}$ or uCi/g by the waste mass.

8.2.1.2 Process Throughput Method for Estimating Tank Waste Inventory. The total amount of irradiated fuel reprocessed at the Hanford Site is known. Also known from historical records is the number of MT of irradiated uranium processed each month through each reprocessing plant. In certain cases, it is possible to relate the number of MT of uranium processed to process wastes sent to a particular tank or a cascade of tanks.

In many of these latter cases the amount, e.g., kilograms, of process chemicals consumed per MTU processed is also known, if the appropriate process flowsheet is known. In these favorable cases, it is possible to calculate directly the inventory for many flowsheet components, in particular those that fully precipitated when the waste was made highly alkaline, in the waste receiver. This method is thus similar to the "concentration factor approach" (see Section 8.2.1.1). This method, however, cannot be used to accurately predict some components that are soluble (e.g., Na) or that may partition significantly (e.g., Al, Cr) since large amounts of these components are transferred from the tanks. Sample data are often required to determine the partitioning behavior of these components.

Inventories calculated by this independent engineering assessment are typically compared to a sample-based inventory to determine the level of consistency. The best-basis inventory for tank 241-B-111 is an example of this method.

8.2.1.3 Examples of Case 1: Waste Analysis Available and Process Flowsheets Known. There are several inventory assessments that have found tank sample data and the flowsheet basis for the waste sample to be consistent. For these assessments sample data were selected as the best-basis inventory for that tank. Two sub-cases are available for this case.

Case 1a. Analytical data are available for the tank being assessed. These data are shown to be consistent with the process flowsheet and also with sample data for same waste type in other tanks. The best-basis inventory is usually based nearly entirely on the tank samples.

Example--Tank 241-BX-112. Sample basis for 1C waste type shown to be consistent with BiPO_4 first cycle flowsheet using CF method. In addition, 1C waste types for other tanks (241-T-104 and 241-T-107) also were found to be consistent with the flowsheet and with tank 241-BX-112.

Other examples--Tank 241-T-111, (2C waste), tank 241-B-201 (224 waste).

Case 1b. Analytical data are not available for tank being assessed. However, analytical data for these waste types from other tanks are found to be consistent with the flowsheet and are used as basis for tank being assessed.

Example -- Tank 241-B-108. Sample basis for 1C waste types in tanks 241-T-104 and 241-T-107 were found to be consistent with flowsheet from independent engineering assessment. Analyte concentrations for these tanks were used as best-basis for 1C waste in tank 241-B-108. Note: Core samples were obtained for waste in tank 241-B-108; however, there was incomplete sample recovery near the bottom of the tank so sample analyses were not provided for the 1C waste type.

Other examples -- Tank 241-T-105 (1C and 2C waste). Note: Core samples were obtained for waste in tank 241-T-105; however, there was incomplete sample recovery so sample analyses were not provided for any waste types.

8.2.2 Case 2: Waste Analyses Available, Process Flowsheets Not Known

This case is encountered for many tanks. Although the process history may strongly indicate what waste type(s) are in a tank, the process history may not be sufficiently complete to determine what process flowsheet was used to generate the waste. For example, process flowsheet changes, modifications, etc. may not have been documented. In addition salt cake waste types and some supernatants do not have a specific defined flowsheet basis. These wastes typically result from evaporation of supernatant from mixed waste types. Salt cake compositions resulting from evaporation of supernatants in the 242-B, 242-A, 242-T, and 242-S evaporators cannot generally be predicted by a flowsheet composition because of the complicated waste transfer histories and the complex and unpredictable partitioning of soluble process components to the supernatants.

Many DST solids and supernatants also fall in this category. Fortunately, particularly for DST wastes, extensive sampling has been performed for some tanks.

A sufficient number of sample analyses of 242-B and 242-BY Evaporator salt cake and some 242-S Evaporator salt cakes have been performed to provide a reasonable estimate of the inventories for tanks with these waste types. As for Case 1, analysis of these waste types in two or more tanks can be compared for consistency as part of the engineering assessment, followed by comparison and reconciliation with a sample (if available) from the tank being assessed, and also comparison and reconciliation with the HDW model basis.

8.2.2.1 Examples of Case 2: Waste Analyses Available but Process Flowsheets Not Known.

Tank 241-U-106. Analytical data for 242-S Evaporator (period 1) salt cake is available from the tank 241-U-106 core sample. These are compared with analyses for other tanks known to contain the same salt cake type. The analytical consistency for most analytes is such that the tank 241-U-106 analytical data are used as the best basis for this tank.

Other examples: Tank 241-BY-102 (BY salt cake), tanks 241-B-104, 241-B-108 (B salt cake), tank 241-AW-106, and 241-AP-102 (non-complexed supernatants), tank 241-A-101 (A salt cake).

As for case 1b, analytical data may not be available for the tank being assessed. However, sample analyses for the same waste types from other tanks are extrapolated for use as the best basis. Tanks that use extrapolated (engineering assessment-based) data include: 241-SX-114 (REDOX) and 241-BX-111 (BY salt cake).

8.2.3 Case 3: Waste Analyses Not available; Process Flowsheets Known

This case was encountered for only a few tanks. It is similar to that described in Section 8.2.1.1, however, this case assumes no sample data are available for the tank being evaluated, nor from other tanks known to contain the same waste that could be extrapolated to the tank being evaluated. The only method, then, for estimating the tank inventory would require knowledge of flowsheet bases and volumes of waste transferred to the tank; or knowledge of MT of uranium processed and kg chemicals used per MTU from the flowsheet. Although there is a sample basis for nearly all waste types in Hanford tanks, little or no data exist for some minor waste types such as AR (washed PUREX process sludge) or P1 (PUREX HLW in SSTs 1956 through 1962). However, a flowsheet basis for these wastes exist and can be used to predict the inventory.

Examples of Case 3: Tanks 241-A-104 (AR and P1 waste) and 241-A-106 (PUREX sludge sent to B Plant (via sludge dissolution in the 244-AR vault) for Sr recovery and returned to 241-A-106).

8.2.4 Case 4: Waste Analyses Not Available, Process Flowsheets Not Known

This case is encountered primarily in DSTs where some settled solids layers generally coming from evaporator slurry materials have not been sampled. The solids often precipitate from complexed or non-complexed supernatants; or they consist of solids heels from previous transfers to which other waste supernatants have been added. These wastes often resulted from evaporation of mixed waste types and do not have a specific defined flowsheet basis.

It is typical for these tanks that the component concentrations are near saturation, and the reasonable assumption is made that the composition of the waste solids is the same as the supernatant.

Examples of Case 4: Tanks 241-AW-101 and 241-AW-106 (DSSF solids), tank 241-AW-102 (DN solids heel), tank 241-AP-105 and 241-AN-101 (DN solids).

8.2.5 Comparison and Reconciliation of Inventory Assessments

Once an independent engineering assessment has been performed to predict the component inventory for a tank, the engineering assessment-based inventory must be compared to other predicted inventories for the tank. As noted, the other sources for predicting the inventory includes: (1) a sample-based inventory for the tank if sample and analyses are available, and (2) the HDW model predicted inventory. As a result of this reconciliation process, the best-basis inventory is selected.

If a sample-based inventory is available for the tank whose inventory is being evaluated, it should first be compared to the engineering assessment-based inventory. Several different conclusions often can be reached by this comparison. If concentrations/inventories compare well, it can generally be concluded that the comparison samples (engineering assessment samples) and tank sample are likely the same waste layer, and were generated by same or similar flowsheets. If there are differences, then these differences should be reconciled and explained in the text. For example, the tank sample may be biased for several reasons, such as poor recovery, not spatially representative of tank content; or it may represent a different waste layer than predicted (e.g., waste was result of modified or different process flowsheet than indicated). The decision must be made whether the predicted engineering assessment-based inventory or sample inventory may best represent the tank. Thus, the sample may represent one part of the tank contents, and an engineering assessment-based inventory may be required for the remainder.

Comparison of both the tank sample-based inventory and engineering assessment inventory with the HDW model prediction is also important. If the evaluation depicted in Section 8.1 indicates that the waste layers (and volumes) predicted by the TLM and/or SMM are incorrect, a comparison to the HDW model inventory may be impractical. However, if the assumed tank waste types and volumes agree with TLM waste types and volumes, differences between the model basis and sample basis or engineering basis could result from (1) biased sample results or (2) invalid assumptions used by the HDW model. Experience has shown that some waste stream flowsheet compositions ("defined wastes") used by the HDW model differ from those assumed for the engineering assessment. The flowsheet compositions assumed for the best-basis inventories are provided in Appendixes C and D of this report.

In addition, the assumptions in the HDW model for component solubilities often differ significantly with those assumed for the engineering assessment. For example, as noted earlier (Section 8.2.1) the Bi, Fe, Si, and other components in 1C waste are assumed to essentially fully precipitate from acidic 1C solutions that are made alkaline. The sample analyses show that the CFs obtained are very consistent which means that the sample matches this flowsheet assumption. The Agnew et al. (1997a) model, however, assumes that approximately 40 percent of the Bi and 60 percent of the Si is soluble so that sample analyses are often not consistent with the HDW model assumptions.

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A task to be completed in FY 1997 will encompass collection of apparent deficiencies in the HDW model. This task is in support of the Defense Nuclear Facilities Safety Board (DNFSB) milestone 5.6.3.1.i. The title of this milestone is "Update tank contents model or define limitations," due December 1998.

Fiscal year 1998 planning also includes addition of uncertainty estimates to the individual tank inventories using a methodology defined by Pacific Northwest National Laboratory.

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APPENDIX A

SUMMARY OF DATA REQUIREMENTS

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APPENDIX A

SUMMARY OF DATA REQUIREMENTS

In order to scope the Standard Inventory task, an extensive review of the tank inventory data requirements was performed in the first quarter of FY 1996. Existing Data Quality Objective (DQO) documents were reviewed and cognizant personnel were contacted to identify specific data requirements associated with: safety assessments, risk assessments, safety analyses, *National Environmental Policy Act of 1969* (NEPA) documentation, facility permits, waste treatment/disposal system design, development and implementation, etc. As a result of these efforts, a summary of data requirements was compiled and is included in Table A-1.

Table A-1. Summary of Data Requirements. (Sheet 1 of 4)

Analyte/ parameter	Function and operations			Safety/risk									Regulatory	
				Tank waste										
	(1) Transfer	(2) Retrieve	(3,4,5) Pretreat/ dispose	(6, 7, 8, 9) Resolution	(22) PA	(1) Transfer	(13-16) Inhalation	(12) Shielding/ direct dose	(15) External radiation hazard	(14-20) Long-term groundwater	(22) Volatile hazard	(10) Critic- ality	(11) Air	(21) Ground water
Al	x		P, H, L	x									B	
Ag			H										B	IA
As			H, L				x						A(I)	II
B			x	x									B	IA
Ba			P	x			x							
Be			H				x						A(I, II)	
Bi			P, H	x										
Ca			P, H, L	x			x			x			B (as	
Ce			P, H				x							
Cd			L				x						A(I, II)	IA
Cl			P, L	x		x								IB
Co							x							
Cr			P, H, L	x										IA
Cr+3							x						B	
Cr+6				x						x			A(I, II)	
Cs			P										B(as	
Cu			x											IB
F	x		P, L	x			x			x			B	IA
Fe			H, L	x						x		x	B	IB
FeCN/CN formate			x	x			x							
Hg			H				x						B	IA
K			P, H	x						x			B(as	
La			H				x							
Li			x											
Mg			H, L											
Mn			P	x								x	B	IB
Mo			H											
Na			P, H, L	x			x			x		x	B(as	
Nd			H				x							
NH4			H											
Ni			P, H	x								x	A(I)	
NO2	x		P, L	x		x								
NO3	x		P, L	x		x				x		x		IA
OH	x		P	x		x	NaOH							
oxalate				x			x							
Pb			H										A(I, III)	IA
Pd			H											
total P as PO4	x		P, H, L	x									B	
Pt			H										B	
Rh			H										B	
Ru			H											
Sb			H											
Se			H				x							
Si			H				x						B	IA
total S as SO4	x		P, H, L	x								x		IB
Sr			P, H											
Te			H				x						B	
TIC (CO3)	x		P, L	x										
Th			H											
Tl			H				x						B	
TOC	x		P, H, L	x			x							
U (total)			P, H	x		x	x			x			B	
V			H				x							
W													B	
Zn			H	x										IB
Zr			H, L	x									B	

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Table A-1. Summary of Data Requirements. (Sheet 2 of 4)

Analyte/ parameter	Function and operations			Safety/risk									Regulatory			
	(1) Transfer	(2) Retrieve	(3, 4, 5) Pretreat/ dispose	Tank waste									(11) Air	(21) Ground water		
				(6, 7, 8, 9) Resolution	(22) PA	(1) Transfer	(13-16) Inhalation	(12) Shielding/ Direct Dose	(15) External Radiation Hazard	(14-20) Long-term Groundwater	(22) Volatile Hazard	(10) Critic ality				
3-H			x	x												IC
14-C			H,L													
59-Ni											x	x				
60-Co			x	x			x	x			x					
63-Ni				x							x					
79-Se			x		x						x	x				
90-Sr/Y	x		P,H,L	x	x		x	x								IC
93-Zr/m-Nb			H		x						x					
99-Tc			P,L	x	x						x					
106-Ru							x					x				
113-Cd/m													x			
125-Sb			H										x			
126-Sn			H		x		x	x	x				x			B
129-I			P,H,L	x	x						x		x			
134-Cs													x			
137-Cs/Ba	x		P,H,L	x	x		x	x								B(mOH)
151-Sm																
152-Eu																
154-Eu			P,L				x	x								
155-Eu			P,L	x				x								
226-Ra					x						x					IC
227-Ac					x											
228-Ra					x						x					IC
229-Th																
231-Pa																
232-Th			P,H,L													
232-U					x						x					
233-U					x						x			x		
234-U					x						x					
235-U			x		x						x			x		
236-U						x					x					
237-Np			P,H,L	x							x					
238-Pu			P,H,L	x												
238-U			x													
239/240-Pu			P,H,L	x	x	x					x			x		
241-Am	x		P,H,L	x										x		
241-Pu			P,H,L	x										x		
242-Pu			P,H,L													
242-Cm				x												
243-Am																
243-Cm			P,H,L	x												
244-Cm			P,H,L	x												
total alpha				x										x		IC
total beta				x												IC

Table A-1. Summary of Data Requirements. (Sheet 3 of 4)

Analyte/ parameter	Function and operations			Safety/risk									Regulatory		
	(1) Transfer	(2) Retrieve	(3, 4, 5) Pretreat/ dispose	Tank waste									(11) Air	(21) Ground water	
				(6, 7, 8, 9) Resolution	(22) PA	(1) Transfer	(13-16) Inhalation	(12) Shielding/ Direct Dose	(15) External Radiation Hazard	(14-20) Long-term Groundwater	(22) Volatile Hazard	(10) Critical- ity			
Centrifuged solids				x											
Cooling Curve	x														
Corrosivity															
Crystalline Compds			x												IB
Density, bulk				x									x		
Density, liquid				x											
Density, slurry		x		x											
Density, solids				x											
DSC		x		x		x									
Enthalpy (bulk)				x											
Oxides, %			x												
Organic Complexants			x												
Organic separable				x		x									
Organic species				x											
Particle size				x											
pH		x	x			x							x		IB
Residual solids, wt%			x												
Settled solids, vol%														x	
Settling rate				x											
Shear strength		x		x											
Sludge height		x													
Sludge profile		x													
Solids, vol%	x	x				x									
Specific Gravity	x		x			x							x		
Statum I.D.				x											
Temperature		x													
Volatiles Content				x											
Viscosity	x	x		x											
Water, wt %	x		x	x									x		
CO				x											
CO, Vol%				x											
CO2				x											
Hydrogen, Vol%				x											
Methane, Vol%				x											
NH3				x											
NH3, Vol %				x											B
NO				x											
NO2				x											
N2O				x											
n-Dodecane				x											
n-Tridecane				x											
TBP				x											
Acetonitrile															
Ammonia															
Benzene															
1,3-Butadiene															
Butanol															
2-Hexanone															
Methylene chloride															
Propane nitrile															

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APPENDIX B

SEPARATIONS PLANT RECONSTRUCTED FUEL PROCESSING HISTORY

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APPENDIX B

SEPARATIONS PLANT RECONSTRUCTED FUEL PROCESSING HISTORY

The separations plant fuel process history used to develop global inventories is provided in the tables at the end of this appendix.

The separations plant fuel process history provided in this appendix was assembled from three primary sources: (1) The fuel processing history developed by the Hanford Environmental Dose Reconstruction Project (HEDR) (Heeb 1994), (2) dissolver charging logbooks for reduction and oxidation (REDOX) and plutonium-uranium extraction (PUREX) (McDonald 1995a and 1995b), and (3) fuel shipping data. Data for 1944 to 1965 were extracted from the HEDR fuel processing history, data for 1966 to 1972 was compiled from dissolver charging logbooks, and data for 1983 to 1989 were extracted from fuel shipping records.

**B1.0 HANFORD ENVIRONMENTAL DOSE RECONSTRUCTION PROJECT
SEPARATIONS PLANT OPERATING HISTORY DATA**

The HEDR separations plant operating history was primarily reconstructed from the following sources. The history covers separation plant operation from 1944 to 1972. The data is reported as a month by month history of separations plant operation.

1. PCHIVE (Jungfleisch 1992) is a computer file containing information on monthly tons processed and burnup for each reprocessing plant over the life of the plant.
2. Gydesen (1992a, 1992b) are compilations of historical Hanford operating documents containing declassified extracts of from classified notebooks. These references provide information on tons processed, burnup, and cooling time for each month of operation.
3. Jaech (1957) is a computer printout that contains reactor discharge data, tons dissolved and burnup.
4. Roberts et al. (1992) contains cooling time, exposure of the fuel (mega-watt days), tons dissolved, burnup, and the amount of plutonium separated by month over the life of each of the four separation plants.
5. Heeb (1993) contains monthly tons dissolved and cooling times for 1944 through 1947.

6. The Hanford Works Monthly Reports for April, June, August, October, and December 1952 (GE 1952a, 1952b, 1952c, 1952d, and 1953) supply tons, burnup, and cooling times that were missing in the other data sources.

The HEDR Project found that these sources contain a significant amount of overlapping information. For instance, the same quantity might be reported in more than one data source. In those few instances where there were conflicts between the data sources, a choice was made by the HEDR Project to use one value instead of another and the reasons were documented in HEDR project records (Schmitt 1993).

With respect to the present tank inventory task, the HEDR reconstructed fuel processing history provided insufficient detail with respect to:

1. Thoria processing at PUREX
2. Differentiation of N Reactor zirconium clad fuel from single pass reactor aluminum clad fuel processed at PUREX between 1967 and 1972.
3. N Reactor fuel processed at PUREX between 1983 and 1989. (This time period was outside the scope of the HEDR study.)

For this report, the HEDR reconstructed fuel processing history was used as reported for the period between 1944 and 1962. Minor additions were made in the 1963 to 1965 data to reconcile zirconium clad fuel processing at REDOX with values reported by Jenkins and Foster (1978).

B2.0 REDOX AND PUREX DISSOLVER CHARGING LOGBOOK DATA

Two logbooks were obtained, containing dissolver charging data for REDOX and PUREX (McDonald 1995a and 1995b). The logbooks record the individual charges made to the PUREX and REDOX dissolvers for the time period between January 1, 1966, and August 25, 1972, when PUREX was placed on standby. The logbooks include data on type of fuel, separations plant charging date, tons/charge, fuel irradiation level, and cooling age. These records were compiled into a spreadsheet that was independently verified for accuracy by a second person against the logbook.

Some minor additions to the logbook record were made. The Jenkins and Foster (1978) report was used to fill in minor gaps in the information with respect to the identity of some of the fuel charges for REDOX. Missing data for the 1969 PRTR run at PUREX were obtained from Hobart and Larsen (1970). Data on the second PRTR run were obtained from the process flowsheet and the canister loading list (Watrous 1969a and 1969b). Data on

processing of Pu-Al alloy fuels came from the flowsheet, process specifications, and monthly reports (Matheison and Nicholson 1972a and 1972b, and ARHCO 1972)

These data have been summarized into a monthly history at the end of this appendix.

B3.0 PUREX 1983 TO 1989 CAMPAIGN RECORDS

The PUREX Plant was re-started in November 1983 and was operated through October 1989 to process N Reactor fuel accumulated since the previous campaign. Operation during the last year was intermittent due to equipment and safety problems. The last dissolver charge was loaded on December 7, 1988, with the last dissolver cut being cleared from the dissolver on December 24, 1988. Solvent extraction was shutdown in March 1989. PUREX was officially declared shutdown in October 1989. Irradiated fuel shipping records were used to compile a monthly fuel processing history for 1983 through 1989.

B4.0 FUEL PROCESSING PRODUCTION DATA

Aluminum-clad fuel was processed in the B, T, REDOX, and PUREX Plants. Zirconium-clad fuel was processed in the REDOX and PUREX Plants.

B4.1 BISMUTH PHOSPHATE (BiPO₄) PROCESS PRODUCTION DATA

The separations plant processing history compiled by Heeb (1994) from Gydesen (1992c) indicates that T Plant processed 5,034 MTU (natural uranium) between startup in December 1944 and shutdown in February 1956. B Plant processed 2,766 MTU between startup in April 1945 and shutdown in June 1952. All of the fuel was aluminum-clad natural uranium. It is assumed, from fuel fabrication documents, that all of this fuel was 8-in. solid elements.

B4.2 REDOX PLANT PRODUCTION DATA

The REDOX Plant ran from January 1952 until November 1966. Comparison of the data base compiled by Heeb (1994) with that of Roberts et al. (1992), Jenkins and Foster (1978), and McDonald (1995a) indicates that the REDOX Plant processed a total of 19,461 MTU of aluminum-clad fuel. Of the aluminum-clad fuel, 11,609 MTU was natural

uranium and 7,852 MTU was enriched uranium. About 40 percent of the fuel was internal and external (I&E) elements.

The REDOX Plant was used to test the Zirflex process that was later implemented in the PUREX Plant to process N Reactor fuel. The first process tests of zirconium-clad fuel decladding occurred at the REDOX Plant in December 1962. The amount of nonradioactive zirconium used in the tests has not been determined. The tests supported the processing of Plutonium Recycle Test Reactor (PRTR) fuel from 1963 to 1966. Details of the zirconium-clad fuel processing at the REDOX Plant were established from Jenkins and Foster (1978) and by review of the monthly production reports. Table B-1 summarizes the processing of zirconium-clad fuel at the REDOX Plant.

Table B-1. Summary of Zirconium-Clad Fuel Processing at the REDOX Plant.

Calendar year	N Reactor fuel (MTU)	PRTR fuel (MTU)	Shippingport fuel (MTU)
1962	--	cold test	--
1963	--	1.5	--
1964	--	1.5	--
1965	8.0	0.5	--
1966	219.6	1.5	12.4
Total	227.6	5.0	12.4

PRTR = Plutonium Recycle Test Reactor
REDOX = Reduction and oxidation.

B4.3 PUREX PLANT PRODUCTION DATA

PUREX was used to process aluminum clad uranium elements, zirconium clad uranium elements and aluminum clad thoria elements. The PUREX Plant ran from January 1956 until August 1972. PUREX was shut down from September 1972 until November 1983. PUREX then was restarted and ran until October 1989. The reconstructed fuel processing history shows that PUREX processed a total of 65,924 MTU of aluminum-clad fuel prior to the August 1972 shutdown. Of the aluminum-clad fuel, 58,748 MTU was natural uranium and 7,176 MTU was enriched uranium.

Zircaloy-clad fuel from N Reactor was first processed in PUREX in 1967. The reconstructed fuel processing history in Table B-5 shows that PUREX processed a 1,572.3 MT of zirconium-clad fuel prior to the August 1972 shutdown and 3,890.4 MTU of zirconium-clad fuel between 1983 and 1989.

B4.4 PUREX PLANT THORIA PRODUCTION DATA

A plant process test of the thoria flowsheet was carried out in the PUREX Plant in January-February 1965 using 4.5 MT of thoria (ThO_2) to test systems capabilities (Duckworth et al. 1965). Details of the 1966 thoria campaign were reported by Isochem (1967). The thoria campaign began in May 1966 and resulted in the processing of 197 MT of thoria. The PUREX Plant was extensively flushed before and after the thoria campaign to minimize cross contamination of products. Summaries of chemical usage are compiled in the report. Details of the 1970 thoria campaign are reported by Jackson and Walser (1977). The 1970 campaign processed 426 MT of thoria. Summaries of the waste discharges are compiled in the report. Table B-2 summarizes the thoria quantities reported by various sources.

Table B-2. Metric Tons of Thoria Fuel Reported by Various Authors.

Campaign	Campaign reports (MT)	Agnew et al. (1996) (MT)	Roberts et al. (1992) (MT)	McDonald (1995a, 1999b) (MT)
1965	4.5			
1966	197	173	180	197.3
1970	426	354	376.9	426.8

Values reported in the campaign reports reconcile well with the dissolver records compiled by McDonald (1995a and 1995b). The most likely explanation for the discrepancies of the values reported by Roberts et al. (1992) is confusion with respect to the reporting of thorium versus thoria when the data was extracted from the original records. The difference in molecular weight between thorium and thoria could account for the difference reported for the 1970 campaign.

The 354 MT reported by Agnew et al. (1996) may be an interpretive error. This value closely matches the 358 MT of thorium product reported by Jackson and Walser (1977) after taking process losses and material unaccounted for (MUF) differences between reactor records and analytical chemistry based plant records into account.

The dissolver charging records supplemented by the campaign reports are considered to be the most reliable source of data on the mass of thoria fuel processed. Thus, the best estimate of thoria processed is 629 MT.

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B5.0 BEST BASIS SEPARATIONS PLANT FUEL PROCESSING HISTORY

The best basis estimate is compiled from three sources. The HEDR separations plant fuel processing history for 1944 to 1965 as amended by this study is believed to be the best available fuel processing history for this time period. The separations plant fuel processing history developed for the HEDR study presented in Heeb 1994 is in very good agreement with the data reported by Roberts et al. (1992) and Gydesen (1992c). However, after 1965 the HEDR fuel processing history did not include detail with respect to aluminum vs. zirconium clad fuel and could not be reconciled with dissolver logbooks for PUREX and REDOX.

Two dissolver logbooks, retrieved from records storage, were used to compile the 1966 to 1972 fuel processing history. The dissolver logbooks are considered to be a more original source of data than were the records compiled by Roberts et al. (1992) and Gydesen (1992c). The dissolver logbooks were more detailed with respect to fuel type than other records examined. Dissolver logbooks that precede 1966 were apparently destroyed and thus were not available for examination.

Fuel shipping records were used to compile the fuel processing history for 1983 to 1989.

Comparison of the fuel processing data used in the HDW model (Agnew et al. 1996) yielded a number of discrepancies.

The REDOX Process Production data file, PCHIVE, provided by LANL, was compared to production data compiled in other reports. During the 1952 to 1955 time period, the PCHIVE file consistently shows higher annual production values than do the Jenkins and Foster (1978), and Roberts et al. (1992) reports. The difference totals to about 160 MTU. Another discrepancy occurs in 1966; the PCHIVE file indicates 360 MTU less than is shown in the other reports.

The PUREX Plant campaign data for 1956 to 1962, reported by Agnew et al. (1996, page E-1), is about 4,700 MT short of the value shown in the PCHIVE file and in the HEDR separations plant fuel processing history. The PCHIVE file is also missing the PUREX Plant production data for 1972.

B6.0 SEPARATIONS PLANT FUEL PROCESSING HISTORY

Table B-3 summarizes the type and enrichment of fuel processed by the various separations facilities. Approximately 99,000 MT of uranium and 630 MT of thoria were processed by the various Hanford Site separations facilities.

Table B-3. Fuel Types at Separations Facilities.

Separations process	Fuel type				
	Aluminum clad fuel				Zirconium clad fuel
(operating campaign)	Solid elements		I&E elements		
	Natural uranium	Enriched uranium	Natural uranium	Enriched uranium	Uranium
	(MT)	(MT)	(MT)	(MT)	(MT)
T Plant (1944-1956)	5,034				
B Plant (1945-1952)	2,766				
REDOX (1952-1966)	10,718	966	891	6,886	245
PUREX (1956-1972)	15,683		43,065	7,176	1572.3
PUREX (1983-1989)					3,890.4
Thoria Processing at PUREX					
Thoria (1965, 1966, and 1970)	629				

I&E = Internal and external
 PUREX = Plutonium-Uranium Extraction
 REDOX = Reduction and Oxidation.

Table B-4 presents the monthly separations plant processing history for the B, T, and REDOX Plants. Table B-5 presents the monthly process history for PUREX.

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
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Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year									
				Al Clad Depleted		Al Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other	
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
T		44	12	0.00	3.27	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	1	0.00	3.36	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	2	0.00	6.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	3	0.00	6.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	4	0.00	21.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	5	0.00	22.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	6	0.00	14.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	7	0.00	17.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	8	0.00	28.85	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	9	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	10	0.00	49.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	11	0.00	48.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		45	12	0.00	43.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	1	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	2	0.00	22.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	3	0.00	33.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	4	0.00	39.83	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	5	0.00	27.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	6	0.00	16.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	7	0.00	25.86	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	8	0.00	10.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	9	0.00	5.99	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	10	0.00	5.99	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	11	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		46	12	0.00	20.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	1	0.00	21.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	2	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	3	0.00	26.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	4	0.00	23.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	5	0.00	23.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	6	0.00	13.97	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	7	0.00	27.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	8	0.00	22.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T		47	9	0.00	21.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 2 of 13)

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Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year													
				Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other	
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
T 47			10	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 47			11	0.00	22.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 47			12	0.00	23.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			1	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			2	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			3	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			4	0.00	26.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			5	0.00	26.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			6	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			7	0.00	26.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			8	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			9	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			10	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			11	0.00	35.38	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 48			12	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			1	0.00	44.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			2	0.00	41.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			3	0.00	57.15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			4	0.00	22.68	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			5	0.00	19.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			6	0.00	14.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			7	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			8	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			9	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			10	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			11	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 49			12	0.00	29.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
				Sum T Plant 1944 - 1949																							
T 50			1	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			2	0.00	21.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			3	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			4	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			5	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		

HNF-SD-WM-TI-740
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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 3 of 13)

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Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year											
				Al Clad Depleted		Al Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad Natural		Al Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other	
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
T 50			6	0.00	18.14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			7	0.00	34.47	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			8	0.00	44.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			9	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			10	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			11	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 50			12	0.00	38.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			1	0.00	58.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			2	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			3	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			4	0.00	50.80	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			5	0.00	48.99	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			6	0.00	43.55	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			7	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			8	0.00	41.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			9	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			10	0.00	37.19	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			11	0.00	47.17	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 51			12	0.00	36.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			1	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			2	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			3	0.00	44.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			4	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			5	0.00	14.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			6	0.00	6.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			7	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			8	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			9	0.00	2.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			10	0.00	11.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			11	0.00	11.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 52			12	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 53			1	0.00	2.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 53			2	0.00	22.68	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
T 53			3	0.00	19.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 4 of 13)

Date	Material Processed by Month												Material Processed by Year																			
	Plant	Year	Month	Al Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad										
				Depleted	Natural	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	Other	MTU									
T	53	4	0.00	15.42	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	5	0.00	15.42	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	6	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	7	0.00	15.42	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	8	0.00	16.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	9	0.00	21.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	10	0.00	15.42	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	11	0.00	20.87	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	53	12	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	1	0.00	33.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	2	0.00	29.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	3	0.00	26.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	4	0.00	28.12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	5	0.00	75.30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	6	0.00	90.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	7	0.00	81.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	8	0.00	90.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	9	0.00	95.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	10	0.00	117.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	11	0.00	117.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	54	12	0.00	137.89	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
Sum T Plant Triple Dip Fuel												0.00	924.43	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T	55	1	0.00	140.62	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	2	0.00	140.62	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	3	0.00	57.15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	4	0.00	71.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	5	0.00	133.36	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	6	0.00	145.15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	7	0.00	90.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	8	0.00	98.88	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	9	0.00	113.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	10	0.00	78.93	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	11	0.00	42.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										
T	55	12	0.00	35.38	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00										

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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 5 of 13)

Date	Material Processed by Month												Material Processed by Year													
	Plant	Year	Month	Al Clad		Zr Clad		Zr Clad		Zr Clad		Al Clad		Al Clad		Zr Clad		Zr Clad		Zr Clad						
				Depleted	Natural	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	Natural	0.947%	Natural	0.947%	1.25%	1.25%	Other	Other					
MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU						
T	56	1	0.00	16.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
T	56	2	0.00	24.49	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
			Sum T Plant 1950 - 1956																							
			Total T Plant Fuel Processing																							
B	45	4	0.00	11.07	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	5	0.00	23.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	6	0.00	16.51	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	7	0.00	19.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	8	0.00	26.76	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	9	0.00	34.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	10	0.00	56.43	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	11	0.00	42.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	45	12	0.00	40.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	1	0.00	33.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	2	0.00	43.09	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	3	0.00	49.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	4	0.00	38.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	5	0.00	56.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	6	0.00	41.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	7	0.00	44.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	8	0.00	54.89	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	9	0.00	35.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	10	0.00	43.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	11	0.00	43.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	46	12	0.00	33.93	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	1	0.00	25.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	2	0.00	21.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	3	0.00	36.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	4	0.00	26.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	5	0.00	26.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
B	47	6	0.00	27.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					

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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 6 of 13)

Date	Material Processed by Month												Material Processed by Year															
	Plant	Year	Month	Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
B	47		7	0.00	28.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	308.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	47		8	0.00	23.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	47		9	0.00	27.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	47		10	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	47		11	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	47		12	0.00	20.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	308.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	48		1	0.00	29.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		2	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		3	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		4	0.00	24.49	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		5	0.00	16.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		6	0.00	35.38	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		7	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		8	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		9	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		10	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		11	0.00	35.38	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	48		12	0.00	41.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	361.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	49		1	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		2	0.00	11.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		3	0.00	50.80	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		4	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		5	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		6	0.00	20.87	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		7	0.00	18.14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		8	0.00	9.07	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		9	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		10	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		11	0.00	23.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	49		12	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	322.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
			Sum B Plant 1944 - 1949																									
B	50		1	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1780.46	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
B	50		2	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
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Date	Material Processed by Month												Material Processed by Year																					
	Plant	Year	Month	Al Clad		Zr Clad		Zr Clad		Zr Clad		Al Clad		Al Clad		Al Clad		Zr Clad		Zr Clad		Zr Clad												
				Depleted	Natural	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	Natural	0.947%	1.25%	Natural	0.947%	1.25%	Other	Spike	Other	Spike	Other										
MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU												
B	50		3	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		4	0.00	20.87	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		5	0.00	29.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		6	0.00	27.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		7	0.00	36.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		8	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		9	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		10	0.00	36.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		11	0.00	44.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	50		12	0.00	50.80	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		1	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		2	0.00	44.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		3	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		4	0.00	42.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		5	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		6	0.00	19.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		7	0.00	14.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		8	0.00	39.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		9	0.00	32.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		10	0.00	41.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		11	0.00	48.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	51		12	0.00	33.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		1	0.00	38.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		2	0.00	36.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		3	0.00	20.87	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		4	0.00	6.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		5	0.00	1.81	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
B	52		6	0.00	3.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00											
Sum B Plant 1950 - 1956												0.00	417.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Total B Plant Fuel Processing												0.00	417.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sum B Plant Triple Dip Fuel												0.00	417.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 9 of 13)

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Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year									
				Al Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad	
				Depleted	Natural	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%	0.947%	1.25%
MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU		
Redox	54	12		0.00	195.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	1		0.00	33.36	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	2		0.00	164.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	3		0.00	222.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	4		0.00	170.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	5		0.00	232.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	6		0.00	133.20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	7		0.00	144.41	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	8		0.00	207.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	9		0.00	281.36	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	10		0.00	256.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	11		0.00	214.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	55	12		0.00	138.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
				Redox Triple Dip Fuel										Total Triple Dip Fuel									
Redox	56	1		0.00	104.74	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	2		0.00	224.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	3		0.00	166.43	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	4		0.00	181.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	5		0.00	194.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	6		0.00	149.30	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	7		0.00	54.28	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	8		0.00	95.21	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	9		0.00	218.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	10		0.00	252.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	11		0.00	252.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	56	12		0.00	224.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	1		0.00	73.28	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	2		0.00	102.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	3		0.00	81.55	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	4		0.00	140.68	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	5		0.00	134.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 10 of 13)

Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year																
				Al Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Al Clad		Al Clad		Al Clad		Al Clad		Zr Clad		Zr Clad		Zr Clad		
				Depleted	MTU	0.947%	MTU	1.25%	MTU	Spike	MTU	Other	MTU	Depleted	MTU	0.947%	MTU	1.25%	MTU	Spike	MTU	Other	MTU	Depleted	MTU	0.947%	MTU	1.25%	MTU	Spike
Redox	57	6		0.00	90.86	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1658.82	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	57	7		0.00	155.85	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	57	8		0.00	138.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	57	9		0.00	199.09	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	57	10		0.00	164.78	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	57	11		0.00	147.97	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	57	12		0.00	229.32	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1658.82	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	58	1		0.00	70.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	2		0.00	133.70	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	3		0.00	93.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	4		0.00	80.32	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	5		0.00	190.85	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	6		0.00	184.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	7		0.00	188.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	8		0.00	108.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	9		0.00	129.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	10		0.00	12.37	8.26	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	11		0.00	0.00	6.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	58	12		0.00	0.00	68.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1191.66	82.36	0.00	0.00	0.00	0.00	0.00	0.00
				Sum REDOX 1952 - 1958																										
Redox	59	1		0.00	0.00	62.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	10645.83	82.36	0.00	0.00	0.00	0.00	0.00	0.00
Redox	59	2		0.00	0.00	48.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	3		0.00	0.00	45.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	4		0.00	72.19	29.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	5		0.00	7.54	38.97	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	6		0.00	0.00	70.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	7		0.00	31.63	51.53	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	8		0.00	0.00	60.12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	9		0.00	0.00	58.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	10		0.00	0.00	67.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	11		0.00	34.07	53.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00									
Redox	59	12		0.00	92.51	4.79	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	237.95	593.57	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60	1		0.00	0.00	82.15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
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Date	Material Processed by Month												Material Processed by Year															
	Plant	Year	Month	Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
Redox	60		2	0.00	0.00	46.55	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	235.13	520.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		3	0.00	0.00	39.12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		4	0.00	0.00	66.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		5	0.00	0.00	33.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		6	0.00	137.48	10.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		7	0.00	61.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		8	0.00	36.62	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		9	0.00	0.00	19.41	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		10	0.00	0.00	69.84	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		11	0.00	0.00	87.19	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	60		12	0.00	0.00	66.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	235.13	520.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		1	0.00	0.00	61.47	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		2	0.00	0.00	76.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		3	0.00	0.00	85.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		4	0.00	0.00	98.07	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		5	0.00	0.00	85.21	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		6	0.00	0.00	152.19	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		7	0.00	0.00	123.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		8	0.00	0.00	114.34	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		9	0.00	0.00	62.61	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		10	0.00	0.00	101.42	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		11	0.00	0.00	124.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	61		12	0.00	0.00	60.17	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		1	0.00	0.00	152.09	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		2	0.00	0.65	101.89	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		3	0.00	5.25	63.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		4	0.00	0.00	3.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		5	0.00	0.00	36.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		6	0.00	0.00	80.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		7	0.00	0.00	37.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		8	0.00	0.00	115.78	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		9	0.00	0.00	102.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		10	0.00	0.00	98.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		11	0.00	0.00	50.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	62		12	0.00	0.00	42.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		235.13	520.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		0.00	1144.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		0.00	5.91	886.20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
																		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

HNF-SD-WM-TI-740
Revision 0

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 12 of 13)

6/13/96

Date	Plant	Year	Month	Material Processed by Month										Material Processed by Year													
				Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other		Al Clad Depleted		Al Clad Natural		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad Spike		Zr Clad Other	
				MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
63	Redox		1	0.00	0.00	205.78	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		2	0.00	0.00	63.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		3	0.00	0.00	44.39	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		4	0.00	0.00	167.19	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		5	0.00	0.00	21.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		6	0.00	0.00	117.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		7	0.00	0.00	138.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		8	0.00	0.00	114.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		9	0.00	0.00	75.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		10	0.00	0.00	38.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		11	0.00	0.00	15.29	1.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
63	Redox		12	0.00	0.00	50.97	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		1	0.00	0.00	173.81	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		2	0.00	0.00	154.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		4	0.00	0.00	108.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		5	0.00	0.00	163.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		6	0.00	0.00	71.21	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		7	0.00	0.00	207.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		8	0.00	0.00	76.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		9	0.00	0.00	210.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		10	0.00	0.00	46.85	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		11	0.00	0.00	204.45	1.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
64	Redox		12	0.00	0.00	275.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
				Sum REDOX 1959 - 1964																							
65	Redox		1	0.00	0.00	225.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		2	0.00	0.00	103.18	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		3	0.00	0.00	152.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		4	0.00	0.00	205.24	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		5	0.00	0.00	175.78	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		6	0.00	0.00	0.00	3.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		7	0.00	0.00	79.97	1.98	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
65	Redox		8	0.00	0.00	185.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		

Table B-4. Fuel Processing History for the B, T, and REDOX Plant.
(Sheet 13 of 13)

Date	Material Processed by Month										Material Processed by Year												
	Plant	Year	Month	Al Clad Depleted	Al Clad Natural	Al Clad MTU	Zr Clad 0.947%	Zr Clad MTU	Zr Clad 1.25%	Zr Clad MTU	Zr Clad Spike	Zr Clad MTU	Zr Clad Other	Zr Clad MTU	Al Clad Natural	Al Clad MTU	Zr Clad 0.947%	Zr Clad MTU	Zr Clad Spike	Zr Clad MTU	Zr Clad Other	Zr Clad MTU	
Redox	65	9	0.00	0.00	43.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	65	10	0.00	0.00	90.19	1.60	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	65	11	0.00	0.00	137.45	0.46	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	65	12	0.00	0.00	0.00	0.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.58	1398.58	8.47	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	1	0.00	251.32	0.00	1.45	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	2	0.00	0.00	35.34	9.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	3	0.00	0.00	0.00	53.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	4	0.00	0.00	0.00	46.44	0.00	0.00	0.00	25.66	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	5	0.00	0.00	104.16	0.00	0.00	0.00	0.00	11.68	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	6	0.00	0.00	225.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	7	0.00	0.00	53.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	8	0.00	0.00	6.72	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	9	0.00	212.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Redox	66	10	20.33	0.00	54.45	23.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	20.33	463.57	479.33	178.23	4.04	37.34	13.91	0.00	0.00	0.00
Redox	66	11	0.00	0.00	0.00	44.57	4.04	0.00	0.00	0.00	0.00	0.00	13.91	20.33	464.15	1877.91	186.70	4.04	37.34	13.91	0.00	0.00	0.00
Sum REDOX 1965 - 1966																							
Total REDOX Fuel Processing																							

Notes:

Uranium data on fuel is the pre-irradiation content of ²³⁵U

Natural uranium is 0.72 wt% ²³⁵U

Zirconium clad "spile" fuel elements were N Reactor 0.947-1.25 wt% ²³⁵U elements

Zirconium clad "other" fuel includes PRTR and Shippingport fuel.

Data is from the HEDR inventory (Heeb 1995) with these modifications

Nov. 1964, Added 1.54 MTU PRTR fuel to reconcile with Jenkins and Foster (1978).

Jun. 1965, Changed 3.90 MTU Al clad 0.947% to Zr clad 0.947% fuel to reconcile with Jenkins and Foster (1978)

Jul. 1965, Changed 1.98 MTU Al clad 0.947% to Zr clad 0.947% fuel to reconcile with Jenkins and Foster (1978)

Nov. 1965, Changed 1.60 MTU Al clad 0.947% to Zr clad 0.947% fuel to reconcile with Jenkins and Foster (1978)

Dec. 1965, Added 0.54 MTU PRTR (Zr clad) to reconcile with Jenkins and Foster (1978).

Jan-Dec 1966, Replaced with data from McDonald (1995a)

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 2 of 11)

Date	Material Processed by Month																								
	Al Clad Depleted		Al Clad 0.947%		Al Clad Thoria		Al Clad Other		Zr Clad 0.72%		Zr Clad 0.80%		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad 2.10%		Zr Clad Spike		Zr Clad 0.947/Spike		Zr Clad Other		
	Year	Month	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
58	5	0.0	263.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	6	0.0	99.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	7	0.0	490.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	8	0.0	171.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	9	0.0	477.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	10	0.0	433.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	11	0.0	466.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
58	12	0.0	501.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1958	0.00	4706.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Subtotal	1956 - 1958	0.00	10982.36	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
59	1	0.0	419.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	2	0.0	232.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	3	0.0	457.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	4	0.0	294.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	5	0.0	325.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	6	0.0	476.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	7	0.0	274.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	8	0.0	585.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	9	0.0	373.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	10	0.0	360.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	11	0.0	374.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
59	12	0.0	459.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1959	0.00	4634.80	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
60	1	0.0	557.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
60	2	0.0	269.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
60	3	0.0	301.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
60	4	0.0	617.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
60	5	0.0	299.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
60	6	0.0	223.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 3 of 11)

Date	Material Processed by Month																									
	Al Clad Depleted		Al Clad 0.947%		Al Clad Thoria		Al Clad Other		Zr Clad 0.72%		Zr Clad 0.80%		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad 2.10%		Zr Clad Spike		Zr Clad 0.947/Spike		Zr Clad Other			
	Year	Month	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	
60	7	0.0	388.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
60	8	0.0	690.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
60	9	0.0	687.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
60	10	0.0	382.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
60	11	0.0	658.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
60	12	0.0	521.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
1960		0.00	5598.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
Subtotal																										
61	1	0.0	396.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	2	0.0	348.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	3	0.0	604.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	4	0.0	289.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	5	0.0	676.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	6	0.0	466.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	7	0.0	454.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	8	0.0	516.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	9	0.0	534.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	10	0.0	343.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	11	0.0	203.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
61	12	0.0	602.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
1961		0.00	5437.92	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
Subtotal																										
62	1	0.0	349.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	2	0.0	161.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	3	0.0	217.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	4	0.0	320.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	5	0.0	560.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	6	0.0	528.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	7	0.0	683.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	8	0.0	214.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	9	0.0	505.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
62	10	0.0	646.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 4 of 11)

Date	Material Processed by Month														
	Year	Month	Al Clad Depleted	Al Clad Natural	Al Clad 0.947%	Al Clad Thoria	Al Clad Other	Zr Clad 0.72%	Zr Clad 0.80%	Zr Clad 0.947%	Zr Clad 1.25%	Zr Clad 2.10%	Zr Clad Spike	Zr Clad 0.947/Spike	Zr Clad Other
		MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
1962	62	11	0.0	651.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	62	12	0.0	644.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1962		0.00	5483.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	63	1	0.0	61.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	2	0.0	448.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	3	0.0	646.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	4	0.0	124.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	5	0.0	592.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	6	0.0	599.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	8	0.0	305.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	9	0.0	540.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	10	0.0	404.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	11	0.0	714.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	63	12	0.0	586.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1963		0.00	5024.29	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	64	1	0.0	140.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	2	0.0	99.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	3	0.0	731.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	4	0.0	278.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	5	0.0	426.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	6	0.0	631.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	7	0.0	306.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	8	0.0	761.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	9	0.0	464.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	10	0.0	765.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	11	0.0	510.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	64	12	0.0	39.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1964		0.00	5157.17	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 5 of 11)

Date	Material Processed by Month																						
	Al Clad		Al Clad		Al Clad		Al Clad		Al Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		Zr Clad		
	Depleted	Natural	0.947%	Thoria	Other	0.72%	0.80%	0.947%	1.25%	2.10%	Spike	0.947/Spike	Other	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	
Year	Month	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	
1959 - 1964		0.00	31335.73	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	65	0.0	38.4	0.0	4.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	365.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	773.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	823.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	801.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	538.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	167.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	449.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	438.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	401.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	211.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	65	0.0	374.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1965	0.00	5385.17	0.00	4.54	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	66	0.0	365.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	86.8	304.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	427.3	131.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	167.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	0.0	0.0	59.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	0.0	0.0	134.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	0.0	0.0	4.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	110.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	525.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	844.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	176.4	300.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	66	0.0	86.0	280.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1966	0.00	2789.14	1016.83	197.32	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	67	0.0	661.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

HNF-SD-WM-TI-740
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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 6 of 11)

Date		Material Processed by Month											
Year	Month	Al Clad	Al Clad	Al Clad	Zr Clad	Zr Clad	Zr Clad	Zr Clad					
		Depleted	Natural	0.947%	0.72%	0.80%	0.947%	1.25%	2.10%	Zr Clad Spike	0.947/Spike	Other	MTU
		MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
67	2	0.0	0.0	143.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	3	0.0	375.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	4	0.0	566.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	5	0.0	421.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	6	0.0	0.0	198.5	0.0	0.0	14.8	0.0	0.0	0.0	0.0	0.0	0.0
67	7	0.0	0.0	74.7	0.0	0.0	66.2	0.0	0.0	41.6	0.0	0.0	0.0
67	8	0.0	0.0	305.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	9	0.0	526.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	10	0.0	446.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	11	0.0	475.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
67	12	0.0	0.0	161.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1967	Subtotal	0.00	3474.80	884.27	0.00	0.00	80.99	0.00	0.00	41.59	0.00	0.00	0.00
68	1	0.0	0.0	67.4	0.0	0.0	85.5	0.0	0.0	52.1	0.0	0.0	0.0
68	2	0.0	0.0	148.3	0.0	0.0	17.5	0.0	26.9	0.0	0.0	0.0	0.0
68	3	0.0	0.0	439.7	0.0	0.0	0.0	0.0	60.0	0.0	0.0	0.0	0.0
68	4	0.0	310.5	13.5	0.0	0.0	0.0	0.0	2.9	0.0	0.0	0.0	0.0
68	5	0.0	250.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
68	6	0.0	516.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
68	7	0.0	0.0	139.3	0.0	0.0	0.0	0.0	27.0	0.0	0.0	0.0	0.0
68	8	0.0	0.0	369.7	0.0	0.0	0.0	0.0	60.6	0.0	0.0	0.0	0.0
68	9	0.0	163.1	27.3	0.0	0.0	0.0	0.0	14.5	0.0	0.0	0.0	0.0
68	10	0.0	507.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
68	11	0.0	29.6	253.1	0.0	0.0	0.0	0.0	45.8	0.0	0.0	0.0	0.0
68	12	0.0	0.0	157.1	0.0	0.0	0.0	0.0	34.0	0.0	0.0	0.0	0.0
1968	Subtotal	0.00	1777.84	1615.40	0.00	0.00	103.04	0.00	271.80	52.15	0.00	0.00	0.00
69	1	44.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
69	2	39.4	104.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.5
69	3	0.0	228.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
69	4	0.0	0.0	272.5	0.0	0.0	0.0	0.0	50.8	0.0	0.0	0.0	0.0
69	5	0.0	12.1	324.8	0.0	0.0	0.0	0.0	36.7	0.0	0.0	0.0	0.0

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 8 of 11)

Date		Material Processed by Month																								
Year	Month	Al Clad Depleted		Al Clad 0.947%		Al Clad Thoria		Al Clad Other		Zr Clad 0.72%		Zr Clad 0.80%		Zr Clad 0.947%		Zr Clad 1.25%		Zr Clad 2.10%		Zr Clad Spike		Zr Clad 0.947/Spike		Zr Clad Other		
		MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU	MTU
71	10	0.0	0.0	73.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
71	11	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	103.2	0.0	0.0	0.0	0.0
71	12	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	183.5	0.0	0.0	0.0	0.0
Subtotal		0.00	1522.43	1465.67	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	286.70	0.00	0.00	0.00	0.00
72	1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	83.6	0.0	0.0	0.0	0.0
72	2	0.0	120.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
72	3	0.0	431.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
72	4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
72	5	0.0	0.0	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.1
72	6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
72	7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	65.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
72	8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	162.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal		0.00	551.92	0.00	0.00	0.00	0.27	0.00	0.27	0.00	0.00	0.00	0.00	228.00	0.00	0.00	0.00	0.00	0.00	0.00	102.89	83.62	0.00	0.00	0.00	1.07
Subtotal		84.25	16346.09	7176.42	628.71	0.27	0.27	0.66	0.66	0.00	0.00	0.05	413.16	0.18	0.18	0.00	0.00	0.00	0.00	0.00	196.63	597.36	0.00	0.00	0.00	5.02
83	11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	48.18	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
83	12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	40.88	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Subtotal		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	89.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	112.83	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	41.46	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	112.61	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	133.09	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	87.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	6	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	95.17	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	7	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	144.69	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	8	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	163.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	65.23	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
84	10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 10 of 11)

Date	Material Processed by Month															
	Year	Month	Al Clad Depleted	Al Clad Natural	Al Clad 0.947%	Al Clad Thoria	Al Clad Other	Zr Clad 0.72%	Zr Clad 0.80%	Zr Clad 0.947%	Zr Clad 1.25%	Zr Clad 2.10%	Zr Clad Spike	Zr Clad 0.947/Spike	Zr Clad Other	MTU
87	1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
87	2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	7.33	0.00	0.00	0.00	0.00	0.00	0.00
87	3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	7.34	0.00	0.00	0.00	0.00	0.00	0.00
87	4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	7.37	0.00	0.00	0.00	0.00	0.00	0.00
87	5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	14.67	0.00	0.00	0.00	0.00	0.00	0.00
87	6	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
87	7	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	29.49	0.00	0.00	0.00	0.00	0.00	0.00
87	8	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	58.59	0.00	0.00	0.00	0.00	0.00	0.00
87	9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
87	10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
87	11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
87	12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Subtotal	1987	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	124.80	0.00	0.00	0.00	0.00	0.00	0.00
88	1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	30.20	0.00	0.00	0.00	0.00	0.00	0.00
88	2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	57.37	0.00	0.00	0.00	0.00	0.00	0.00
88	3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	48.22	0.00	0.00	0.00	0.00	0.00	0.00
88	4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
88	5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	9.80	0.00	0.00	0.00	0.00	0.00	0.00
88	6	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	75.98	0.00	0.00	0.00	0.00	0.00	0.00
88	7	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	38.38	0.00	0.00	0.00	0.00	0.00	0.00
88	8	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	48.57	0.00	0.00	46.59	0.00	0.00	0.00
88	9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	68.46	0.00	0.00	24.32	0.00	0.00	0.00
88	10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	19.15	0.00	0.00	32.61	0.00	0.00	0.00
88	11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	18.05	0.00	0.00	40.63	-0.00	0.00	0.00
88	12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	28.35	0.00	0.00	0.00	0.00	0.00	0.00
Subtotal	1988	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	442.54	0.00	0.00	144.15	0.00	0.00	0.00
Subtotal	1983 - 1988	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	3298.94	0.00	0.00	591.46	0.00	0.00	0.00
Total PUREX Fuel Processing		84.25	58664.19	7176.42	628.71	0.27	0.66	0.05	0.18	3712.10	0.00	0.00	788.08	597.36	5.02	0.00

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Table B-5. Fuel Processing History for the PUREX Plant.
(Sheet 11 of 11)

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Date	Material Processed by Month																			
	AI Clad Depleted	AI Clad Natural	AI Clad MTU	AI Clad 0.947%	AI Clad Thoria	AI Clad MTU	Zr Clad 0.72%	Zr Clad MTU	Zr Clad 0.80%	Zr Clad MTU	Zr Clad 0.947%	Zr Clad MTU	Zr Clad 1.25%	Zr Clad MTU	Zr Clad 2.10%	Zr Clad MTU	Zr Clad Spike	Zr Clad 0.947/Spike	Zr Clad MTU	Zr Clad Other

Notes:

Uranium data on fuel is the pre-irradiation content of ²³⁵U

Natural uranium is 0.72 wt% ²³⁵U

Zirconium clad "spike" fuel elements were N Reactor 0.947-1.25 wt% ²³⁵U elements

Zirconium clad 2.10 wt% ²³⁵U elements were N Reactor driver elements for tritium production

Zirconium clad "0.947/spike" is 0.947% and spike fuel processed in combined runs.

Zirconium clad "other" fuel includes PRTR and other miscellaneous fuel.

1956-1965 data is from the HEDR inventory (Heeb 1995) with these modifications

Jan. 1965, Added Thoria data from Duckworth (1965)

1966-1972 data is from dissolver logbooks (McDonald 1995a and 1995b) with these modifications.

Feb. 1969, PRTR fuel determined from Hobart and Larsen (1970).

Feb. 1970, PRTR data established from Waitrous (1969a and 1969b).

May 1972, Pu-AI fuels data from Mathieson and Nicholson (1972a and 1972b) and ARHCO (1972).

1983-1989 data is from fuel shipping records.

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APPENDIX C

BISMUTH PHOSPHATE PROCESS FLOWSHEET

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APPENDIX C

C1.0 BISMUTH PHOSPHATE PROCESS FLOWSHEET

Flowsheet Estimation of Bismuth Phosphate Process Wastes

The waste constituents added by the bismuth phosphate process were estimated from: (1) the Schneider (1951) process flowsheet, (2) the flowsheet contained in the 1944 bismuth phosphate process technical manual (GE 1944), and (3) a revised flow diagram (date unknown) that was contained in the technical manual. The quantities of chemicals added per the flowsheets/flow diagram were calculated in a spreadsheet and compared on the basis of a MT uranium processed (Table C-1). Whenever possible the chemical additions, rather than the waste summary information, were used for these calculations, with the following exceptions: (1) uranium and plutonium losses, (2) the final volume of the wastes streams, and (3) the chemical reactions occurring during reactor fuel decladding and uranium dissolution. The two flowsheets and the flow diagram actually compared quite favorably for major waste components, although the predicted minor components (such as lanthanum, cerium, chromium, and mercury) were generally higher for the Schneider (1951) flowsheet.

The Schneider (1951) flowsheet was issued after seven years of processing experience and is therefore thought to be more reliable than GE (1944). The predicted quantities of minor components are also higher and therefore more conservative for most purposes. The essential material usage and waste constituents predicted for the Schneider (1951) flowsheet are summarized in Tables C-2 and C-3. An additional degree of confidence is provided by actual chemical analyses of the 1C waste stream (Healy 1953). The measured nitrate, sodium, phosphate, sulfate, and fluoride concentrations are almost identical to the spreadsheet predictions based on the Schneider (1951) flowsheet.

The Schneider (1951) flowsheet waste compositions predicted from chemical additions also were compared with the waste summaries provided in the same flowsheet document (Table C-4). The predicted chemical molarities were generally in good agreement with the following exceptions:

<u>Waste Stream</u>	<u>Components</u>
Metal Waste (MW)	NO_3^{-1}
First Cycle Waste (1C)	Fe
	SO_4^{-2}
Second Cycle Waste (2C)	SO_4^{-2}
224 Building Waste	$\text{C}_2\text{O}_4^{-2}$
	Mn
	NO_3^{-1}
	SO_4^{-2}

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Table C-1. Comparative Summary BiPO₄ Waste Additions.
(Based on 7,800 MTU processed)

Component	1951 Flowsheet added (MT)	Tech. manual added (MT)	Flow diagram added (MT)
Al	257	257	248
Bi	563	420	451
C ₂ O ₄ ⁻²	461	228	228
Ce	3.1	2.5	3.0
CO ₃	8,809	9,988	8,588
Cr	47	16	20
F	1,284	967	968
Fe	387	237	234
Hg	1*	0	0
K	995	676	682
La	60	22	22
Mn	32	16	24
Na	24,417	24,970	20,759
NO ₃	28,877	28,713	21,491
NO ₂	754	651	803
Si	180	157	157
PO ₄	8,734	11,315	7,459
SO ₄	4,385	3,410	3,848
Zr	3.1	2.5	4.7

*Per discussions with the flowsheet author, mercuric nitrate was not initiated until sometime in the July 1950 to December 1951 time period. Quantity was reduced from 2.1 to 1 MTU for inventory purposes.

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Table C-2. Essential Material Usage Per Schneider (1951) Flowsheet.

Chemical	Coating waste	Metal waste	1C waste	2C waste	224 waste	Total usage
	Kilograms per MT Uranium					
Al	33					33
$(\text{NH}_4)_2\text{SiF}_6$			75	64		139
$(\text{NH}_4)_2\text{SO}_4$					1.4	1.4
$(\text{NH}_4)_2\text{SO}_3$					0.32	0.32
BiONO_3		30	25	16		70
$(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$				1.6		1.6
$\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$				132	120	252
HF					80	80
H_2O_2			8		10	18
$\text{La}(\text{NO}_3)_3 \cdot 2\text{NH}_4\text{NO}_3 \cdot 4\text{H}_2\text{O}$					31	31
$\text{Hg}(\text{NO}_3)_2$		0.21				0.21
HNO_3		1,947	1,209	765	591	4,512
$\text{H}_2\text{C}_2\text{O}_4$					60	60
H_3PO_4		414	369	332	41	1,155
KOH					179	179
KMnO_4					12	12
Si	1.2					1.2
NaBiO_3			12	7	9	28
Na_2CO_3		1,995				1,995
$\text{Na}_2\text{Cr}_2\text{O}_7$			6	2	7	15
NaOH	84	765	1,121	893	946	3,808
NaNO_3	157					157
NaNO_2		86				86
H_2SO_4		397			0.7	398
$\text{ZrO}(\text{NO}_3)_2$			1.0			1.0

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Table C-3. Bismuth Phosphate Waste Components
Per Schneider (1951) Flow Sheet.

Component	Coating waste	Metal waste	1C waste	2C waste	224 waste	Total	Total added (MT)
	Kilograms per MTU						
Al	33					33	257
Bi		0.08	36	18	18	72	563
C ₂ O ₄ ⁻²					59	59	461
Ce			0.399			0.40	3.1
CO ₃		1129				1,129	8,809
Cr			2.4	0.9	2.8	6.0	47
F			48	41	76	165	1,284
Fe			26	24		50	387
Hg		0.13				0.13	1
K					128	128	995
La					7.7	7.7	60
Mn					4.1	4.1	32
Na	91	1,334	646	514	545	3,130	24,417
NO ₃	47	561	1,268	860	965	3,702	28,877
NO ₂	39	57				97	754
Si	1.2		12	10		23	180
PO ₄		391	361	322	45	1,120	8,734
SO ₄		389	89	81	2.0	562	4,385
Zr			0.40			0.40	3.1

Table incorporates carry-over of HNO₃ and BiPO₄ between cycles, evolution of NO_x during fuel dissolution and reaction of the nitrate ion during fuel decladding.

Table C-4. Bismuth Phosphate Waste Molarities.
(Based on Schneider (1951) flow sheet)

Component	Coating Waste (molar)		Metal (molar)		1C (molar)		2C (molar)		224 (molar)	
	This evaluation ^f	Schneider ^b								
Al	1.17 E+00	1.16 E+00								
Bi			2.46 E-05	0.00 E+00	1.24 E-02	1.24 E-02	6.23 E-03	6.27 E-03	5.95 E-03	5.65 E-03
C ₂ O ₄ ²⁻									4.58E-02	1.47E-02
Ce					2.07E-04	2.14E-04				
CO ₃			1.17 E+00	NA						
Cr					3.30 E-03	3.08 E-03	1.23 E-03	1.15 E-03	3.62 E-03	3.27 E-03
F					1.83 E-01	1.84 E-01	1.54 E-01	1.55 E-01	2.72 E-01	2.95 E-01
Fe					3.39 E-02	2.45 E-02	3.01 E-02	3.26 E-02		
Hg			4.00 E-05	6.67 E-05	NA	1.66 E-05				
K									2.23 E-01	2.18 E-01
La									3.76 E-03	3.53 E-03
Mn									5.14 E-03	6.01 E-03
Na	3.79 E+00	3.83 E+00	3.62 E+00	3.62 E+00	2.05 E+00	2.06 E+00	1.59 E+00	1.60 E+00	1.62 E+00	1.60 E+00
NO ₃	7.34 E-01	7.30 E-01	5.65 E-01	1.56 E-01	1.49 E+00	1.50 E+00	9.87 E-01	9.89 E-01	1.06 E+00	6.84 E-01
NO ₂	8.19 E-01	8.10 E-01	7.79 E-02	NA						
Si	4.10 E-02	4.00 E-01			3.05 E-02	3.06 E-02	2.57 E-02	2.58 E-02		
PO ₄			2.57 E-01	2.65 E-01	2.77 E-01	2.76 E-01	2.41 E-01	2.42 E-01	3.22 E-02	3.21 E-02
SO ₄			2.53 E-01	2.54 E-01	6.78 E-02	4.92 E-02	6.02 E-02	3.76 E-02	1.40 E-03	3.64 E-03
Zr					3.19 E-04	3.29 E-04				
Volumes (gal) from 7,800 MTU	2.14 E+06		3.30 E+07		2.83 E+07		2.90 E+07		3.02 E+07	

^fSpreadsheet based on chemical additions in the Schneider (1951) flowsheet and summarized in this appendix. Use of these values is recommended for performing best-basis tank inventory evaluations.

^bValues from the waste summaries contained in the Schneider (1951) flowsheet.

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The nitrate ion associated with the uranyl nitrate hexahydrate (UNH) was not accounted for in the waste summaries, which explains the difference in the metal waste (MW) nitrate ion. The reasons for the other differences are unknown, but probably the result of simple mathematical errors. The spreadsheet entries/calculations for these stream components were rechecked, and no errors were found.

The waste concentrations predicted from the Schneider (1951) flowsheet were also compared to HDW model values (Agnew et al. 1996) in Table C-5. For comparison with the HDW model values, the coating waste and first cycle wastes were combined, as was the actual practice until late 1954. After December 1954, the two waste streams were sent to separate underground storage tanks to improve the performance of the ferrocyanide scavenging process (Anderson 1990).

The pre/post 1950 HDW model compositions (e.g., MW1 and MW2) for the MW, 1C/CW, and 2C streams were combined into a single value for each waste stream using the waste volumes provided in the HDW document (Agnew et al. 1996)

Overall, the spreadsheet and HDW model concentrations do not compare well. The HDW aluminum concentration in the combined CW/1C stream is roughly a factor of three higher than the Schneider (1951) flowsheet. This difference is likely the result of misinterpreting the basis for the volume of the coating waste. The flowsheet coating waste is based on "6,600 lbs" (3.0 MT) of fuel elements charged, rather than the "2,200 lbs" (1.0 MT) uranium basis used for subsequent fuel dissolution steps. The HDW volume for the 224 Building wastes is also extremely low. This value probably does not account for the supernatant which overflowed the 241-B/T-361 tanks to dry wells or cribs (Anderson 1990).

Other differences between the HDW document and the current spreadsheet calculations are likely the result of differences in source documents used as the bases for calculations. The HDW chemical inventory estimates were based on TRAC (Jungfleisch 1980, 1983, 1984), other waste transaction records (Anderson 1990) and waste composition estimates (Lucas 1989 draft, which for bismuth phosphate processing was based on the 1944 technical manual). Also, the HDW estimates add allowances for corrosion products (Fe and Cr) and calcium impurities in essential materials, both of which are not included in the flowsheets.

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Table C-5. Comparison of Estimated Bismuth Phosphate Waste Molarities.

Component	Metal waste (molar)		1C/CW (molar)		2C waste (molar)		224 waste (molar)	
	This Evaluation ^a	Agnew et al. ^b	This Evaluation ^a	Agnew et al. ^b	This Evaluation ^a	Agnew et al. ^b	This Evaluation ^a	Agnew et al. ^b
Al		0	8.26E-02	2.33E-01		0		0
Bi	2.46E-05	0	1.15E-02	1.40E-02	6.23E-03	6.60E-03	5.95E-03	6.20E-03
Ca		1.80E-02		1.81E-02		1.81E-02		1.82E-02
Ce			1.93E-04					0.00019
Cr		3.20E-03	3.06E-03	5.20E-03	1.23E-03	5.07E-03	3.62E-03	4.10E-03
Fe		1.60E-02	3.15E-02	4.60E-02	3.01E-02	3.18E-02		1.60E-02
Hg	4.00E-05	0		1.07E-05		0		0
K		7.10E-04		3.81E-03		4.49E-03	2.23E-01	2.71E-01
La							3.76E-03	1.50E-02
Mn		0		0		0	5.14E-03	4.60E-02
Na	3.63E+00	2.43E+00	2.17E+00	2.24E+00	1.59E+00	1.55E+00	1.62E+00	1.80E+00
Ni		1.60E-03		1.60E-03		1.60E-03		1.60E-03
Si		4.00E-03	3.12E-02	3.80E-02	2.57E-02	2.44E-02		0
U	2.63E-01	2.41E-01	9.63E-04	7.67E-04	2.41E-05	6.70E-05	0	0
Zr		0	2.96E-04	4.00E-03		0		0
C ₂ O ₄ ⁻²							4.59E-02	3.00E-02
Cl		3.25E-03		7.67E-05		2.07E-02		2.97E-02
CO ₃	1.18E+00	6.57E-01		1.81E-02		1.81E-02		1.82E-02
F		0	1.70E-01	2.28E-01	1.54E-01	1.45E-01	2.72E-01	3.10E-01
NO ₃	5.66E-01	1.85E-01	1.44E+00	5.88E-01	9.88E-01	8.48E-01	1.06E+00	1.58E+00
NO ₂	7.80E-02	0	5.77E-02	1.74E-01		0		0
PO ₄	2.57E-01	1.80E-01	2.58E-01	3.26E-01	2.41E-01	1.39E-01	3.23E-02	4.92E-02
SO ₄	2.53E-01	2.12E-01	6.31E-02	6.16E-02	6.02E-02	3.33E-02	1.40E-03	1.60E-03
Pu(μCi/l)	9.66E+00	4.38E+00	3.86E+01	1.67E+01	1.10E+01	1.29E+01	1.13E+01	1.27E+01
¹³⁷ Cs(Ci/l)		9.27E-04		2.75E-02		2.72E-04		0
⁹⁰ Sr(Ci/l)		8.09E-03		2.43E-04		4.47E-05		0

Volumes (gal) 3.30E+07 3.04E+07 2.90E+07 3.02E+07
from 7,800 MTU
Agnew et al. 3.59E+07 2.83E+07 3.17E+07 8.30E+06
(1996)^b volumes

Conversion factors to convert Agnew results to single concentration:

Waste 1 (1944-1949)	MW1	43%	1C1	42%	2C1	28%
Waste 2 (1950-1956)	MW2	57%	1C2	58%	2C2	72%

Note: The coating wastes were sent to the same underground storage tank as the First-Cycle waste until late 1954.
^aSpreadsheet is based on chemical additions in the Schneider (1951) flowsheet, which is summarized in this appendix (plutonium and uranium are based on flowsheet waste compositions). Use of these values is recommended for performing best-basis tank inventory evaluations.

^bAgnew et al. (1996) *Hanford Tank Chemical and Radionuclide Inventories*, Rev. 3.

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APPENDIX D

REDOX PROCESS FLOWSHEETS

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APPENDIX D

REDOX PROCESS FLOWSHEETS

D1.0 REDOX PROCESS WASTE BACKGROUND

The Reduction and Oxidation (REDOX) Plant underwent several significant equipment and flowsheet changes over the fifteen year operating life. This contrasts with the Bismuth Phosphate Plants (B and T Plants) which ran essentially the same flowsheet throughout their operating lives. Additionally, the REDOX process incorporated waste recycle streams and product rework capabilities. Six REDOX process flowsheets were issued (Nos. 4 through 9) between 1951 and 1966 (GE 1951, Merrill and Stevenson 1955, Crawley and Harmon 1960, Isaacson 1965, Boldt 1966).

Despite the flowsheet changes, there were basically two major waste streams generated by the REDOX process: (1) a coating waste stream with high concentrations of aluminum and sodium and (2) a high-level waste (HLW) stream resulting from solvent extraction operations also with high concentrations of aluminum and sodium, but containing several other chemical components in low concentrations and much higher in volume and radioactivity.

The coating waste stream from aluminum-clad fuel elements had generally the same characteristics as coating waste streams generated at the bismuth phosphate plants (B and T Plants) and Plutonium-Uranium Extraction (PUREX). Only a small quantity (245 MTU) of zirconium-clad fuel elements was processed.

The HLW stream was significantly different from the other fuel reprocessing facilities as the REDOX solvent extraction process used aluminum nitrate nonahydrate (ANN) as a salting agent. Substantial sludge volumes were generated by the precipitated aluminum and other metals and introduced into 46 of the 149 single-shell tanks (SSTs) (Hill et al. 1995). The sludges in these 46 tanks represent 28 percent of the total waste sludge in SSTs. Additionally, sodium dichromate was used as an oxidizing agent, adding significant quantities of chromium to the tank wastes. Chromium is of concern both from an environmental standpoint (*Resource Conservation and Recovery Act of 1976* [RCRA] toxic metal) and interference with proposed waste treatment processes (i.e., glass).

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D2.0 FLOWSHEET ESTIMATION OF REDOX PROCESS WASTES

The flowsheets were first analyzed to confirm an overall material balance and to establish essential material usage rates and waste compositions using a spreadsheet model. The final neptunium decontamination cycle was not included as it occurs only about twice per year and only 11 kL (3 kgal) of feed per cycle is processed. The essential material usage is summarized in Table D2-1. The six flowsheets did in general balance; the stated flowsheet waste stream compositions closely matched that predicted from the chemical input streams. It was not possible to independently estimate the nitrate/nitrite compositions for the coating wastes due to the chemical reactions that are taking place, so the flowsheet values were assumed to be correct for all coating waste. Due to slight discrepancies in material balance and the lack of values for some minor components, the spreadsheet values were presumed to be more correct for the solvent extraction (high-level) waste stream. The waste components are reported on the basis of a metric ton of fuel processed in Table D2-2 and on a concentration basis in Table D2-3. Corresponding values used in the HDW model (Agnew et al. 1996) and in Allen (1976) are included for comparison and further discussed later in this appendix (Sections D3.0 and D4.0 respectively).

Ideally, it would be desirable to assign specific time periods and fuel quantities reprocessed to each of the six flowsheets in order to complete an independent estimate of wastes generated by the REDOX process. The following three significant problems arise when trying to apply the REDOX process flowsheets directly to the prediction of wastes generated:

1. **Non-flowsheet Chemical Usage.** Semiannual Special Nuclear Material (SNM) accountability flushes with concentrated nitric acid solutions would be an example of such a situation.
2. **Process Rework.** Continuous processes such as the REDOX process have provisions for recycle of out-of-specification uranium or plutonium products, recycle of purified uranium to dilute recycled plutonium (criticality prevention) or to keep the solvent extraction systems operating during short disruptions in uranium feed from the head-end operations. The control features of a particular flowsheet can significantly affect these recycle rates.
3. **Flowsheet Implementation Dates.** The date of application of the various flowsheet changes is uncertain. The changes can be made earlier or later than flowsheet issuance. Flowsheet No. 5 in particular was used for project justification, and the ozonation equipment required for implementation was not available until 1-1/2 years later.

Due to the above complications, it was not possible to determine the MTU processed under each of the six flowsheets or to estimate rework values.

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Table D2-1. REDOX Plant Essential Material Usage Predicted from Flowsheets.
(2 Sheets)

Solvent Extraction Essential Material Usage by Flowsheet							
Chemical	REDOX Flowsheet Number						
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 (kg/MTU)	No. 9 (kg/MTU)	
ANN	5,894	1,805	1,358	798	1,169	1,094	
Cr(NO ₃) ₃ · 9H ₂ O	---	---	---	---	---	---	
Fe(NH ₂ SO ₃) ₂	63	6.9	7.1	11	9.1	9.5	
H ₂ C ₂ O ₄	---	3	3	3	3	3	
HIO ₄	---	---	0.031	0.026	0.032	0.032	
HNO ₃	91	68	178	240	202	266	
KMnO ₄	---	---	---	---	---	---	
Na ₂ Cr ₂ O ₇	14	28	18	27	23	22	
NaBiO ₃	---	---	0.052	---	0.052	0.052	
NaNO ₃	107	---	---	---	---	---	
NaOH	3,283	985	1,083	541	681	638	
NH ₂ SO ₃ H	---	2.9	2.5	---	---	---	
Head End Essential Material Usage by Flowsheet							
Chemical	REDOX Flowsheet Number						
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 (kg/MTU)	No. 9-Al (kg/MTU)	No. 9-Zr (kg/MTU)
ANN	---	---	---	---	---	---	700
Cr(NO ₃) ₃ · 9H ₂ O	2.6	---	---	---	---	---	---
Fe(NH ₂ SO ₃) ₂	23	---	---	---	---	---	---
H ₂ C ₂ O ₄	---	---	---	---	---	---	---
HIO ₄	---	---	---	---	---	---	---
HNO ₃	1,537	1,296	657	639	761	733	694
KMnO ₄ *	7	---	---	---	---	---	---
Na ₂ Cr ₂ O ₇	45	56	48	34	44	39	62
NaBiO ₃	---	---	---	---	---	---	---

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Table D2-1. REDOX Plant Essential Material Usage Predicted from Flowsheets.
(2 Sheets)

Head End Essential Material Usage by Flowsheet							
Chemical	REDOX Flowsheet Number						
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 (kg/MTU)	No. 9-Al (kg/MTU)	No. 9-Zr (kg/MTU)
NaNO ₃	166	150	169	135	---	---	---
NaOH	139	124	71	79	90	74	296
NH ₂ SO ₃ H	---	---	---	---	---	---	---
NH ₄ F	---	---	---	---	---	---	244
NH ₄ NO ₃	---	---	---	---	---	---	48
Overall REDOX Essential Material Usage by Flowsheet							
Chemical	REDOX Flowsheet Number						
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 (kg/MTU)	No. 9-Al (kg/MTU)	
ANN	5,894	1,805	1,358	798	1,169	1,094	
Cr(NO ₃) ₃ · 9H ₂ O	2.6	0	0	0	0	0	
Fe(NH ₂ SO ₃) ₂	85	7	7	11	9	9	
H ₂ C ₂ O ₄	0	2.6	2.7	2.7	2.7	2.7	
HIO ₄	0	0	0.031	0.026	0.032	0.032	
HNO ₃	1,628	1,365	835	879	963	999	
KMnO ₄ *	7	0	0	0	0	0	
Na ₂ Cr ₂ O ₇	59	84	65	61	67	61	
NaBiO ₃	0	0	0.052	0	0.052	0.052	
NaNO ₃	273	150	169	135	0	0	
NaOH	3,422	1,109	1,154	621	771	711	
NH ₂ SO ₃ H	0	2.9	2.5	0	0	0	

REDOX = Reduction and Oxidation

*Although not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

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Table D2-2. REDOX Process Waste Generation Predicted from Flowsheets. (2 Sheets)

Solvent Extraction Waste Components by Flowsheet.								
Species	REDOX Flowsheet Number						HDW	HDW
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 (kg/MTU)	No. 9 (kg/MTU)	R1 (kg/MTU)	R2 (kg/MTU)
Al	423	130	98	57	84	79	154	142
Bi	—	—	0.039	—	0.039	0.039	0	0
C ₂ O ₄ ⁻²	—	2.5	2.7	2.7	2.7	2.7	—	—
Cr	24	33	26	24	27	24	31	27
F	—	—	—	—	—	—	0	0
Fe	19	1.5	1.6	2.5	2.0	2.1	23	14
I	—	—	0.021	0.017	0.021	0.021	—	—
K*	1.6	—	—	—	—	—	0	0
Mn*	2.3	—	—	—	—	—	0	0
Na	1955	606	634	322	413	377	594	573
NO ₂	0	0	0	0	0	0	0	0
NO ₃	3108	987	904	657	803	744	1344	1209
Si	0.088	0	0	0	0	0	4	6
SO ₄	66	8.1	8.0	8.8	7.0	7.3	16	13
U	3.5	3.3	0.60	1.1	0.55	0.45	10	10
Zr	—	—	—	—	—	—	0	0
Gal/MTU	3252	984	1005	549	699	644	2321	1234
L/MTU	12308	3723	3802	2080	2647	2437	8787	4669
Issue Date	7/51	8/55	10/60	1/65	1/65	9/66		

HDW = Hanford Defined Waste

REDOX = Reduction and Oxidation

*Although not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

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Table D2-2. REDOX Process Waste Generation Predicted from Flowsheets. (2 Sheets)

Coating Waste Components by Flowsheet									
Species	REDOX Flowsheet Number							HDW	HDW
	No. 4 (kg/MTU)	No. 5 (kg/MTU)	No. 6 (kg/MTU)	No. 7 (kg/MTU)	No. 8 ^a (kg/MTU)	No. 9-Al ^a (kg/MTU)	No. 9-Zr ^a (kg/MTU)	CRW1 (kg/MTU)	CRW2 (kg/MTU)
Al	35	35	39	46	46	46	15	49	20
Bi	—	—	—	—	—	—	—	0	0
C ₂ O ₄ ²⁻	—	—	—	—	—	—	—	—	—
Cr	—	—	—	—	—	—	—	0.14	0.15
F	—	—	—	—	—	—	112	0	0
Fe	—	—	—	—	—	—	1.4	0.8	0.8
I	—	—	—	—	—	—	—	—	—
K ^b	—	—	—	—	—	—	—	0.078	0.12
Mn ^b	—	—	—	—	—	—	—	0	0
Na	96	88	83	82	101	99	247	82	39
NO ₂	45	46	50	47	47	50	21	59	12
NO ₃	82	36	37	11	58	50	220	120	50
Si	0.42	0.42	0.58	1.6	1.6	1.6	—	0.77	0
SO ₄	—	—	—	—	—	—	—	1.1	1.1
U	0.45	0.30	1.0	1.7	1.0	1.0	7.2	4.0	4.0
Zr	—	—	—	—	—	—	77	0	0
Gal/MTU	430	261	198	176	196	196	458	240	248
L/MTU	1629	988	751	668	741	741	1734	909	937
Issue Date	7/51	8/55	10/60	1/65	1/65	9/66	9/66		

HDW = Hanford Defined Waste

REDOX = Reduction and Oxidation

^aIncludes Tank Farm salt solution recycled to provide nitrate ion for Al coating removal or Al fuel bucket dissolution.

^bAlthough not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

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Table D2-3. REDOX Process Waste Concentrations Predicted from Flowsheets. (2 Sheets)

REDOX Solvent Extraction Waste Concentrations by Flowsheet									
Species	REDOX Flowsheet Number						HDW	HDW	Allen (1976)
	No. 4 (molar)	No. 5 (molar)	No. 6 (molar)	No. 7 (molar)	No. 8 (molar)	No. 9 (molar)	R1 (molar)	R2 (molar)	Basis (molar)
Al	1.27	1.29	0.95	1.02	1.18	1.20	0.65	1.13	1.2
Bi	0	0	0.000049	0	0.000070	0.000076	0	0	—
C ₂ O ₄ ⁻²	0	0.0077	0.0080	0.015	0.011	0.012	—	—	—
Cr	0.037	0.17	0.13	0.22	0.19	0.19	0.068	0.11	0.18
F	0	0	0	0	0	0	0	0	—
Fe	0.028	0.0074	0.0075	0.022	0.014	0.016	0.048	0.053	0.016
I	0	0	0.000043	0.000066	0.000063	0.000068	0	0	—
K*	0.0034	0	0	0	0	0	0.011	0.019	—
Mn*	0.0034	0	0	0	0	0	0	0	—
Na	6.9	7.1	7.3	6.7	6.8	6.7	2.9	5.3	6.9
NO ₂	0	0	0	0	0	0	0	0	—
NO ₃	4.1	4.3	3.8	5.1	4.9	4.9	2.5	4.2	4.8
Si	0.00025	0	0	0	0	0	0.015	0.042	—
SO ₄	0.056	0.023	0.022	0.044	0.028	0.031	0.019	0.030	0.031
Issue Date	7/51	8/55	10/60	1/65	1/65	9/66			

HDW = Hanford Defined Waste

REDOX = Reduction and Oxidation

*Although not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

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Table D2-3. REDOX Process Waste Concentrations Predicted from Flowsheets.
(2 Sheets)

REDOX Coating Waste Concentrations by Flowsheet											
Species	REDOX Flowaheet Number							HDW	HDW	Allen 1976	Allen 1976
	No. 4 (molar)	No. 5 (molar)	No. 6 (molar)	No. 7 (molar)	No. 8 ^a (molar)	No. 9-Al ^a (molar)	No. 9-Zr ^a (molar)	CWR1 (molar)	CWR2 (molar)	Basis-Al (molar)	Basis-Zr (molar)
Al	0.80	1.73	1.91	2.56	2.31	2.32	0.33	2.0	0.8	1.2	—
Bi	0	0	0	0	0	0	0	0	0	—	—
C ₂ O ₄ ²⁻	0	0	0	0	0	0	0	—	—	—	—
Cr	0	0	0	0	0	0	0	0.0030	0.0030	—	—
F	0	0	0	0	0	0	3.4	0	0	—	0.071
Fe	0	0	0	0	0	0	0.014	0.015	0.015	—	—
I	0	0	0	0	0	0	0	—	—	—	—
K ^b	0	0	0	0	0	0	0	0.0022	0.0034	—	0.01
Mn ^b	0	0	0	0	0	0	0	0	0	—	—
Na	2.6	3.6	4.8	5.3	5.9	5.8	6.2	3.9	1.8	3.74	0.72
NO ₂	0.60	1.0	1.4	1.5	1.4	1.5	0.26	1.4	0.28	0.9	—
NO ₃	0.81	0.17	0.80	0.26	1.3	1.1	2.0	0.88	0.89	0.6	0.02
Si	0.0091	0.059	0.027	0.088	0.079	0.079	0	0.030	0.000	0.02	—
SO ₄	0	0	0	0	0	0	0	0.013	0.013	—	—
U	0.0012	0.0072	0.0056	0.011	0.0057	0.0057	0.017	0.019	0.018	—	0.0005
Zr	0	0	0	0	0	0	0.49	0	0	—	0.1
Issue Date	7/51	8/55	10/60	1/65	1/65	9/66	9/66				

HDW = Hanford Defined Waste

REDOX = Reduction and Oxidation

^aIncludes Tank Farm salt solution recycled to provide nitrate ion for Al coating removal or Al fuel bucket dissolution

^bAlthough not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

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Despite this limitation, important conclusions can be drawn from the comparison of the REDOX process flowsheets. The flowsheets indicate that the HLW concentrations for major components (Na, Al, and NO_3) were relatively constant, even though the waste volume generation rate (volume/MTU) varied substantially over the plant's operating life. In contrast, the minor components were fairly constant on the basis of a MTU processed.

Actual essential material usage rates are available for aluminum nitrate nonahydrate, nitric acid and sodium hydroxide (Jenkins and Foster 1978). These usage rates were converted to kgs of chemical per MTU processed and compared to solvent extraction wastes predicted from the six flowsheets (Table D2-4). Even though the flowsheet implementation dates are not exactly known, significant process rework is evident.

The waste concentrations for Al, NO_3 , and Na were also calculated from the essential material usage (Jenkins and Foster 1978) and the waste volumes reported for 1955 through 1965 (Roberts et al. 1992) for comparison with the flowsheet waste predictions (Table D2-5). The actual plant data indicates relatively constant waste concentrations for the three major waste constituents (Al, Na, and NO_3), although concentrations for all three species are slightly more dilute than predicted from the flowsheets (Table D2-3). The agreement between the estimated concentrations based on plant data and the flowsheet predictions is good.

From an operating perspective, the concentration of sodium, aluminum, and nitrate ion would be expected to remain fairly constant regardless of the flowsheet being used or the amount of process recycle. The waste concentrator would be operated to a specific gravity, which essentially determined by the nitrate ion concentration since the atomic weights of aluminum and hydrogen are relatively low. Aluminum nitrate nonahydrate contributes roughly 70 percent of the nitrate ion, so its concentration will also be relatively constant. Since four moles of caustic are required to neutralize the aluminum and convert it to sodium aluminate, the caustic demand is also fixed and the waste sodium is also fairly constant. Improvements in plant operating efficiency are reflected in the reduction of waste volume transferred to the underground storage tanks, not a change in concentration for these three components.

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Table D2-4. Actual REDOX Process Essential Material Usage.
(based on Foster et al. 1978)

Year	ANN Consumption (kg/MTU)	HNO ₃ Consumption (kg/MTU)	NaOH Consumption (kg/MTU)
1952 (Last nine months only)	8,700	1,750	4,350
1953	6,750	1,650	3,250
1954	6,100	1,550	3,040
1955	4,150	1,550	2,200
1956	2,800	1,520	1,700
1957	2,150	1,450	1,400
1958	1,800	1,010	1,300
1959	1,550	1,200	1,250
1960	1,600	1,280	1,450
1961	1,200	1,085	1,150
1962	1,400	1,240	1,250
1963	1,200	1,390	1,150
1964	1,450	1,170	1,100
1965	1,475	1,050	1,120
1966	2,000	1,200	1,350

REDOX = Reduction and Oxidation.

Essential Material Usage per REDOX Process Flowsheets
(includes head-end and solvent extraction)

Flowsheet & Issue Date	ANN Consumption (kg/MTU)	HNO ₃ Consumption (kg/MTU)	NaOH Consumption (kg/MTU)
No. 4, 7/51	5,894	1,628	3,422
No. 5, 8/55	1,805	1,365	1,109
No. 6, 10/60	1,358	835	1,154
No. 7, 1/65	798	879	621
No. 8, 1/65	1,169	963	771
No. 9, 9/66	1,094	999	711

Table D2-5. REDOX High-Level Waste Concentrations Predicted from Production Data.

Year	Aluminum concentration (molarity)	Nitrate concentration ^{d,e} (molarity)	Sodium concentration ^{b,c} (molarity)	REDOX production (MTU)	Waste volume (gal/MTU)
1955	1.34	4.68	6.37	2,144	2,187
1956	1.25	4.58	6.72	2,182	1,583
1957	1.17	4.32	6.69	1,659	1,293
1958	1.04	4.11	6.56	1,274	1,218
1959	0.89	4.30	6.25	840	1,227
1960	0.76	3.85	6.05	746	1,485
1961	0.89	4.27	7.37	1,137	950
1962	0.83	4.34	6.48	889	1,182
1963	0.69	4.38	5.75	1,043	1,219
1964	0.85	4.10	5.54	1,605	1,204
1965	1.04	4.49	6.80	1,523	1,000
Average	0.98	4.31	6.42		
Std. Dev.	0.20	0.22	0.49		
Flowsheet and Issue Date					
No. 4, 7/51	1.27	4.07	6.91		
No. 5, 8/55	1.29	4.28	7.08		
No. 6, 10/60	0.95	3.83	7.25		
No. 7, 1/65	1.02	5.10	6.73		
No. 8, 1/65	1.18	4.89	6.78		
No. 9, 9/66	1.20	4.92	6.73		
Average ^a	1.15	4.52	6.91		

REDOX = Reduction and Oxidation

^aThis is a simple arithmetic average of the six flowsheets and is NOT weighted for the actual reactor fuel processed under each flowsheet.

^bSodium concentration includes only NaOH.

^cNaOH was reduced by a nominal 90 kg/MTU used for fuel element decladding.

^dThe nitrate ion reduced to account for NO_x evolved during uranium fuel dissolution: 660 kg/MTU through 1957 when the 293-S NO_x recovery facility became operational, 190 kg/MTU thereafter.

^eThe nitrate ion was also reduced by 521 kg/MTU to account for the nitrate associated with the UNH product.

D3.0 COMPARISON TO THE HDW MODEL

The HDW model (Agnew et al. 1996) divides the solvent extraction (high-level) waste into two time periods: 1952 to 1958 (R1) and 1959 to 1967 (R2). There is a substantial increase in the sodium and aluminum concentrations between the two time periods coupled with a large decrease in the waste volume produced per MTU. The basis for the REDOX solvent extraction process (high-level) waste stream composition and volume is unclear (Agnew et al. 1996). The coating waste composition and volume were based on TRAC (Jungfleisch 1984).

The HDW model R1 and R2 concentrations values for Na, Al, and NO_3 show a sharp increase between the two time periods and a reduction in waste volume per MTU processed. There should be little change between the two time periods. The concentrations for R2 are more reasonable, but are still low for Na and NO_3 . The waste volume generation rates appear reasonable, but the date separating R1 and R2 should be about 1956.

The minor components (such as chromium) unfortunately do not have a constant concentration in the HLW stream (Table D2-3). A review of the flowsheets indicates that these components are more constant on the basis of a MTU processed (Table D2-2). Process rework would increase chemical usage and the waste generation rates above the flowsheet values.

The HDW model divides coating wastes into two time periods: 1952 through 1960 (CWR1) and 1961 through 1967 (CWR2). Although the coating waste volumes per MTU did not change appreciably, the HDW model shows a substantial reduction in the waste concentrations of aluminum and sodium between the two periods. There is no practical justification for this reduction in aluminum as the decladding process did not change appreciably and the flowsheet evaluations summarized in this appendix show that the aluminum concentrations were increasing for REDOX process flowsheets No. 5 through 7 and fairly constant for flowsheets No. 7 through 9 (Table D2-3).

The coating waste volumes for flowsheet No. 4 are inflated by an acid rinse of the fuel elements after decladding (which also reduces the aluminum concentration). The acid rinse was probably not used for a significant time period (if at all) since the acid rinse had already been deleted from the bismuth phosphate flowsheet in 1951 (Schneider 1951).

Tank Farm salt waste solutions were recycled to the REDOX Plant dissolvers to provide the nitrate ion required for coating removal from aluminum-clad fuels beginning with flowsheet No. 8 issued in 1965. Production records show this practice was actually initiated in the first quarter of 1963 (GE 1963) or the third quarter of 1963 (Roberts et al. 1992). Recycle of salt waste would have reduced the net addition of sodium, hydroxide and nitrate to wastes, but this would not explain the drop in the concentrations used in the HDW model.

The coating waste concentrations used in the HDW model also do not reflect the approximately 245 MTU of zirconium-clad fuel elements processed as no zirconium or fluoride is reported.

The HDW model reports the total REDOX Plant production as 19,464 MTU which agrees very closely with the 19,700 MTU estimated in Section 4.2.1 of this report.

D4.0 COMPARISON TO ALLEN 1976

A 1976 estimate of the inventory of chemicals added to the Hanford underground storage tanks includes waste inputs from the REDOX process (Allen 1976). The REDOX process waste estimates were based on a production data notebook (Roberts et al. 1992), REDOX process flowsheet No. 9 (Boldt 1966) for the composition of the HLW and an unknown data source for the composition of coating wastes. The compositions used for the coating wastes (aluminum and zirconium-clad fuel elements) are identical to those reported in Anderson 1990 and may for aluminum-clad fuels be based on the 1951 bismuth phosphate flowsheet (Schneider 1951).

The waste volumes were based in HWN-1991 (Roberts et al. 1992) which covers only SX Tank Farm and does not include the first 3 years of REDOX Plant production. Additionally, sporadic adjustments were made to the "Accum. gal." column to account for transfers of HLW to other tank farms, recalibrations, etc., so the accumulation values used by Allen (1976) are less than the sum of the monthly waste volume transfers from the REDOX Plant. Overall, the volumes used by Allen are probably over 33 percent too low for the HLW and over 20 percent low for coating wastes.

The use of REDOX process flowsheet No. 9 for HLW is a reasonable approximation for major waste components since these were to be used with production waste volumes (Roberts et al. 1992). The use of flowsheet No. 9 would tend to cause an overestimation of minor components (such as chromium or iron) added under the early flowsheets.

The waste compositions assumed coating wastes from aluminum-clad fuel (source uncertain) appear to be roughly 33 percent low when compared to the REDOX process flowsheet values. The compositions are most likely based on the 1951 bismuth phosphate flowsheet (Schneider 1951), which is more dilute than the flowsheets used at either the REDOX or PUREX Plants (with the exception of REDOX process flowsheet No. 4 as discussed previously in Appendix D3.0).

The coating waste composition used for zirconium-clad fuel elements is relatively dilute and does not approximate flowsheet compositions at either REDOX or PUREX Plants. The source of this composition data is unknown. However, only 245 MTU of zirconium-clad fuel were processed at the REDOX Plant.

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APPENDIX E
SUPPORTING DATA FOR ALUMINUM (SECTION 5.1)

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APPENDIX E
SUPPORTING DATA FOR ALUMINUM (SECTION 5.1)

**E1.0 COMPARISON OF HANFORD DEFINED WASTE ESTIMATE
VERSUS STANDARD ESTIMATE**

Table E-1. Comparison of Hanford Defined Waste Estimate
Versus Standard Estimate.

Process	MT aluminum	
	HDW ^a	Standard estimate ^b
BiPO ₄ CW	629	251
TFeCN	33	--
REDOX CW	750	747
REDOX ANN	2,903	4,210
PUREX CW	2,551	2,639
PUREX ThO ₂	47	43
PUREX NPR	39	87
PFP PRF (Z Plant)	85	88
B Plant HLW	87	--
B Plant LLW	851	--
Sum of aluminum inputs	7,975	8,065
Aluminum to ground	886 ^c	220
Remaining tank inventory	7,089	7,845
Aluminum input to balance	629 ^d	--
Reported inventory	7,718	7,845

HDW = Hanford Defined Waste

HLW = High level waste

LLW = Low level waste

PUREX = Plutonium Uranium Extraction

^aThe values in this column are the product of the volumes shown in pages B-2 through B-4 times the concentrations shown in pages B-5 through B-7 of Agnew et al. (1996), e.g., for BiPO₄ CW, the BiPO₄ campaigns shown as 1C1, 1C2, and 1CFeCN are thus evaluated.

^bThese values are from Table 5.1-1 of this document.

^cThis is the sum of the aluminum shown to cribs and from leaks as shown on page E-1a of Agnew et al. (1996).

^dThis additional aluminum input is required to arrive at the value shown (sum of SSTs and DDTs) on page E-1a of Agnew et al. (1996).

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E2.0 CALCULATION OF ALUMINUM INPUT FROM PUREX THORIA RUNS**E2.1 ALUMINUM IN CLADDING**

Data from Jackson and Walser (1977) show 400 lbs Al per 6,430 lb thoria (ThO₂) or 5,660 lbs Th. From Appendix B of this document, the total MT ThO₂ processed = 629 MT. With approximately 5 MT recycled, the total MT dissolved = 624 MT.

$$(400 \text{ lb Al} / 6,430 \text{ lb ThO}_2)(\text{lb}/0.4536 \text{ kg})(0.4536 \text{ kg}/\text{lb}) = 400 \text{ kg Al}/6,430 \text{ kg ThO}_2$$

$$= 400 \text{ MT Al}/6,430 \text{ MT ThO}_2 = 6.22 \text{ E-2 MT Al}/\text{MT ThO}_2$$

$$(6.22 \text{ E-2 MT Al}/\text{MT ThO}_2)(624 \text{ MT ThO}_2) = 38.8 \text{ MT Al in Cladding}$$

E2.2 ALUMINUM IN FLUSHING/PROCESSING

Isochem (1967) gives data for ANN usage in flushing and processing for the 1966 PUREX Thorium/²³³U Campaign. These data show (lbs of 100 percent ANN):

Preflushes	8,160 + 30,500	=	38,660 lbs ANN
Processing	14,860 + 3,500	=	18,360 lbs ANN
			57,020 lbs ANN
		Take	57,000 lbs ANN

$$\text{The wt\% Al in 100 percent ANN } [\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}] = \\ (1)(27)(100)/375.14 = 7.20 \text{ percent}$$

$$\text{ANN Al in 1966 campaign} = (5.7 \text{ E}+4 \text{ lbs ANN})(0.4536 \text{ kg}/\text{lb})(0.072 \text{ Al}/\text{ANN}) \\ = 1.86 \text{ E}+3 \text{ kg Al} = 1.86 \text{ MT Al}$$

This usage was for processing 197 MT ThO₂ (Appendix B).

The MT ThO₂ processed in the 1970 campaign was 427 MT (Appendix B). Assume that the ANN used for flushing was the same as for the 1966 campaign, and ratio the ANN used for processing based on the relative amounts of ThO₂ processed.

ANN Al in 1970 campaign

Flushing	=	38,660 lbs ANN
Processing = (18,360 lb/197 MT)(427 MT)	=	39,796
		78,456 lbs ANN
	Take	78,500 lbs ANN

$$\text{Al} = (78,500)(0.4536)(0.072) = 2.56 \text{ E}+3 \text{ kg} = 2.56 \text{ MT Al}$$

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Total Aluminum Added in ThO₂ Processing

Cladding (all campaigns)	=	38.8 MT
1966 Processing Campaign	=	1.9
1970 Processing Campaign	=	<u>2.6</u>
		43.3 MT
approximately		43 MT Al

E3.0 ESTIMATE OF ALUMINUM IN TANKED Z PLANT WASTE

E3.1 FY 1973 THROUGH FY 1976 (TO 241-TX-118)

1. Volume estimates:
 - a. From Anderson (1990) and Brevick (1996a) $6.90 \text{ E}+6 \text{ L}$ (1,823 kgal). These are the same except Brevick (1996a) has 230.9 kL (61 kgal) in the second quarter of 1976 that is not shown in Anderson (1990) (included in 1.823 Mgal).
 - b. From G. G. Bergquist (1996 electronic mail) $5.22 \text{ E}+6 \text{ L}$ (1,379 kgal)
 - c. From HDW Rev. 3, $6.27 \text{ E}+6 \text{ L}$ (1,656 kgal)
Take average of 3 = $6.13 \text{ E}+6 \text{ L}$
2. Al concentration estimates
 - a. From G. G. Bergquist (1996 electronic mail)
 $0.3M$
 $0.66M$ (range 0.45 to $0.92M$)
 $0.3M$
 - b. From flowsheets, one (Jones 1993) shows CAW as $0.55M \text{ Al}$. Another (RHO 1987) gives it as $0.72M$. In both cases, the CAW would be diluted from 1 to approximately 1.75 (at least) so the tanked waste Al M would range from approximately $0.25M$ to ~ 0.35
 - c. HDW Rev. 3, uses $0.5M$, no reference.
Best estimate - use $0.3M \text{ Al}^{+3}$

E3.2 FY 1973 THROUGH 1976 AL

$$\begin{aligned} (6.13 \text{ E}+6 \text{ L})(0.3M) &= 1.84 \text{ E}+6 \text{ g mol Al} \\ &= 4.97 \text{ E}+7 \text{ g Al} \\ &= 49.7 \text{ MT Al} \end{aligned}$$

E3.3 FY 1977 THROUGH 1981

(Z Plant was down in FY 1977)

Note: Post 1976, all wastes were routed to tank 241-SY-102

1. Volume/concentration estimates
Only data are volume projections given in Bergquist (1996 electronic mail) for FY 1978 through FY 1984. Per discussion with T. J. Venetz (5/29/96), it is thought that the only one of these years that Al was used was in 1979 with $3.94 \text{ E}+5 \text{ L}$ (104 kgal) - assumed $0.3M \text{ Al}$

$$\begin{aligned}
 (3.94 \text{ E}+5 \text{ L})(0.3M) &= 1.18 \text{ E}+5 \text{ g mol Al} \\
 &= 3.19 \text{ E}+6 \text{ g Al} \\
 &= 3.2 \text{ MT Al}
 \end{aligned}$$

E3.4 FY 1982 AND FY 1983

There are some Z Plant waste volumes reported, however, per 5/29/96 discussion with T. J. Venetz, PRF did not run in this time period (or at least did not use ANN). Therefore, no Al usage is calculated.

E3.5 FY 1985

PRF ran and the Remote Mechanical C (RMC) line tried using an ANN scrubber. Therefore, an Al concentration of 0.5M was used.

1. Volume estimates
 - a. G. G. Bergquist (1996 electronic mail) showed 7.57 E+5 L (200,000 gal)
 - b. The waste volume projections file for Tank 241-SY-102 shows 7.76 E+5 L (205,000 gal) as the sum of the Z Lab, Z PRFL, and ZRMCL wastes. Brevick (1996b) confirms this, so use this.

FY 1985

$$\begin{aligned}
 (7.76 \text{ E}+5 \text{ L})(0.5M) &= 3.8 \text{ E}+5 \text{ g mol Al} \\
 &= 1.05 \text{ E}+7 \text{ g Al} \\
 &= 10.5 \text{ MT Al}
 \end{aligned}$$

E3.6 FY 1984 THROUGH 1989 (EXCLUDING FY 1985)

An Al concentration of 0.3M was used. G. G. Bergquist (1996 electronic mail) gives a total volume for FY 1986 through FY 1989 of 680 kgal (no actual volume for FY 1984 was shown). The Waste Volume Projection (WVP) file and Brevick (1996b) show (sum of Z Lab, ZPRFL, Z High and ZRCML) 2,195 kL (580 kgal) for these years plus 734.3 kL (194 kgal) for FY 1984 for a total of 2.93 E+6 L (774 kgal). Use the following:

FY 1984 and FY 1986 through FY 1989

$$\begin{aligned}
 (2.93 \text{ E}+6 \text{ L})(0.3M) &= 8.79 \text{ E}+5 \text{ g mol Al} \\
 &= 2.37 \text{ E}+7 \text{ g Al} \\
 &= 23.7 \text{ MT Al}
 \end{aligned}$$

E3.7 FY 1990 AND FY 1991

PRF did not run, the WVP file and Brevick (1996b) show no entries other than Z Lab.

E3.8 FY 1992

A training run in PRF was performed. The WVP file and Brevick (1996b) show a ZPRFL volume of $1.06 \text{ E}+5 \text{ L}$ (28 kgal)

Using 0.3M ANN

FY 1992

$$\begin{aligned} (1.06 \text{ E}+5 \text{ L})(0.3\text{M}) &= 3.8 \text{ E}+4 \text{ g mol Al} \\ &= 8.6 \text{ E}+5 \text{ g Al} \\ &= 0.9 \text{ MT Al} \end{aligned}$$

E3.9 TOTAL AL FROM Z PLANT TO UGS TANKS

FY 1973 through FY 1976	49.7 MT
FY 1977 through FY 1978	0
FY 1979	3.2
FY 1980 through FY 1983	0
FY 1985	10.5
FY 1984 and FY 1986 through FY 1989	23.7
FY 1992	<u>0.9</u>
Total	88 MT

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APPENDIX F

SUPPORTING DATA FOR LEAD (SECTION 5.12)

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APPENDIX F

SUPPORTING DATA FOR LEAD (SECTION 5.12)

Amount of lead introduced into tanks as a result of operation of PbSO_4 carrier precipitation process.

Time frame: 1961 through 1964

Phase I. Recover/purify 1 MCi ^{90}Sr (1961 through 1962) (Moore 1964).

1. PbSO_4 process:

- Use 90 gal 0.5M $\text{Pb}(\text{NO}_3)_2$ per 1,000 gal PUREX process 1WW solution
90 gal 0.5M $\text{Pb}(\text{NO}_3)_2$ equivalent to 35 kg Pb/ 1,000 gal batch
- ^{90}Sr concentration during 1961 through 1962 in 1WW solution
 - 1,922.2 Ci ^{90}Sr /ton of fuel (Isochem 1964, Table II-6)
 - 1WW volume = 80 gal/ton (Van Tuyl 1958)
 - ^{90}Sr concentration in 1WW = $\frac{1,922 \text{ Ci/ton}}{80 \text{ gal/ton}} = 24 \text{ Ci/gal}$
 - Number of 10^3 gal batches of 1WW to recover 10^6 Ci at process efficiency of 55 percent.
 $10^6 \text{ Ci}/24,500 \text{ Ci/batch}/0.55 = 75.76 \times 10^3 \text{ gal batches}$
 - MT of Pb used to treat 75.76 batches
 $75.76 \text{ batch} \times 35 \text{ kg Pb/batch} \times 10^{-3} \text{ MT/kg} = 2.65 \text{ MT of Pb}$

Phase II. Recover/purify 8 MCi ^{90}Sr (1963 through 1964) (Moore 1964).

- Use 90 gal 0.5M $\text{Pb}(\text{NO}_3)_2$ per 1,500 gal PUREX process acid waste (Beard and Judson 1963). Equivalent to 35 kg Pb/1,500 gal batch
- ^{90}Sr concentration of PUREX process acid waste = 24 Ci/gal
- Number of 1,500 gal batches of PUREX process acid waste to recover 8 MCi ^{90}Sr at 90 percent process efficiency.
 $8 \times 10^6 \text{ Ci}/36,000 \text{ Ci/batch}/0.9 = 247 \text{ batches}$
- MT of Pb used to treat 370 batches PUREX process acid waste
 $247 \text{ batch} + 35 \text{ kg Pb/batch} + 10^{-3} \text{ MT/kg} = 8.642 = \text{approximately } 8.64 \text{ MT of Pb}$

Therefore, MT of Pb used in PUREX Plant during 1961 through 1964 = 2.65 + 8.64
= 11.3 MT

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APPENDIX G

SAMPLE FUEL ACTIVITY RECORDS

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APPENDIX G

SAMPLE FUEL ACTIVITY RECORDS

Table G-1. Generic Factors Splitting Radionuclides Between Cladding Waste and Separations Waste Streams (Input to DKPRO Code).

Plant	Fuel Type	Fraction of Core Radionuclides Split to Cladding Waste (CW).		
		U CW	Pu and others CW	Th CW
T or B	AL-0.71U	0.00081	0.00102	0.00102
REDOX	AL-0.71U AL-0.94U	0.00081	0.00102	0.00102
REDOX	ZR-0.94U ZR-1.1U	0.0072	0.0052	0.0052
PUREX	AL-0.71U AL-0.94U	0.00081	0.00102	0.00102
PUREX	AL-0.0TH	0.0206	0.0206	0.0084
PUREX	ZR-0.94U ZR-1.1U ZR-2.1U	0.014	0.023	0.023

CW = Cladding waste

PUREX = Plutonium-Uranium Extraction

REDOX = Reduction-Oxidation

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Table G-2 (Schneider 1951) gives fractional split factors for radionuclides in extraction feed to various bismuth phosphate process waste streams.

Table G-2. Extraction Split Factors for T and B Plants. (2 Sheets)
Created: June 7, 1996.

Fracton of extraction feed routed to stream:				
Nuclide	MW	1C	2C	224
³ H	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁴ C	6.600E-01*	1.970E-01	1.000E-03	1.000E-05
⁵⁹ Ni	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁶⁰ Co	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁶³ Ni	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁷⁹ Se	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁹⁰ Sr	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁹⁰ Y	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁹³ Zr	8.020E-01	1.970E-01	1.000E-03	1.000E-05
^{93m} Nb	8.020E-01	1.970E-01	1.000E-03	1.000E-05
⁹⁹ Tc	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁰⁶ Ru	8.020E-01	1.970E-01	1.000E-03	1.000E-05
^{113m} Cd	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹²⁵ Sb	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹²⁶ Sn	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹²⁹ I	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹³⁴ Cs	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹³⁷ Cs	8.020E-01	1.970E-01	1.000E-03	1.000E-05
^{137m} Ba	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁵¹ Sm	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁵² Eu	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁵⁴ Eu	8.020E-01	1.970E-01	1.000E-03	1.000E-05
¹⁵⁵ Eu	8.020E-01	1.970E-01	1.000E-03	1.000E-05

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Table G-2. Extraction Split Factors for T and B Plants. (2 Sheets)
Created: June 7, 1996.

Fracton of extraction feed routed to stream:				
Nuclide	MW	1C	2C	224
²²⁶ Ra	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²²⁷ Ac	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²²⁸ Ra	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²²⁹ Th	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²³¹ Pa	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²³² Th	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²³² U	9.968E-01	3.200E-03	0.000	0.000
²³³ U	9.968E-01	3.200E-03	0.000	0.000
²³⁴ U	9.968E-01	3.200E-03	0.000	0.000
²³⁵ U	9.968E-01	3.200E-03	0.000	0.000
²³⁶ U	9.968E-01	3.200E-03	0.000	0.000
²³⁷ Np	9.968E-01	3.200E-03	0.000	0.000
²³⁸ Pu	3.940E-03	1.390E-02	3.900E-03	3.900E-03
²³⁸ U	9.968E-01	3.200E-03	0.000	0.000
²³⁹ Pu	3.940E-03	1.390E-02	3.900E-03	3.900E-03
²⁴⁰ Pu	3.940E-03	1.390E-02	3.900E-03	3.900E-03
²⁴¹ Am	9.968E-01	3.200E-03	0.000	0.000
²⁴¹ Pu	3.940E-03	1.390E-02	3.900E-03	3.900E-03
²⁴² Pu	3.940E-03	1.390E-02	3.900E-03	3.900E-03
²⁴² Cm	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²⁴³ Am	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²⁴³ Cm	8.020E-01	1.970E-01	1.000E-03	1.000E-05
²⁴⁴ Cm	8.020E-01	1.970E-01	1.000E-03	1.000E-05

*0.66 = Assuming same factor as for Plutonium-Uranium Extraction (PUREX) (Watrous 1984). To be revised.

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Table G-3 gives fractional split factors for radionuclides in REDOX SX feed to various waste streams.

Table G-3. Extraction Split Factors for REDOX Plant. (2 Sheets)
Created: June 7, 1996

Fracton of extraction feed routed to stream		
Nuclide	High-Level Waste (D8)	Reference
³ H	7.800E-01	0.78 Calculated from flowsheet #6 flows, HW-66203 (Crawley 1960)
¹⁴ C	6.600E-01	PUREX Factor, to be Revised (Watrous 1984).
⁵⁹ Ni	1.000E+00	
⁶⁰ Co	1.000E+00	
⁶³ Ni	1.000E+00	
⁷⁹ Se	1.000E+00	
⁹⁰ Sr	1.000E+00	
⁹⁰ Y	1.000E+00	
⁹³ Zr	1.000E+00	
^{93m} Nb	1.000E+00	
⁹⁹ Tc	6.300E-01	Colby and Petersen (1995) Figure 1.2
¹⁰⁶ Ru	1.000E+00	
^{113m} Cd	1.000E+00	
¹²⁵ Sb	1.000E+00	
¹²⁶ Sn	1.000E+00	
¹²⁹ I	1.000E+00	
¹³⁴ Cs	1.000E+00	
¹³⁷ Cs	1.000E+00	
^{137m} Ba	1.000E+00	
¹⁵¹ Sm	1.000E+00	
¹⁵² Eu	1.000E+00	
¹⁵⁴ Eu	1.000E+00	
¹⁵⁵ Eu	1.000E+00	
²²⁶ Ra	1.000E+00	

Table G-3. Extraction Split Factors for REDOX Plant. (2 Sheets)

Created: June 7, 1996

Fracton of extraction feed routed to stream		
Nuclide	High-Level Waste (D8)	Reference
²²⁷ Ac	1.000E+00	
²²⁸ Ra	1.000E+00	
²²⁹ Th	1.000E+00	
²³¹ Pa	1.000E+00	
²³² Th	1.000E+00	
²³² U	1.990E-03	Jenkins and Foster (1978)
²³³ U	1.990E-03	Jenkins and Foster (1978)
²³⁴ U	1.990E-03	Jenkins and Foster (1978)
²³⁵ U	1.990E-03	Jenkins and Foster (1978)
²³⁶ U	1.990E-03	Jenkins and Foster (1978)
²³⁷ Np	1.000E+00	
²³⁸ Pu	4.500E-03	Jenkins and Foster (1978)
²³⁸ U	1.990E-03	Jenkins and Foster (1978)
²³⁹ Pu	4.500E-03	Jenkins and Foster (1978)
²⁴⁰ Pu	4.500E-03	Jenkins and Foster (1978)
²⁴¹ Am	1.000E+00	
²⁴¹ Pu	4.500E-03	Jenkins and Foster (1978)
²⁴² Pu	4.500E-03	Jenkins and Foster (1978)
²⁴² Cm	1.000E+00	
²⁴³ Am	1.000E+00	
²⁴³ Cm	1.000E+00	
²⁴⁴ Cm	1.000E+00	

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Table G-4 gives fractional split factors for radionuclides in PUREX SX feed to various waste streams.

Table G-4. Extraction Split Factors for PUREX Plant. (2 Sheets)
Created: June 7, 1996

Fracton of extraction feed routed to stream				
Nuclide	High-level waste (F16)	OWW	Low-Level Waste (F18)	Reference
³ H	1.400E-02	6.500E-05	3.000E-04	0.014 calculated from ARH-214 (Crawley and Harmon 1960) flows.
¹⁴ C	6.600E-01	6.500E-05	3.000E-04	0.66, (Watrous 1984). To be revised.
⁵⁹ Ni	9.996E-01	6.500E-05	3.000E-04	OWW and LLW from Allen et al. (1985), SK-2-23944
⁶⁰ Co	9.996E-01	6.500E-05	3.000E-04	
⁶³ Ni	9.996E-01	6.500E-05	3.000E-04	
⁷⁹ Se	9.996E-01	6.500E-05	3.000E-04	
⁹⁰ Sr	9.996E-01	6.500E-05	3.000E-04	
⁹⁰ Y	9.996E-01	6.500E-05	3.000E-04	
⁹³ Zr	9.996E-01	6.500E-05	3.000E-04	
^{93m} Nb	9.996E-01	6.500E-05	3.000E-04	
⁹⁹ Tc	7.596E-01	6.500E-05	3.000E-04	Colby and Petersen (1995), Figure 1.1
¹⁰⁶ Ru	9.996E-01	6.500E-05	3.000E-04	
^{113m} Cd	9.996E-01	6.500E-05	3.000E-04	
¹²⁵ Sb	9.996E-01	6.500E-05	3.000E-04	
¹²⁶ Sn	9.996E-01	6.500E-05	3.000E-04	
¹²⁹ I	3.600E-02	5.200E-02	7.000E-02	
¹³⁴ Cs	9.996E-01	6.500E-05	3.000E-04	
¹³⁷ Cs	9.996E-01	6.500E-05	3.000E-04	
^{137m} Ba	9.996E-01	6.500E-05	3.000E-04	
¹⁵¹ Sm	9.996E-01	6.500E-05	3.000E-04	
¹⁵² Eu	9.996E-01	6.500E-05	3.000E-04	
¹⁵⁴ Eu	9.996E-01	6.500E-05	3.000E-04	
¹⁵⁵ Eu	9.996E-01	6.500E-05	3.000E-04	

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Table G-4. Extraction Split Factors for PUREX Plant. (2 Sheets)
Created: June 7, 1996

Fracton of extraction feed routed to stream				
Nuclide	High-level waste (F16)	OWW	Low-Level Waste (F18)	Reference
²²⁶ Ra	9.996E-01	6.500E-05	3.000E-04	
²²⁷ Ac	9.996E-01	6.500E-05	3.000E-04	
²²⁸ Ra	9.996E-01	6.500E-05	3.000E-04	
²²⁹ Th	9.996E-01	6.500E-05	3.000E-04	
²³¹ Pa	9.996E-01	6.500E-05	3.000E-04	
²³² Th	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³² U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³³ U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁴ U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁵ U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁶ U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁷ Np	9.989E-01	6.500E-05	1.000E-03	
²³⁸ Pu	2.400E-03	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁸ U	9.000E-04	6.500E-05	1.000E-03	Allen et al. (1985)
²³⁹ Pu	2.400E-03	6.500E-05	1.000E-03	Allen et al. (1985)
²⁴⁰ Pu	2.400E-03	6.500E-05	1.000E-03	Allen et al. (1985)
²⁴¹ Am	9.996E-01	6.500E-05	3.000E-04	
²⁴¹ Pu	2.400E-03	6.500E-05	1.000E-03	Allen et al. (1985)
²⁴² Pu	2.400E-03	6.500E-05	1.000E-03	Allen et al. (1985)
²⁴² Cm	9.996E-01	6.500E-05	3.000E-04	
²⁴³ Am	9.996E-01	6.500E-05	3.000E-04	
²⁴³ Cm	9.996E-01	6.500E-05	3.000E-04	
²⁴⁴ Cm	9.996E-01	6.500E-05	3.000E-04	

LLW = Low-Level Waste
OWW = Organic Wash Waste.

Table G-5 gives p/M levels for activation product target impurities in SPR and NPR fuels. The intent is that target p/M values will be used to correct ORIGEN2 output tables by a simple ratio multiplier. The SEPARATIONS MODEL will also use these values to reconstruct the split of activation products between fuel core and cladding.

Table G-5. Activation Product Target Values. Created: June 7, 1996

Activation Product	Target Element	Time Period	Single Pass Reactors		N-Reactor		
			Core (p/M)	Al Clad (p/M)	Core (p/M)	Zr Clad (p/M)	Braze (p/M)
¹⁴ C ^{b,d,e}	N	1944-1949	90.00	0 - 250 ^a	90.00	80.00	200.00
	N	1950-1954	65.00	0 - 250	65.00	80.00	200.00
	N	1955-1959	40.00	0 - 250	40.00	80.00	200.00
	N	1960-1969	17.50	0 - 250	17.50	80.00	200.00
	N	1970-1978	8.00	0 - 250	8.00	80.00	200.00
	N	1979-1989	9.30	0 - 250	9.30	80.00	200.00
^{59/63} Ni ^e	Ni	1944-1958	100.00	100.00	100.00	300-800	280-800
^{59/63} Ni ^e	Ni	1959-1972	100.00	9000-13000	100.00	300-800	280-800
⁶⁰ Co ^f	Co		1.00	10.00	1.00	10.44	
Cladding mass factors:							
kg Clad Al/MTU				37.62			
kg Clad Zr/MTU ZR-0.94U (Mk IV)				70.35			
kg Clad Zr/MTU ZR-1.1U (Mk 1a)				85.65			
kg Clad Zr/MTU ZR-2.1U (Mk II)				86.99			

^aNo data exist on N p/M in Al cladding. 250 p/M is a guess that the maximum N value might be as high as 50 percent of the specification (500 p/M) for all other impurities.

^bMcNeece (1975), Table 4.1-3

^cWatrous (1984)

^dFort (1987)

^eSee Section 7.1.1 for discussion of nickle impurity levels

^fT10.44 p/M is a weighted average of 10 p/M in Zr plus 300 p/M in the Fe shoes.

Table G-6. Sample of Separations Production File.
(Input to DKPRO code)

KEY	TYPE	MTU	240	MUM/MTU	PLANT	MO	YR.	SEP	KEY	Mid-Mo*	COOLING DAYS	Ref.	WASTE FRACTION:								
													Th	U	Hp	Pu					
3702.0	AL-0.71U	1.647E+02		2.240E+02	Redox	2	55	3794.01	92	SPLANT.DAT	19348	9/30/93	1:199	1	0.0033	1	0.0079	1	0.0000	1	0.0000
3748.0	AL-0.71U	2.268E+02		2.289E+02	Redox	3	55	3824.01	82	SPLANT.DAT	19348	9/30/93	1:199	1	0.0046	1	0.0082	1	0.0000	1	0.0000
3829.0	AL-0.71U	1.701E+02		7.104E+02	Redox	4	55	3855.01	147	SPLANT.DAT	19348	9/30/93	1:199	1	0.009	1	0.0131	1	0.0000	1	0.0000
3815.0	AL-0.71U	2.324E+02		8.934E+02	Redox	5	55	3885.01	456	SPLANT.DAT	19348	9/30/93	1:199	1	0.0043	1	0.0087	1	0.0000	1	0.0000
3844.0	AL-0.71U	1.332E+02		9.499E+02	Redox	6	55	3915.01	100	SPLANT.DAT	19348	9/30/93	1:199	1	0.0059	1	0.0096	1	0.0000	1	0.0000
3867.0	AL-0.71U	1.444E+02		3.988E+02	Redox	7	55	3946.01	102	SPLANT.DAT	19348	9/30/93	1:199	1	0.0079	1	0.0121	1	0.0000	1	0.0000
3907.0	AL-0.71U	2.076E+02		2.876E+02	Redox	8	55	3976.01	109	SPLANT.DAT	19348	9/30/93	1:199	1	0.0079	1	0.0121	1	0.0000	1	0.0000
3937.0	AL-0.71U	2.813E+02		2.858E+02	Redox	9	55	4007.01	100	SPLANT.DAT	19348	9/30/93	1:199	1	0.0043	1	0.0076	1	0.0000	1	0.0000
3997.0	AL-0.71U	2.169E+02		9.003E+02	Redox	10	55	4037.01	157	SPLANT.DAT	19348	9/30/93	1:199	1	0.0043	1	0.0076	1	0.0000	1	0.0000
4012.0	AL-0.71U	1.389E+02		1.000E+03	Redox	11	55	4068.01	101	SPLANT.DAT	19348	9/30/93	1:199	1	0.0063	1	0.0092	1	0.0000	1	0.0000
4045.0	AL-0.71U	1.047E+02		2.634E+02	Redox	12	55	4098.01	117	SPLANT.DAT	19348	9/30/93	1:199	1	0.0087	1	0.0182	1	0.0000	1	0.0000
4061.0	AL-0.71U	2.240E+02		2.736E+02	Redox	1	56	4129.02	114	SPLANT.DAT	19348	9/30/93	1:199	1	0.0081	1	0.0112	1	0.0000	1	0.0000
4075.0	AL-0.71U	1.664E+02		3.564E+02	Redox	2	56	4159.02	128	SPLANT.DAT	19348	9/30/93	1:199	1	0.008	1	0.008	1	0.0000	1	0.0000
4161.0	AL-0.71U	1.820E+02		9.039E+02	Redox	3	56	4189.02	145	SPLANT.DAT	19348	9/30/93	1:199	1	0.0048	1	0.0048	1	0.0000	1	0.0000
4171.0	AL-0.71U	1.943E+02		8.437E+02	Redox	4	56	4220.02	109	SPLANT.DAT	19348	9/30/93	1:199	1	0.0018	1	0.0021	1	0.0000	1	0.0000
4187.0	AL-0.71U	1.493E+02		7.800E+02	Redox	5	56	4250.02	124	SPLANT.DAT	19348	9/30/93	1:199	1	0.0025	1	0.0025	1	0.0000	1	0.0000
4208.0	AL-0.71U	5.427E+01		8.266E+02	Redox	6	56	4281.02	110	SPLANT.DAT	19348	9/30/93	1:199	1	0.0044	1	0.0049	1	0.0000	1	0.0000
4224.0	AL-0.71U	9.519E+01		2.776E+02	Redox	7	56	4311.02	134	SPLANT.DAT	19348	9/30/93	1:199	1	0.0049	1	0.0049	1	0.0000	1	0.0000
4245.0	AL-0.71U	2.189E+02		2.776E+02	Redox	8	56	4342.02	148	SPLANT.DAT	19348	9/30/93	1:199	1	0.007	1	0.007	1	0.0000	1	0.0000
4296.0	AL-0.71U	2.525E+02		2.740E+02	Redox	9	56	4372.02	157	SPLANT.DAT	19348	9/30/93	1:199	1	0.0067	1	0.0067	1	0.0000	1	0.0000
4349.0	AL-0.71U	2.526E+02		2.732E+02	Redox	10	56	4402.02	137	SPLANT.DAT	19348	9/30/93	1:199	1	0.0023	1	0.0023	1	0.0000	1	0.0000
4371.0	AL-0.71U	2.241E+02		2.732E+02	Redox	11	56	4433.02	123	SPLANT.DAT	19348	9/30/93	1:199	1	0.0017	1	0.0017	1	0.0000	1	0.0000
4404.0	AL-0.71U	1.022E+01		7.627E+02	Redox	12	56	4463.02	120	SPLANT.DAT	19348	9/30/93	1:199	1	0.0023	1	0.0023	1	0.0000	1	0.0000
4427.0	AL-0.71U	7.327E+01		7.040E+02	Redox	1	57	4494.02	128	SPLANT.DAT	19348	9/30/93	1:199	1	0.0017	1	0.0017	1	0.0000	1	0.0000
4466.0	AL-0.71U	8.153E+01		6.249E+02	Redox	2	57	4524.02	119	SPLANT.DAT	19348	9/30/93	1:199	1	0.004	1	0.004	1	0.0000	1	0.0000
4495.0	AL-0.71U	1.407E+02		6.207E+02	Redox	3	57	4555.02	121	SPLANT.DAT	19348	9/30/93	1:199	1	0.001	1	0.001	1	0.0000	1	0.0000
4522.0	AL-0.71U	1.342E+02		6.347E+02	Redox	4	57	4585.02	124	SPLANT.DAT	19348	9/30/93	1:199	1	0.0018	1	0.0018	1	0.0000	1	0.0000
4558.0	AL-0.71U	9.085E+01		6.102E+02	Redox	5	57	4616.02	118	SPLANT.DAT	19348	9/30/93	1:199	1	0.0023	1	0.0023	1	0.0000	1	0.0000
4594.0	AL-0.71U	1.558E+02		5.617E+02	Redox	6	57	4646.02	113	SPLANT.DAT	19348	9/30/93	1:199	1	0.0009	1	0.0009	1	0.0000	1	0.0000
4623.0	AL-0.71U	1.389E+02		6.678E+02	Redox	7	57	4676.02	110	SPLANT.DAT	19348	9/30/93	1:199	1	0.0024	1	0.0024	1	0.0000	1	0.0000
4633.0	AL-0.71U	1.991E+02		6.124E+02	Redox	8	57	4707.02	104	SPLANT.DAT	19348	9/30/93	1:199	1	0.0016	1	0.0016	1	0.0000	1	0.0000
4658.0	AL-0.71U	1.648E+02		6.903E+02	Redox	9	57	4737.02	110	SPLANT.DAT	19348	9/30/93	1:199	1	0.0024	1	0.0024	1	0.0000	1	0.0000
4695.0	AL-0.71U	1.679E+02		6.413E+02	Redox	10	57	4768.02	103	SPLANT.DAT	19348	9/30/93	1:199	1	0.0017	1	0.0017	1	0.0000	1	0.0000
4729.0	AL-0.71U	2.293E+02		4.364E+02	Redox	11	57	4798.02	100	SPLANT.DAT	19348	9/30/93	1:199	1	0.0016	1	0.0016	1	0.0000	1	0.0000
4755.0	AL-0.71U	7.025E+01		5.734E+02	Redox	12	57	4829.02	104	SPLANT.DAT	19348	9/30/93	1:199	1	0.0025	1	0.0025	1	0.0000	1	0.0000
4799.0	AL-0.71U	1.337E+02		5.724E+02	Redox	1	58	4859.02	90	SPLANT.DAT	19348	9/30/93	1:199	1	0.0022	1	0.0022	1	0.0000	1	0.0000
4828.0	AL-0.71U	9.299E+01		5.897E+02	Redox	2	58	4889.02	92	SPLANT.DAT	19348	9/30/93	1:199	1	0.0027	1	0.0027	1	0.0000	1	0.0000
4857.0	AL-0.71U	8.031E+01		5.897E+02	Redox	3	58	4920.02	93	SPLANT.DAT	19348	9/30/93	1:199	1	0.0031	1	0.0031	1	0.0000	1	0.0000
4886.0	AL-0.71U	1.708E+02		6.167E+02	Redox	4	58	4950.02	95	SPLANT.DAT	19348	9/30/93	1:199	1	0.0024	1	0.0024	1	0.0000	1	0.0000
4905.0	AL-0.71U	1.840E+02		5.454E+02	Redox	5	58	4981.02	106	SPLANT.DAT	19348	9/30/93	1:199	1	0.0015	1	0.0015	1	0.0000	1	0.0000
4934.0	AL-0.71U	1.840E+02		5.990E+02	Redox	6	58	5011.02	106	SPLANT.DAT	19348	9/30/93	1:199	1	0.0016	1	0.0016	1	0.0000	1	0.0000
4966.0	AL-0.71U	1.889E+02		6.042E+02	Redox	7	58	5042.02	106	SPLANT.DAT	19348	9/30/93	1:199	1	0.0014	1	0.0014	1	0.0000	1	0.0000
4994.0	AL-0.71U	1.293E+02		6.012E+02	Redox	8	58	5072.02	109	SPLANT.DAT	19348	9/30/93	1:199	1	0.0025	1	0.0025	1	0.0000	1	0.0000
						9	58	5103.02						1	0.0025	1	0.0025	1	0.0000	1	0.0000

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APPENDIX H

CORROSION PRODUCTS
(IRON, NICKEL, AND CHROMIUM)

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APPENDIX H
CORROSION PRODUCTS (IRON, NICKEL, AND CHROMIUM)

H1.0 INTRODUCTION

In Hanford Site waste, iron (Fe), nickel (Ni), and chromium (Cr) are generated from two sources:

1. Process chemicals, such as ferrous sulfamate, nickel sulfate, or sodium dichromate added in fuel processing and tank operations.
2. Corrosion of stainless steel piping and equipment during fuel processing operations and waste evaporator campaigns, and (for Fe) in-tank corrosion of mild steel waste storage tanks.

Flowsheets for the bismuth phosphate (BiPO_4), Uranium Recovery (UR), reduction and oxidation (REDOX), and plutonium-uranium extraction (PUREX) processes provide some information concerning the amounts of Fe, Ni, and Cr introduced into chemical processes. Analytical data obtained from core samples, especially from tanks containing only one or two characteristic waste types (Agnew et al. 1995a, Agnew et. al. 1996), provide information concerning the amounts of Fe, Ni, and Cr in the tanks. Additional information is provided by essential material usage records. The amount of corrosion-generated Fe, Ni, and Cr introduced to wastes in the PUREX and REDOX processes can be estimated from the nickel content of those wastes.

H1.1 CORROSION PRODUCT INVENTORY EVALUATION

The global amount of corrosion-generated Fe, Ni, and Cr is summarized in Table H-1.

Table H-1. Global Inventory of Corrosion-Generated Iron, Nickel, and Chromium.*

Process	Iron, MT	Nickel, MT	Chromium, MT
BiPO_4	32.4	4.6	8.7
Uranium Recovery	44.8	6.3	12.0
REDOX	8.5	1.2	2.3
PUREX (1956 to 1972)	141.7	20.0	37.9
PUREX (1983 to 1989)	36.0	5.1	9.6
Total	263.4	37.2	70.5

PUREX = Plutonium-uranium extraction

REDOX = Reduction and oxidation

*Based on the composition of 304 stainless steel (71 percent Fe, 10 percent Ni, and 19 percent Cr).

H1.1.1 Bismuth Phosphate Process Wastes

The bismuth phosphate process operated at T Plant from December 1944 through 1956 and at B Plant from April 1945 through 1952. These processes collectively produced 393.6 ML (104,000 kgal) of metal waste from the bismuth phosphate process (MW), first cycle decontamination waste from the bismuth phosphate process (1C), second cycle decontamination waste from the bismuth phosphate process (2C), final concentration/decontamination cycle waste from the bismuth phosphate process (224), and cladding waste (CW) (Agnew et al. 1996). The Hanford Site production records indicate that 7,800 metric tons of uranium (MTU) were processed in the bismuth phosphate plants from 1944 to 1956 (Heeb 1994).

The Bismuth Phosphate Process Flowsheet (Schneider 1951) is discussed in Appendix C. This analysis indicates that 146.5 MT of Fe from ferrous ammonium sulfate use reported to the 1C waste and 199.1 MT to the 2C waste, adding a total of 345.5 MT of Fe from both sources. Furthermore, 17.2 MT of Cr from sodium dichromate use were added to the 1C waste, 6.6 MT to the 2C waste and 19.4 MT to the 224 waste, producing an estimated total of 43.2 MT of Cr from chemical sources (derived from waste volume and waste summary concentrations given in Appendix C, Table C-4).

Process flowsheets are of limited value with respect to predicting the amount of corrosion related Fe in the waste. Since corrosion estimates are not generally available from other sources, estimates of total Fe should be based as much as possible on the actual composition of the sludge in the tanks. For these estimates, tank core samples and waste volumes from the Tank Layer Model (TLM) section of the HDW model were used to estimate the total amount of Fe in Hanford Site's BiPO₄ waste.

The HDW model utilizes tank transaction records, chemical and physical properties of wastes, and derived compositions for 55 of the most important wastes produced at the Hanford Site (chemicals used and wastes produced). In this model, sludge accumulation measurements in the tanks were used to determine the volume of solids precipitated from each waste. While component solubilities and the likely distribution of soluble components between sludge and supernate were estimated from limiting solubilities of key components in the supernate. The solids vol% is also very important since this value determines the sludge void fraction and the amount of interstitial liquid in each sludge. Solids vol% values were derived from HDW analysis based on the known inventory of sludge in the tanks. The HDW model uses observed waste volumes, both liquid and settled solids, to establish an actual waste generation rate in L/MT waste of uranium processed for each waste stream.

Most transfers of waste material from the processing plants to the tanks and subsequent transfers between tanks or back to the processing plants or waste evaporators are represented in transaction records that were compiled into the Waste Status and Transaction Record Summaries (WSTRS) (Agnew et al. 1995a). These transaction records were used to estimate the volume of the solids layers in each tank (Agnew et al. 1995b). It is implicitly assumed

in this model that each layer has a unique composition that corresponds to one of the 55 characteristic or defined wastes.

Bismuth phosphate process wastes can be generally defined as metal waste, 1C, 2C, and 224 waste. Table H-2 summarizes the basis for computing the Fe inventory of BiPO₄ waste in the TLM model (Agnew et al. 1995b, Appendix B).

Table H-2. Basis for Estimating the Iron Inventory of BiPO₄ Waste in the Tank Layer Model.

TLM Basis	MW1	MW2	1C1	1C2	2C1	2C2	224
Date	1944-49	1950-56	1944-49	1950-56	1944-49	1950-56	1944-56
MTU	3,341	4,457	3,341	4,457	3,341	4,457	7,798
kgal total waste	15,325	20,551	11,767	16,531	8,962	22,727	8,300
kgal Solids	736	0	1,732	1,407	762	821	322
Fe, ppm	3,794	3,794	13,981	8,165.3	25,018	38,973	14,647
Density	1.7466	1.7466	1.2908	1.2223	1.2512	1.125	1.3763
Fe, MT	18.46	-	118.3	53.15	90.36	136.33	24.57

MW1 = Metal waste from the bismuth phosphate process, 1944 to 1951

MW2 = Metal waste from the bismuth phosphate process, 1952 to 1956

TLM = Tank layer model

1C1 = First decontamination cycle of the bismuth phosphate process, 1944 to 1951

1C2 = First decontamination cycle of the bismuth phosphate process, 1952 to 1956

2C1 = Second decontamination cycle of the bismuth phosphate process, 1944 to 1951

2C2 = Second decontamination cycle of the bismuth phosphate process, 1952 to 1956

224 = Final decontamination/concentration cycle of the bismuth phosphate process.

There are about 17 tanks with simple and reasonably well defined HDW sludge layers from which representative core samples have been obtained. Eleven of these tanks were used to estimate the composition of the TLM sludge layers, based on the volume of each layer in the tank and the general volume of these characteristic layers in all other tanks.

Tanks 241-BX-105 and 241-B-201 were used to estimate the composition of MW and 224 waste. Tanks 241-C-110 and 241-U-110 were used to establish the composition of 1C waste generated between 1944 and 1949 (1C1), with the MW contribution in 241-U-110 being derived from 241-BX-105. Tanks 241-T-104 and 241-BX-107 were used to derive the compositions of 1C waste generated between 1950 and 1956 (1C2), while 241-B-110 and 241-T-111 were used to estimate 2C waste generated between 1944 and 1949 (2C1) and 2C waste generated between 1950 and 1956 (2C2), with PUREX high-level waste generated

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between 1963 and 1967 (P2) and 224 contributions in 241-B-110 and 241-T-111 being derived from the HDW model and 241-BX-105, respectively. Tanks 241-TY-101, 241-TY-103, and 241-TY-104 were used to estimate the composition of ferrocyanide sludge produced by in-plant scavenging of 1C supernatant waste using 0.005M ferrocyanide (1CFeCN) and UR/Tri-butyl phosphate (UR/TBP) waste, with the HDW model being needed to adjust for salt cake generated from 242-T between 1965 and 1976 (T2SltCk) in tank 241-TY-103. Table H-3 compares the HDW model and core sample derived estimates for BiPO₄ waste.

Table H-3. Comparison of Hanford Defined Waste and Core Sample Estimates for BiPO₄ Waste.

BiPO ₄ waste	Fe concentration, kg/kgal (based on TLM)	Fe concentration, kg/kgal (based on tank core samples) ^a	Reference tanks (241-)
MW1	25.1	39.9	BX-105
MW2	0		
1C1	68.3	54.8	C-110/U-110
1C2	37.8	35.4	T-104/BX-107
2C1	118.5	115.5	B-110/T-111
2C2	166.0	57.9	B-110/T-111
224	76.3	71.5	B-201
1CFeCn	22.4	317.5	TY-101/TY-103
UR/TBP	332.6	208.0	TY-104

MW1 = Metal waste from the bismuth phosphate process, 1944 to 1951

MW2 = Metal waste from the bismuth phosphate process, 1952 to 1956

TBP = Tri-butyl phosphate

TLM = Tank layer model

UR = Uranium recovery

1CFeCN = Ferrocyanide sludge produced by in-plant scavenging of 1C supernatant wastes; used 0.005M ferrocyanide

1C1 = First decontamination cycle of the bismuth phosphate process, 1944 to 1951

1C2 = First decontamination cycle of the bismuth phosphate process, 1952 to 1956

2C1 = Second decontamination cycle of the bismuth phosphate process, 1944 to 1951

2C2 = Second decontamination cycle of the bismuth phosphate process, 1952 to 1956

224 = Final decontamination/concentration cycle of the bismuth phosphate process

^a Core sample results derived from Colton 1995.

The results for BiPO₄ wastes are summarized in Table H-4.

Table H-4. Estimated Iron Inventory in BiPO₄ Waste Based on Tank Core Samples.

BiPO ₄ waste	Waste volume, kgal (TLM)	Fe concentration, kg/kgal (core samples) ^a	BiPO ₄ Fe inventory, kg
MW	736	39.9	29,300
1C1	1,732	54.8	94,900
1C2	1,407	35.4	49,900
2C1	762	115.5	88,000
2C2	822	57.9	47,600
224	322	71.5	23,000
1CFeCN	119	380.8	45,300
Total			378,000

MW = Metal waste from the bismuth phosphate process

TLM = Tank layer model

1CFeCN = Ferrocyanide sludge produced by in-plant scavenging of 1C supernatant wastes; used 0.005M ferrocyanide

1C1 = First decontamination cycle of the bismuth phosphate process, 1944 to 1951

1C2 = First decontamination cycle of the bismuth phosphate process, 1952 to 1956

2C1 = Second decontamination cycle of the bismuth phosphate process, 1944 to 1951

2C2 = Second decontamination cycle of the bismuth phosphate process, 1952 to 1956

224 = Final decontamination/concentration cycle of the bismuth phosphate process

^a Core sample results derived from Colton 1995.

This analysis indicates that 378 MT of Fe should be contained in the BiPO₄ tank waste. Based on the flowsheet, about 345.5 MT of Fe were added in the form of ferrous sulfamate to this waste; while the remaining 32.4 MT of Fe were probably produced from corrosion. An estimated corrosion Fe inventory can also be derived from the Ni inventory of the BiPO₄ wastes (based on tank samples), together with characteristic sludge types and volume estimates in the HDW model (Agnew et al. 1996). Based on this approach, the corrosion Fe inventory for BiPO₄ wastes appears to be 20.17 MT. Since the flowsheet and sample derived estimates for BiPO₄ waste yield the highest prediction, this value (32.4 MT) will be used as a basis for estimating the total amount of corrosion Fe in BiPO₄ process waste.

Three of the 17 tanks were not used for core sample analysis because of inconsistencies with other tanks. Tank 241-T-107 was not chosen for 1C1 waste analysis because it contains three times as much Fe, on a 1C1 volume basis, as 241-C-110 and 241-U-110. This suggests that the tank 241-T-107 core sample results or waste description and volume may be in error. Tank 241-T-107 evidently contained 109 kgal of 1C waste in 1976 and 167 kgal of solids by 1980, with subsequent transfers from tank 241-BX-104 in 1973 and salt well

pumping from 1976 to 1977 (Anderson 1991). It is not clear from this description that all of the waste in tank 241-T-107 can be accurately described as 1C1 waste, since Anderson also describes this waste as cladding waste (CW), ion exchange waste, evaporator concentrates (EVAP), and noncomplexed waste (NCPLX). Tank 241-B-111 was not used for analysis of 2C2 waste because the Fe contained in P2 waste had a dominant affect on the analysis for 2C2 waste and the results were not consistent with tanks 241-B-110 and 241-T-111. Finally, tank 241-C-109 was not chosen for analysis of ferrocyanide sludge produced by in-tank or in-farm scavenging (TFeCN) because the large Fe contributions from hot semi-works waste (HS) (Agnew et al. 1996).

H1.1.2 Uranium Recovery Wastes

The UR process began in 1952 to extract uranium from the BiPO_4 MW stored in B, BX, BY, C, T, TX, and U tank farms. Uranium Recovery wastes can be generally described as UR/TBP wastes, scavenged in-plant wastes from the UR process (PFeCN1 and PFeCN2), and TFeCN waste. Wastes described as 1CFeCN waste are not included with the UR wastes.

According to values in the UR flowsheet, the "RA" solvent extraction stream injects 0.05 mol of $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ and 0.1 mol of NH_3SO_3 at a flow value of 50 into the RA column to treat the MW feed that contains 0.26 mol of uranium at a flow value of 104. Essentially, the UR flowsheet requires 0.05 gmol of $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ to treat 0.52 gmol of uranium. On this basis, 167.5 MT of Fe would have been needed to process 7,370 MTU (estimated fraction of MW from sluicing records treated in the UR process).

From process records, Allen estimated the amount of $\text{Na}_4\text{Fe}(\text{CN})_6$ and $\text{K}_4\text{Fe}(\text{CN})_6$ used for in-plant and in-tank scavenging of UR wastes (Allen 1976). Altogether, 395,900 kg of $\text{Na}_4\text{Fe}(\text{CN})_6$ and 60,000 kg of $\text{K}_4\text{Fe}(\text{CN})_6$ were reportedly used. This is equivalent to adding 81.9 MT of Fe to UR waste from scavenging (36 MT from in-tank and 45.86 MT from in-process scavenging). Another 167.5 MT of Fe were added as $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$, bringing the total amount of Fe added to 249.4 MT. Of this total, about 21.7 MT of Fe were transferred to cribs with UR waste, leaving a net total of 227.65 MT of Fe in the UR tank waste, based on this method of calculation.

The HDW model estimates for Fe are summarized in Table H-5.

Table H-5. Iron Inventory Estimates for Uranium Recovery Waste Assumed in the Hanford Defined Waste Model.

	Waste types			
	UR/TBP	PFeCN1	PFeCN2	TFeCN
Date	1952 to 1958	1955 to 1958	1955 to 1958	1955 to 1958
Kgal total waste	23,090 kgal	13,180 kgal	20,540 kgal	11,600 kgal
Kgal solids	636 kgal	359 kgal	437 kgal	112 kgal
Fe, p/M	66,800 p/M	36,400 p/M	38,800 p/M	13,610 p/M
Density	1.31537	1.37007	1.374663	1.4655
Fe, MT	211.5 MT	67.8 MT	88.2 MT	8.55 MT

MT = Metric tons

PFeCN1 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.005M ferrocyanide

PFeCN2 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.0025M ferrocyanide

TBP = Tri-butyl phosphate

TFeCN = Ferrocyanide sludge produced by in-tank or in-farm scavenging

UR = Uranium recovery.

Tanks 241-C-112 and 241-C-105 were used to estimate the composition of TFeCN and PUREX cladding waste generated between 1956 and 1960 (CWP1), respectively, while other common wastes such as PUREX cladding waste generated between 1961 and 1972 (CWP2) and HS waste were derived from HDW model estimates. As previously discussed, tank 241-TY-104 was used to estimate the composition of UR/TBP waste. In-plant scavenging waste from UR that used 0.005M and 0.0025M ferrocyanide (PFeCN1 and PFeCN2, respectively) primarily reside in tanks 241-BY-104, 241-BY-105, 241-BY-106, 241-BY-107, 241-BY-108, and 241-BY-110, along with a substantial volume of BY salt cake (BYSltCk). Tank core samples cannot be used as a basis for estimating the composition of these wastes because representative samples have not been obtained from these tanks.

Table H-6 Comparison of Hanford Defined Waste and Core Sample Estimates for Uranium Recovery Waste.

Uranium recovery waste	Iron concentration, kg/kgal (based on HDW)	Iron concentration, kg/kgal (based on tank core samples)	Reference tanks
UR/TBP	332.58	208.34	241-TY-104
PFeCN1	188.95	-	
PFeCN2	201.89	-	
TFeCN	75.49	159.97	241-C-112

HDW = Hanford Defined Waste

PFeCN1 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.005M ferrocyanide

PFeCN2 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.0025M ferrocyanide

TBP = Tri-butyl phosphate

TFeCN = Ferrocyanide sludge produced by in-tank or in-farm scavenging

UR = Uranium recovery.

The results for Uranium Recovery waste are summarized in Table H-7.

Table H-7. Estimated Iron Inventory in Uranium Recovery Waste.

Uranium recovery waste	Waste volume, kgal (TLM)	Fe concentration, kg/kgal	Uranium recovery Fe inventory, kg
UR/TBP	636	208.3 (Sample)	132,500
PFeCN1	359	188.9 (TLM)	67,830
PFeCN2	437	201.9 (TLM)	88,230
TFeCN	112	160 (Sample)	17,920
Total			306,500

PFeCN1 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.005M ferrocyanide

PFeCN2 = Ferrocyanide sludge produced by in-plant scavenging of waste from uranium recovery; used 0.0025M ferrocyanide

TBP = Tri-butyl phosphate

TFeCN = Ferrocyanide sludge produced by in-tank or in-farm scavenging

TLM = Tank Layer Model

UR = Uranium recovery.

These results show that 306.5 MT of Fe should be found in UR tank wastes. 167.5 MT of Fe were added as ferrous sulfamate to the "RA" solvent extraction stream, 63.9 MT added from the UR scavenging campaign, while 21.7 MT were transferred to cribs. Based on the Ni inventory in tank 241-TY-104, it appears that 44.83 MT of corrosion iron were added to UR/TBP waste and an indeterminate amount of corrosion Fe to other UR wastes (PFeCN1, PFeCN2, and TFeCN).

H1.1.3 Reduction and Oxidation Process Wastes

The REDOX process used solvent extraction to recover uranium and plutonium from irradiated fuel. The process was started in January 1952, at S Plant. According to the Hanford Site production records, 19,790 MTU were processed through the REDOX plant from 1952 to 1966 (Heeb 1994). The REDOX process flowsheets can be used as a basis for estimating the total amount of Fe consumed as $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ in the REDOX process (Merrill and Stevenson 1955, Crawley 1960, Isaacson 1965, Boldt 1966). Based on the flowsheet results in Table H-8, some 33.1 MT of Fe is thought to have been added as ferrous sulfamate to the REDOX process. This corresponds to 1.67 kg of Fe/MTU from chemicals (assuming negligible recycle or rework).

Table H-8. Amount of Iron Consumed in the REDOX Process.

Flowsheet	MTU	Flow, gal/MTU	$\text{Fe}(\text{OH})_3M$	Fe in Neutralized Waste, MT
No. 4 and 5	11,422	977	0.0075	17.69
No. 6	3,104	996	0.006	3.92
No. 7	1,057	545	0.021	2.55
No. 8	1,696	693	0.014	3.48
No. 9A	2,239	638	0.016	4.82
No. 9B	269	638	0.016	0.65
Total	19,787			33.11

Tank 241-S-104 wastes can also be used as a reference baseline for high-level REDOX process wastes (R1 and R2 wastes in the TLM). This tank contains 108 kgal of R1 waste, 24 kgal of REDOX process cladding waste generated between 1952 and 1960 (CWR1), and 161 kgal of salt cake precipitate from self concentration in S and SX tank farms (RSltCk) (Agnew et al. 1996). Based on analysis of three composite core samples, tank 241-S-104

sludge also contains about 2,820 kg of Fe and 91.8 kg of Ni, or 26.11 kg of Fe/kgal of R1 waste (DiCenso et al. 1994).

The corrosion source term can be estimated from the Ni inventory in tank 241-S-104. Nickel was chosen as the principal indicator of corrosion because the only known source of nickel in REDOX process waste, other than incidental contamination and post 1958 aluminum coating waste, is from the corrosion of 304 L stainless steel. Type 304 stainless steel contains 71 wt% Fe, 19 wt% Cr, and 10 wt% Ni. Based on these values, corrosion processes may have added about 652 kg of Fe to tank 241-S-104 waste. If this value is converted into an equivalent source term for all REDOX process wastes, corrosion processes could have added another 8.5 MT of Fe to REDOX process waste. REDOX corrosion processes were relatively mild because 2 mol $\text{Al}(\text{NO}_3)_3$ waste solutions were the only wastes processed in the REDOX waste evaporators.

Based on the results to-date, it appears that REDOX process wastes probably contain about 52 MT of Fe from all sources, including 8.5 MT of corrosion Fe.

H1.1.4 Plutonium-Uranium Extraction Process Wastes

The PUREX plant processing campaign can be broken down into three main production periods: 1983 to 1989, 1965 to 1972, and 1956 to 1964. For the most recent period (1983 to 1989), Fe estimates can be developed from multiple sources of information, including tank core samples, flowsheet estimates and purchase records. Table H-9 summarizes the current estimates for Neutralized Current Acid Waste (NCAW) and Neutralized Cladding Removal Waste (NCRW), based on tank core samples and flowsheet estimates (Hodgson 1995, Ryan 1995, Schofield 1991).

Table H-9. Estimated Amount of Iron in PUREX Process Waste (1983 to 1989).

Tank	Waste type	Total Fe, kg	Fe in Heel,* kg
241-AZ-101	NCAW	24,100 (Hodgson 1995)	5,408 (Hodgson 1995)
241-AZ-102	NCAW	46,800 (Ryan 1995)	4,339 (Ryan 1995)
241-AW-103	NCRW	1,440 (Schofield 1991)	-
241-AW-105	NCRW	517 (Schofield 1991)	-
Total		72,857	9,748

NCAW = Neutralized current acid waste

NCRW = Neutralized cladding removal waste

*The heel is the waste in the tank prior to 1983.

If the heel inventory is subtracted, these results show that 63,100 kg of Fe were added to PUREX process waste from 1983 to 1989. This total includes both chemical and corrosion sources. The essential materials records show that 23,300 kg of iron were used as $\text{Fe}(\text{NH}_4\text{SO}_4)_2$ from 1983 to 1989 (Hogan 1994). Since 6,350 MTU (including cold uranium feed) were processed through the PUREX Plant, the chemical consumption rate must have averaged about 3.66 kg of Fe/MTU. The amount of corrosion Fe therefore equals 63,100 kg to 23,300 kg, or 39,800 kg of iron.

Table H-10 shows the estimated Ni inventory in NCAW and NCRW wastes. From the information contained in this table, it can be established that 4,517 kg of nickel (4,666 kg - 149 kg) were added to the PUREX process waste, presumably from corrosion processes in the plant. Based on the quantities of Ni and Fe in 304 L stainless steel, about 32,100 kg of iron were apparently added to PUREX process waste from corrosion processes. This estimate, based on the Ni content of the waste, is about 19 percent lower than the previous estimate from tank samples, flowsheets and purchase records (39,800 kg of corrosion Fe). Based on an average of these estimates, corrosion processes probably added about 5.66 kg of Fe/MTU to PUREX process waste.

Table H-10. Estimated Amount of Nickel in PUREX Process Waste
(1983 to 1989).

Tank	Waste type	Total nickel, kg	Nickel in heel ^a , kg
241-AZ-101	NCAW	1,074 (Hodgson 1995)	Not determined
241-AZ-102	NCAW	3,165 (Ryan 1995)	149 (Ryan 1995)
241-AW-103	NCRW	355 (Schofield 1991)	Not determined
241-AW-105	NCRW	72 (Schofield 1991)	Not determined
Total		4,666	149

NCAW = Neutralized current acid waste

NCRW = Neutralized cladding removal waste

^aThe heel is the waste in the tank prior to 1983.

The site production records for the next period indicate 24,322 MTU were processed in PUREX (and B Plant) from 1965 to 1972 (Heeb 1995). Purchase records show that 1,041,000 lb of $\text{Fe}(\text{NH}_2\text{SO}_3)_2$ were consumed by these processes over this period of time (McDonald 1968). This corresponds to 106,600 kg of Fe from chemical sources, or 4.38 kg of Fe/MTU, based on the quantity of fuel processed.

The PUREX corrosion term can be estimated from several sources, including the compositions of PUREX acid waste (PAW) and PUREX acidified sludge (PAS) and a sample of 1963 to 1967 vintage P2 sludge from tank 241-AX-104 (Larson 1967, Buckingham et al. 1967, Brevick et al. 1994). About 4,000 kgal of PAW were transferred to B Plant from 1968 to 1972 (Agnew et al. 1995a). This estimate is consistent with the total volume of

PAW produced during this period (based on 378 gal of PAW per ton of uranium and 11,798.8 tons of uranium processed) (Agnew et al. 1995a, Heeb 1995). Another 716 kgal of PAS were also retrieved from the 241-A and 241-AX tank farms and processed through the AR vault and B Plant for Sr separation (Agnew et al. 1995a). Table H-11 shows the average composition of each waste.

Table H-11. Estimated Composition of PUREX Waste (1965 to 1972).

Component	241-AX-104 (Brevick et al. 1994)	PAW ^a (Larson 1967)	PAW (Buckingham et al. 1967)	PAS ^a (Larson 1967)	PAS (Buckingham et al. 1967)
Fe, gmol/L	5.95	0.3	0.315	0.35	0.36
Ni, gmol/L	0.26	0.011	0.016	0.013	0.012
Cr, gmol/L	0.059	0.019	0.032	0.0061	0.21

PAS = PUREX acidified sludge

PAW = PUREX acid waste

PUREX = Plutonium-uranium extraction

^aBased on tank 241-A-103 sludge.

The corrosion term can be estimated from Ni by assuming the remaining Fe represents the chemical term, which is equivalent to 4.38 kg Fe/MTU. Corrosion rates could also be derived from Cr, but the ratio of Fe/Ni is conserved following acid dissolution of the PUREX acidified sludge (PAS) while the ratio of Fe/Cr is not (Larson 1967). Apparently, most of the Cr is water soluble or potentially susceptible to oxidation during the PAS sludge washing process (Larson 1967). The corrosion estimates derived from Ni are presented in Table H-12.

Table H-12. Corrosion Estimates for PUREX (1965 to 1972).

Iron	241-AX-104 (Brevick et al. 1994)	PAW ^a (Larson 1967)	PAW (Buckingham et al. 1967)	PAS ^a (Larson 1967)	PAS (Buckingham et al. 1967)
Chem. Fe, kg/MTU	4.38	4.38	4.38	4.38	4.38
Corrosion Fe, kg/MTU	2.12	1.65	2.67	1.68	1.45
Total Fe, kg/MTU	6.50	6.03	7.05	6.06	5.83

PAS = PUREX acidified sludge

PAW = PUREX acid waste

PUREX = Plutonium-uranium extraction

^aBased on tank 241-A-103 sludge.

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From these results, it appears the corrosion term ranges from 1.45 to 2.67 kg of Fe/MTU, with an average value of 1.91 kg of Fe/MTU. Another set of corrosion estimates was developed from PUREX waste stream compositions from May to October 1962 (Oberg 1962). Based on these results, the corrosion rate was found to be 2.25 kg Fe/MTU before September 1962, when the E-F6 and E-F11 concentrators were used in two stage configuration, and 1.15 kg of Fe/MTU after conversion to single stage operation with the E-F6 concentrator.

Another approach is to consider the evaporator tube bundle replacement statistics for PUREX (Schofield 1989). From 1956 to 1962, 6-H4 (backcycle waste evaporator) bundles, 16-F6 (waste concentrator) bundles and 2-2J8 (intercycle uranium concentrator) bundles were replaced. From 1963 to 1972, 17-H4 bundles, 13-F6 and 1-J8 bundles were changed out. For the last PUREX production campaign, 8-H4, 3-F6 and 3-J8 bundles were replaced. One can estimate the earlier corrosion rates by comparing the number of bundles per MTU replaced from 1956 to 1962 and from 1963 to 1972 to the number per MTU replaced and the known corrosion rates during the last PUREX campaign (1983 to 1989). Based on this approach, the corrosion rate appears to be 1.44 kg of Fe/MTU from 1956 to 1962 and 1.83 kg of Fe/MTU from 1963 to 1972.

A PUREX plant corrosion term of 1.91 kg of Fe/MTU will be assumed for 1965 to 1972, and 2.25 kg of Fe/MTU for 1956 to 1964. Based on the amount of fuel processed, approximately 157,000 kg of Fe were added to PUREX waste from 1965 to 1972 (based on 6.29 kg of total Fe/MTU), with 46,500 kg of this total from corrosion.

The third period covers the PUREX production process from 1956 to 1964. During this period, a total of 42,310 MTU were processed by PUREX (Heeb 1995). Since purchase and consumption records are not available, it must be assumed that the overall production process was much the same as during 1965 to 1972. Based on this scenario, about 95,200 kg of corrosion iron (2.25 kg/MTU) and 185,300 kg of chemical iron (4.38 kg/MTU) were added to the waste, producing a total of 280,500 kg of Fe from 1956 to 1964.

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H2.0 BEST-BASIS FOR ESTIMATING CORROSION-GENERATED IRON, NICKEL, AND CHROMIUM INVENTORIES

Table H-13 summarizes the results from this evaluation.

Table H-13. Global Inventory of Corrosion-Generated Iron, Nickel, and Chromium.*

Process	Iron, MT	Nickel, MT	Chromium, MT
BiPO ₄	32.4	4.6	8.7
Uranium Recovery	44.8	6.3	12.0
REDOX	8.5	1.2	2.3
PUREX (1956 to 1972)	141.7	20.0	37.9
PUREX (1983 to 1989)	36.0	5.1	9.6
Total	263.4	37.2	70.5

PUREX = Plutonium-uranium extraction

REDOX = Reduction and oxidation

*Based on the composition of 304 stainless steel (71 percent Fe, 10 percent Ni and 19 percent Cr).

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APPENDIX I

STANDARD INVENTORY CONTENT LIMITATIONS

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APPENDIX I

STANDARD INVENTORY CONTENT LIMITATIONS

The following sections describe the justification for analytes included in the Standard Inventory.

II.0 STANDARD INVENTORY CONTENT LIMITATIONS ON ANALYTES

This task focuses on providing standard inventory values for those analytes that comprise the bulk of the waste mass. As indicated in column 3 of Table II.0-1, these 28 analytes account for 99.89 percent of the waste (less water) by mass (Shelton 1996). Table II.0-1 primarily includes those components introduced from Hanford waste separation processes. The list is also influenced by safety, risk and regulatory needs, as well as disposal needs.

As depicted in column 5 of Table II.0-1, inventory reconciliation will be conducted on both a global and tank-specific basis for analytes accounting for at least 0.02 weight percent of the waste (column 2). Inventories of analytes accounting for less than 0.02 weight percent will be reconciled on a tank-specific basis only if it is known that the inventory distribution is concentrated in a few of the 177 tanks (e.g., La, Hg). Global inventories for additional minor components (e.g., Cd, Ag, Cu, Se, Te, Co, Be, Ba, etc.) are to be identified through review of fuel fabrication and flowsheet records.

Table II.0-1. Analytes Targeted by the Standard Inventory Task. (2 Sheets)

Column 1	Column 2	Column 3	Column 4		Column 5	
Analyte	%Δ Mass ^a	Σ% Mass	Standard inventory		Cited in	
			Global	Tank	HDW Model Rev. 3 (Agnew et al. 1996)	Shelton 1996
NO ₃	46.83	46.83	X	X	X	X
Na	29.96	76.78	X	X	X	X
OH	8.62	85.41	X	X	X	X
NO ₂	4.12	89.53	X	X	X	X
PO ₄	2.18	91.71	X	X	X	X
Al	2.05	93.76	X	X	X	X
CO ₃	1.39	95.15	X	X	X	X

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Table II.0-1. Analytes Targeted by the Standard Inventory Task. (2 Sheets)

Column 1	Column 2	Column 3	Column 4		Column 5	
Analyte	%Δ Mass ^a	Σ% Mass	Standard inventory		Cited in	
			Global	Tank	HDW Model Rev. 3 (Agnew et al. 1996)	Shelton 1996
SO ₄	0.88	96.03	X	X	X	X
U	0.71	96.75	X	X	X	X
F	0.52	97.27	X	X	X	X
TOC	0.50	97.76	X	X	X	X
Fe	0.34	98.11	X	X	X	X
K	0.32	98.43	X	X	X	X
Zr	0.30	98.73	X	X	X	X
Si	0.30	99.03	X	X	X	X
Cl	0.30	99.32	X	X	X	X
Cr	0.15	99.48	X	X	X	X
Bi	0.11	99.59	X	X	X	X
Ni	0.09	99.68	X	X	X	X
Mn	0.08	99.76	X	X	X	X
Ca	0.07	99.83	X	X	X	X
Pb	0.02	99.85	X	X	X	X
Sr	0.02	99.87	X	X	X	X
La	0.01	99.88	X	X	X	X
Cd	0.01	99.89	X			X
Ce	0.00 ^b	99.89	X			X
Ag	0.00	99.89	X			
Hg	0.00	99.89	X	X	X	X

^aShelton (1996).

^bRanking based on revised global inventory defined by the Standard Inventory effort (8.5 MT).

12.0 STANDARD INVENTORY CONTENT LIMITATIONS ON RADIONUCLIDES

The *Predominant Radionuclides in Hanford Site Waste Tanks* document (Boothe 1996) provides the basis to consider 40 radionuclides as part of the Standard Inventory Estimate task. The predominant radionuclides defined by Boothe (1996) are radionuclides that are estimated to present over 99 percent of the inherent risks in each of five categories: (1) long-term or short-term inhalation hazards; (2) long-term groundwater hazards; (3) long-term external radiation hazards; (4) short-term shielding concerns; and (5) short-term volatile hazards. An additional six (6) radionuclides (Y-90, Ba-137m, Eu-155, Cm-242, Cm-243 and Pu-242) were added to the list of 40 due to their impact on source term. The resulting 46 radionuclides are listed in Table I2.0-1. As depicted in column 3 of Table I2.0-1, approximately 100 percent of the tank waste activity (DOE 1996) is associated with 16 of the 46 radionuclides.

As indicated in Table I2.0-1 (column 4), both global and tank-specific inventories will be developed for each of the 46 radionuclides. The ORIGIN2 (Wittekind 1989) model is utilized to generate the total production values, and the HDW model (Rev. 4) will partition these total inventories among the 177 tanks and cribs. An in-depth discussion of the methodology used to determine radionuclide inventories is included in Section 6.0.

Table I2.0-1. Radionuclides Targeted by the Standard Inventory Task. (3 Sheets)

Column 1	Column 2	Column 3	Column 4		Column 5		
Radionuclide	%Δ Activity ^a	Σ% Activity	Standard inventory		Cited in		
			Global	Tank	HDW model Rev. 4 ^c	Boothe (1996)	Source term
Sr-90	30.39	30.39	X	X	X ^b	X	
Y-90	30.39	60.78	X	X	X ^b		X
Cs-137	19.75	80.53	X	X	X ^b	X	
Ba-137m	18.73	99.26	X	X	X		X
Sm-151	0.35	99.61	X	X	X	X	
Ni-63	0.15	99.76	X	X	X	X	
Am-241	0.06	99.82	X	X	X ^b	X	
Pu-241	0.04	99.66	X	X	X ^b	X	
Eu-155	0.03	99.89	X	X	X		X
Eu-154	0.03	99.92	X	X	X ^b	X	

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Table I2.0-1. Radionuclides Targeted by the Standard Inventory Task. (3 Sheets)

Column 1	Column 2	Column 3	Column 4		Column 5		
Radionuclide	%Δ Activity ^a	Σ% Activity	Standard inventory		Cited in		
			Global	Tank	HDW model Rev. 4 ^c	Boothe (1996)	Source term
Sb-125	0.02	99.94	X	X	X	X	
Tc-99	0.02	99.96	X	X	X	X	
Pu-239	0.01	99.97	X	X	X ^b	X	
Cd-113m	0.01	99.98	X	X	X	X	
H-3	0.01	99.99	X	X	X	X	
Cs-134	0.01	100.00	X	X	X	X	
Co-60	0.00	100.00	X	X	X ^b	X	
Pu-240	0.00	100.00	X	X	X ^b	X	
C-14	0.00	100.00	X	X	X	X	
Ni-59	0.00	100.00	X	X	X	X	
Zr-93	0.00	100.00	X	X	X	X	
Nb-93m	0.00	100.00	X	X	X	X	
Eu-152	0.00	100.00	X	X	X	X	
Pu-238	0.00	100.00	X	X	X ^b	X	
Se-79	0.00	100.00	X	X	X	X	
Sn-126	0.00	100.00	X	X	X	X	
U-238	0.00	100.00	X	X	X	X	
Pa-231	0.00	100.00	X	X	X	X	
Cm-244	0.00	100.00	X	X	X ^b	X	
U-232	0.00	100.00	X	X	X	X	
Np-237	0.00	100.00	X	X	X ^b	X	
Cm-242	0.00	100.00	X	X	X		X
Am-243	0.00	100.00	X	X	X	X	
I-129	0.00	100.00	X	X	X	X	

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Table I2.0-1. Radionuclides Targeted by the Standard Inventory Task. (3 Sheets)

Column 1	Column 2	Column 3	Column 4		Column 5		
Radionuclide	%Δ Activity ^a	Σ% Activity	Standard inventory		Cited in		
			Global	Tank	HDW model Rev. 4 ^c	Boothe (1996)	Source term
Cm-243	0.00	100.00	X	X	X		X
U-234	0.00	100.00	X	X	X	X	
U-235	0.00	100.00	X	X	X	X	
Ru-106	0.00	100.00	X	X	X	X	
Ac-227	0.00	100.00	X	X	X	X	
U-233	0.00	100.00	X	X	X	X	
U-236	0.00	100.00	X	X	X	X	
Pu-242	0.00	100.00	X	X	X		X
Th-229	0.00	100.00	X	X	X	X	
Ra-226	0.00	100.00	X	X	X	X	
Th-232	0.00	100.00	X	X	X	X	
Ra-228	0.00	100.00	X	X	X	X	

^aDOE 1996 and Boothe 1996

^bFinal Safety Analysis Report (FSAR) radionuclides which will receive more in-depth treatment in HDW model, Rev. 4

^cThis table was initially prepared in June 1996. As of this date, the HDW Rev. 4 document had not been issued, but the Standard Inventory team was providing input to support inclusion of these radionuclides in the HDW model (refer to Section 6.0).

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APPENDIX J

GUIDELINES FOR PREPARING BEST-BASIS TANK-BY-TANK WRITE-UPS

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APPENDIX J

**GUIDELINES FOR PREPARING BEST-BASIS
TANK-BY-TANK WRITE-UPS**

J1.0 BACKGROUND

Chemical and radionuclide inventory estimates are generally derived from one of three sources of information: (1) sample analysis and sample-derived inventory estimates, (2) component inventories predicted by the Hanford Defined Waste (HDW) model (Agnew et al. 1996, 1997a¹) based on process knowledge and historical tank transfer information, or (3) tank-specific process estimates based on process flowsheets, reactor fuel data, essential materials records, or comparable sludge layers and associated sample information from other tanks.

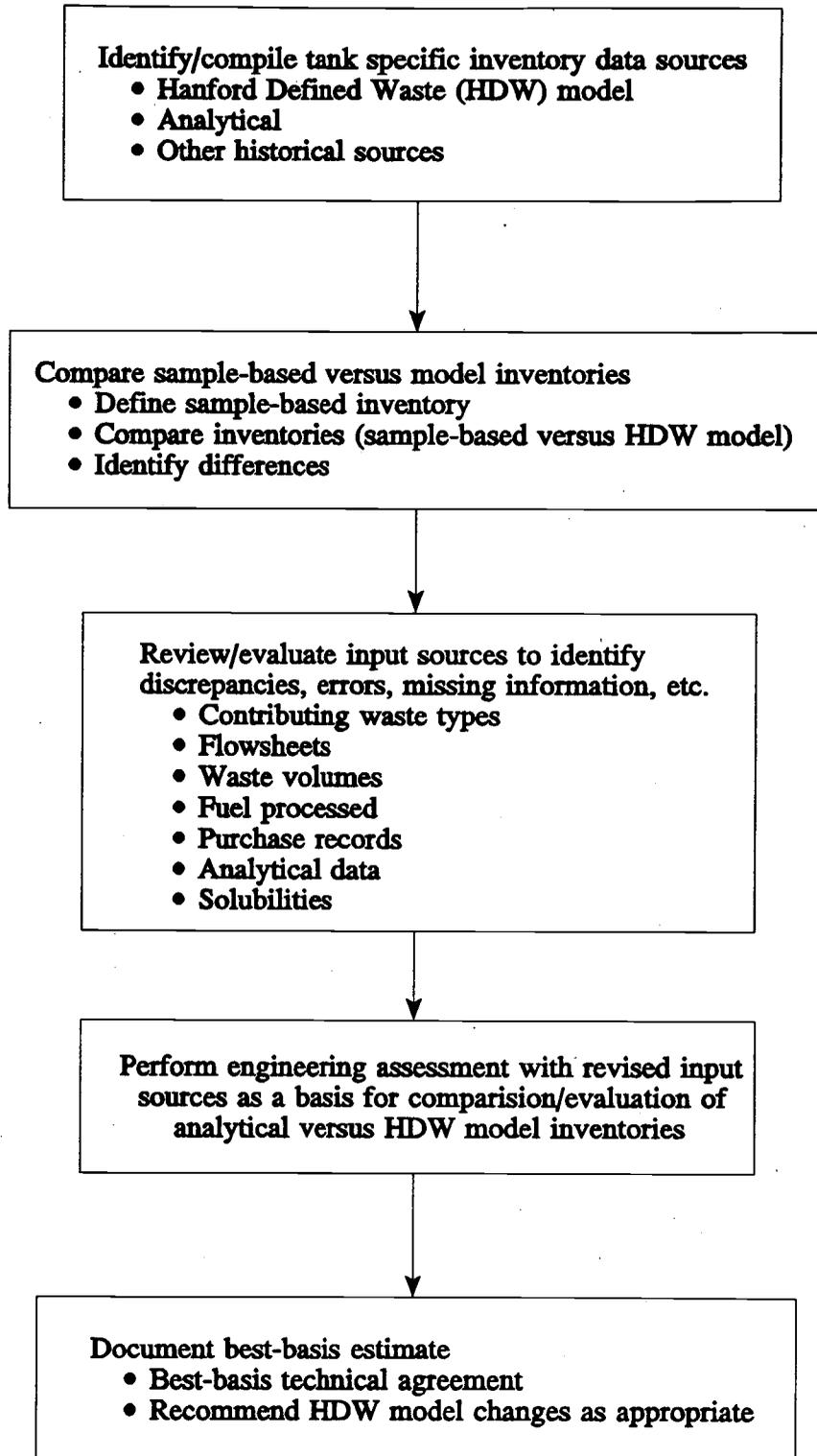
An effort is currently underway to provide waste inventory estimates that will serve as the standard characterization data for various waste management activities. The main objective of this standard inventory is to provide best-basis estimates that will serve as standard inventory values for all waste management and disposal activities. Guidelines for providing best-basis tank-by-tank estimates are described in this appendix. The methodology typically used to reconcile tank-by-tank inventories is depicted schematically in Figure J-1.

The following provides a discussion of the contents and presentation relative to each section of the best-basis tank-by-tank write-up. The intent of these guidelines is to clarify expectations and assure consistency among such write-ups. It should be noted that the amount of available source data information varies significantly from tank to tank (see Section 8.0). Consequently, it may not be possible to adhere to all of the suggested elements of the inventory reconciliation process described in this appendix.

Since the tank-specific best-basis write-ups are intended for publication within Appendix D of the respective Tank Characterization Report (TCR), the following discussion makes reference to the relevant section of a TCR Appendix D.

¹Many of the tank-by-tank best-basis write-ups prepared in fiscal year (FY) 1996, utilized Agnew et al. 1996 data. Tank-by-tank write-ups prepared in FY 1997 utilized data from Agnew et al. 1997. Reference will be made to the most current source document (Agnew et al. 1997) throughout this appendix.

Figure J-1. Tank-by-Tank Reconciliation Process.



revertmain/FIG-J-1.WPG

The approved outline for the best-basis tank-by-tank appendix is as follows:

APPENDIX D
EVALUATION TO ESTABLISH BEST-BASIS INVENTORY FOR
____-SHELL TANK 241-__-__

D1.0 CHEMICAL INFORMATION SOURCES

D2.0 COMPARISON OF COMPONENT INVENTORY VALUES

D3.0 COMPONENT INVENTORY EVALUATION

D4.0 DEFINE THE BEST-BASIS AND ESTABLISH COMPONENT INVENTORIES

D5.0 APPENDIX D REFERENCES

J2.0 INTRODUCTION TO APPENDIX D

The tank number should be clearly identified within the Appendix D title. To be consistent with the TCR outline, the title of Appendix D should be "EVALUATION TO ESTABLISH BEST-BASIS INVENTORY FOR ____-SHELL TANK 241-XX-YYY." The introductory paragraph which follows should clearly state the purpose for the write-up. The purpose is likely to be as follows:

- Review the data, both process-based and sample-based, to identify errors, biases, inconsistencies, and attempt to reconcile them if possible.
- Assess the data in their entirety and define the best-basis inventory (i.e., baseline inventory) for the individual tank.

When working with a "basis" tank (i.e., a tank with one waste type or simple process history) the following may be an additional purpose:

- Generalize findings between sample and process history data and establish a consistent set of assumptions/parameters which can be extended to tanks with no samples or partial samples.

All best-basis evaluations will review and reconcile data sources. They will also define the best-basis inventory. However, some tanks are "basis" tanks and will have a much more detailed narrative (especially regarding process documentation), whereas other tanks will not have this level of detail (see Section 8.0).

J3.0 SECTION D1.0 CHEMICAL INFORMATION SOURCES

List available chemical and radionuclide information sources for the tank waste. This list can be a set of bullets defining each of the samples obtained from the tank along with the corresponding sample date and sample analysis data reference. Sample analysis results for other tanks known to contain the same waste type(s) as the tank being studied should also be listed when used in the component inventory evaluation (Section J5.0). One of the bullets should reference the HDW model results (Agnew et al. 1997a). For a list of the types of references which might be used to prepare a best-basis write-up, see Section J7.0.

J4.0 SECTION D2.0 COMPARISON OF COMPONENT INVENTORY VALUES

This section provides a quantitative comparison of the various analyte inventories available for the tank. Existing tank inventory data should be reported in tabular form. (See example Tables D2-1 and D2-2 below). Normally a sample-based inventory and the HDW model predicted inventory are shown.

Clarify the source of the sample-based inventory by referencing the associated inventory or mean tables which would appear in Appendix B of the TCR (if one is available). State the respective volume and mass bases for both the sample-based and HDW model-based inventories. Note discrepancies between source bases (e.g., density, tank volume), explicitly define the source bases used in the best-basis evaluation, and briefly discuss why. The HDW model bases are consistent with their own assumptions/values.

Comparison need not be limited to the standard inventory analytes. Additional analyte data may be quite useful in assessing the various inventories. (Only results reported in the best-basis summary tables are limited to the standard analyte list, as discussed in Section J6.0.)

If the sample-based inventory is biased (based on other than complete core) and the missing element of the core sample is not represented by the portion of the core sample which was obtained, then clarify within the text the nature of the comparison being made in the table. For example, the core did not include the sludge fraction, therefore, the sample inventory is represented only by the portion of the core sample which was obtained, and available sample data were applied to the full volume in the tank.

Example Table D2-1. Sample- and Hanford Defined Waste Model-based Inventory Estimates for Nonradioactive Components in Tank 241-X-YYY^a.

Analyte	Sample-based ^b inventory estimate (kg)	HDW ^c inventory estimate (kg)
Al	16.75	8.30
...		

^a Concentration is obtained from Table XX-Y, Appendix B. (or reference data package)

^b Mass basis is ___ kg (___ kL of waste with a density of ___ g/mL). See Appendix B for sample information

^c Agnew et al. (1997a).

Example Table D2-2. Sample- and Hanford Defined Waste Model-based Inventory Estimates for Radioactive Components in Tank 241-X-YYY^a.

Analyte	Sample-based ^b inventory estimate (Ci)	HDW ^c inventory estimate (Ci)
¹⁴ C	YXY	ZZZ
...		

^a Concentration is obtained from Table XX-Y, Appendix B. (or reference data package)

^b Mass basis is ___ kg (___ kL of waste with a density of ___ g/mL). See Appendix B for sample information

^c Agnew et al. (1997a).

If there are no data available for analytes of importance, simply state so in the text (e.g., "no inventory data were available for bismuth, lanthanum, phosphate...") rather than listing each as a row within the comparison table. Comparisons may not be applicable if all the sample-based results are "NR." If no sample-based inventory exist (i.e., there is no sample analysis from which to base an inventory), state this in the text and list only the HDW model inventory values in the table. Finally, conclude this section by summarizing pertinent observations, e.g., note major discrepancies between HDW model and sample-based inventory estimates. The discrepancies should be further discussed and reconciled in Section D3.0, Component Inventory Evaluation.

When completing the inventory comparison section, keep the following in mind:

- Reference a single identifiable source (e.g., specific sample basis, HDW model version, etc.) for the given inventory value.
- Clearly note exceptions or basis changes.
- Verify that no concentrations are greater than 1,000,000 $\mu\text{g/g}$.
- Verify that the sum of the analyte concentrations is $\leq 1,000,000 \mu\text{g/g}$.
- Verify that the system is approximately charge balanced (macro scale).
- Check for analyte correlations (e.g. Na and NO_3) and note inconsistencies, biases, errors and differing assumptions based on review of analytical observations, process knowledge/documentation and HDW model results.

J5.0 SECTION D3.0 COMPONENT INVENTORY EVALUATION

This section should present the logical arguments, calculations, and other bases used to arrive at the best-basis inventory for the tank. All assumptions are to be stated and conclusions need to be substantiated. Cite unique circumstances applicable to a tank, document applicable references, and assure that qualitative descriptions are given a quantitative context. In addition, any data interpretation or conclusions should be reasonable in the context of the tank environment or unique conditions (e.g., in-tank heaters used for supernatant evaporation).

J5.1 CONTRIBUTING WASTE TYPES AND VOLUMES

This section should start by reviewing the waste transactions and process history associated with the tank. This is done to verify the Agnew et al. (1997a) tank layers and to revise them if necessary. Much important historical documentation exists that can be used to supplement interpretation of waste transaction records. Tank sample analyses (if available) combined with the process flowsheet basis for the tank waste and the transaction history for the tank can be used to arrive at the basis for the type(s) and amounts of waste presently in the tank.

Section 8.0, "Method for Estimating Individual Tank Waste Inventories," describes in more detail the historical-based and sample-based sources of information that can be used to formulate the basis for tank waste types and volumes. The following sections describe the

engineering assessment-based inventory development (for tanks with little or no sample data) and the inventory reconciliation step.

J5.2 ENGINEERING ASSESSMENT-BASED INVENTORIES

This section of the inventory evaluation reviews available information sources to estimate the best-basis chemical and radionuclide inventory for the tank. Two key sources of information for calculating tank waste inventories are: (1) analytical data from samples taken from the tanks, and (2) the HDW model predictions (Agnew et al. 1997a). The HDW model estimates the inventory for tanks using primarily process history rather than sample data as the basis.

An engineering assessment is also performed, however, to predict the waste inventory. The engineering assessment is an "independent" method for predicting the inventory in a tank based on: (1) process history (e.g., process flowsheets and process throughput) and/or (2) tank sample analyses for other tanks known to contain the same waste type as the tank being evaluated. The process history information assessed as part of the independent engineering evaluation may use different input assumptions than those used by the HDW model (e.g., flowsheet assumptions, component solubilities, etc.).

To determine the best-basis inventory, the engineering assessment-based inventory prediction is compared to and reconciled with the sample-based inventory (if available) for the tank being assessed, and it is also compared to and reconciled with the HDW model estimates for the tank.

For tanks with little or no sample data, it is prudent to utilize process and transaction history to estimate a tank inventory. Based on the common layer principle, it may be possible to state that the composition of the tank's contents (by layer) is the same as (or nearly the same as) a specific "basis" tank(s). In this case, the parameters obtained for the "basis" tank(s) are directly applied to the tank having no analytical data. For example, tank 241-T-105 contains cladding waste (CW), and first and second cycle decontamination waste from the bismuth phosphate process (1C and 2C). The basis tank for CW is 241-C-104; for 1C waste, 241-T-104; and for 2C waste, 241-B-110. The appropriate parameters of tanks 241-C-104, 241-T-104, and 241-B-110 will be applied to tank 241-T-105.

For tanks with partial or biased sample data, specify how it is that you are using the sample data to refine the engineering assessment-based inventories. You may choose to use the available sample data to develop inventories for specific layers of the tank and apply "basis" tank data to those layers for which there is no sample data.

Section 8.0 of this report provides additional detail on the approach used for estimating tank waste inventories.

J5.3 INVENTORY RECONCILIATION

Once the engineering assessment-based inventory has been developed, it is important to identify errors and other reasons for the discrepancies among the various inventories. Review the general process history (transfer activity and waste types), flowsheet data, and HDW model estimates. Consider differences in the following bases when comparing inventories:

- Waste volume
- Waste types used/"basis" tanks identified/used
- Analyte concentrations
- Quality of tank samples (sample type, recovery, analyses performed, etc.).

Verify bases (e.g., volume, density, concentration) used in HDW model, sample-based inventory, and engineering assessment calculations. Reconcile discrepancies first in terms of differences in the bases before attributing these differences to other causes (e.g., lab error, spacial variability, sampling bias, incorrect solubilities, incorrect model assumptions, etc.).

In cases where the sample-based result is considered acceptable and the engineering assessment-based result is a value that is within the uncertainty of the sample-based result, you can accept the sample-based inventory as the best-basis value. Rationale: Defer to sample-based value because you perceive the associated error and biases are acceptable for interpreting the contents of the tank and are less than those associated with the engineering assessment-based value. (The sample-based inventory error band has been quantified whereas the engineering assessment-based result is only qualified.)

For each key analyte, document notable observations based on the comparison of the various inventories and respective bases. Specifically state your conclusions regarding the preferred inventory (sample-based, engineering assessment-based, or model-based) for each analyte. This documentation needs to substantiate conclusions about a preferred inventory, therefore, it must be more than a simple statement of available inventory numbers and whether one is twice as large as another, etc. (Comparisons should be performed on a mass basis for the tank and on a concentration basis for waste types.) Distinguish between conclusions based on fact and those based on informed speculation. Lines of evidence for a hypothesis should be clearly spelled out and supported. Reasonable alternatives should be presented, where possible.

Once the best-basis inventories are determined, the hydroxide inventory can be calculated by performing a charge balance with the valences of other analytes. In some cases, this approach requires that other analyte (e.g., sodium or nitrate) inventories be adjusted to achieve the charge balance. During such adjustments, the number of significant

figures is not increased. This charge balance approach is consistent with that used by Agnew et al. (1997a).

J6.0 SECTION D4.0 DEFINE THE BEST-BASIS AND ESTABLISH COMPONENT INVENTORIES

This section of the tank-by-tank inventory evaluation is intended to summarize the best-basis analyte inventories for a given tank. Section D4.0 should be written as a stand-alone section as it will become the text for Section 3.0 of the TCR. It should summarize the evaluation documented in the previous sections including the data sources used in the evaluation, differences among reported inventories, and conclusions regarding the best-basis inventory values. The best-basis inventory and associated bases should be documented in the inventory summary tables as described in Sections J6.1 and J6.2.

J6.1 BEST-BASIS SUMMARY TABLE

Report your best-basis inventories, associated basis, and relevant comments in the standard tables. If no inventory values are reported, designate the inventory as "NR" for "not reported." The standard best-basis summary tables template is provided in example Tables D4-1 and D4-2.

Example Table D4-1. Best-Basis Inventory Estimates for Nonradioactive Components in Tank 241-XX-YYY (Effective May 31, 1997). (X Sheets)

Analyte	Total inventory (kg)	Basis (S, M, C, or E) ¹	Comment
Al			
Bi			
Ca			
Cl			
TIC as CO ₃			
Cr			
F			
Fe			
Hg			
K			
La			
Mn			

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Example Table D4-1. Best-Basis Inventory Estimates for Nonradioactive Components in Tank 241-XX-YYY (Effective May 31, 1997). (X Sheets)

Analyte	Total inventory (kg)	Basis (S, M, C, or E) ¹	Comment
Na			
Ni			
NO ₂			
NO ₃			
OH _{TOTAL}		C	
Pb			
P as PO ₄			
PO ₄			
Si			
S as SO ₄			
SO ₄			
Sr			
TOC			
U _{TOTAL}			
Zr			

- ¹S = Sample-based
M = Hanford Defined Waste model-based
E = Engineering assessment-based
C = Calculated by charge balance; includes oxides as hydroxides, not including CO₃, NO₂, NO₃, PO₄, SO₄, and SiO₃
NR = Not reported.

Example Table X.Y-Z. Best-Basis Inventory Estimates for Radioactive Components in Tank 241-XX-YYY Decayed to January 1, 1994 (Effective May 31, 1997). (X Sheets)

Analyte	Total inventory (Ci)	Basis (S, M, or E) ¹	Comment
³ H			
¹⁴ C			
⁵⁹ Ni			
⁶⁰ Co			
⁶³ Ni			
⁷⁹ Se			
⁹⁰ Sr			
⁹⁰ Y			
⁹³ Zr			
^{93m} Nb			
⁹⁹ Tc			
¹⁰⁶ Ru			
^{113m} Cd			
¹²⁵ Sb			
¹²⁶ Sn			
¹²⁹ I			
¹³⁴ Cs			
¹³⁷ Cs			
^{137m} Ba			
¹⁵¹ Sm			
¹⁵² Eu			
¹⁵⁴ Eu			
¹⁵⁵ Eu			
²²⁶ Ra			
²²⁷ Ac			
²²⁸ Ra			
²²⁹ Th			

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Example Table X.Y-Z. Best-Basis Inventory Estimates for Radioactive Components in Tank 241-XX-YYY Decayed to January 1, 1994 (Effective May 31, 1997). (X Sheets)

Analyte	Total inventory (Ci)	Basis (S, M, or E) ¹	Comment
²³¹ Pa			
²³² Th			
²³² U			
²³³ U			
²³⁴ U			
²³⁵ U			
²³⁶ U			
²³⁷ Np			
²³⁸ Pu			
²³⁸ U			
²³⁹ Pu			
^{239/240} Pu			
²⁴⁰ Pu			
²⁴¹ Am			
²⁴¹ Pu			
²⁴² Cm			
²⁴² Pu			
²⁴³ Am			
²⁴³ Cm			
²⁴⁴ Cm			

- ¹S = Sample-based
- M = Hanford Defined Waste model-based
- E = Engineering assessment-based
- NR = Not reported.

J6.2 INVENTORY BASIS

Based on the comparisons and conclusions documented for analytes (in Section D3.0), the author will have fixed on the best-basis inventory and corresponding basis (sample-based, engineering assessment-based or model-based). However, as these terms are consistently applied, the following clarification is provided.

- **Sample-Based Inventory Values.** Sample-based inventory values are based strictly on the sample data available for a tank. Such inventories are designated with an "S" in the best-basis inventory summary tables (see Section J6.1).
- **Engineering Assessment-Based Inventory Values.** Engineering assessment-based values are derived by data manipulation on the part of the author (see Section 8.0). If any manipulation was done to augment the sample data for a tank (i.e., to generate an inventory for a missing sample segment) or apply sample analysis results from a "basis" tank to a tank having like waste layers, then the basis for the resulting inventory is engineering assessment-based. Such inventories are designated with an "E" in the best-basis inventory summary tables (see Section J6.1).
- **Model-Based Inventory Values.** Model-based inventory values are those reported in the HDW model Rev. 4 (Agnew et al. 1997a). These inventories are designated with an "M" in the standard inventory summary tables (see Section J6.1).

J7.0 SECTION D5.0, APPENDIX D REFERENCES

The following provides a partial listing of references which may be used to develop best-basis inventories:

- Process-specific references:
 1. Plant Technical Manuals (BiPO₄ plant, U plant, B plant, REDOX, etc.)
 2. Schneider (1951)
 3. Rodenhizer (1987)
- Tank-specific references:
 1. HDW model documents (Agnew et al. 1996, Agnew et al. 1997a)

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2. Waste Status and Transaction Records Summary (WSTRS) (Agnew et al. 1997b)
3. Hanford Tank Content Estimates (HTCEs) and Supporting Documents (Brevick et al. 1997a, 1997b, 1997c, 1997d)
4. Sort on Radioactive Waste Types (SWORT) (Hill, et al. 1995)
5. Anderson (1990)
6. Latest Monthly Waste Tank Summary Report (e.g., Hanlon 1997)
7. Specific TCRs or Analytical Data Reports
8. 200 Area monthly reports regarding wastes/tanks.

J8.0 REFERENCES

- Agnew, S. F., J. Boyer, R. A. Corbin, T. B. Duran, J. R. FitzPatrick, K. A. Jurgensen, T. P. Ortiz, and B. L. Young, 1996, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 3*, LA-UR-96-858, Los Alamos National Laboratory, Los Alamos, New Mexico.
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- Hill, J. G., G. S. Anderson, and B. C. Simpson, 1995, *The Sort on Radioactive Waste Type Model: A Method to Sort Single-Shell Tanks into Characteristic Groups*, PNL-9814, Rev. 2, Pacific Northwest Laboratory, Richland, Washington.

Rodenhizer, D. G., 1987, *Hanford Waste Tank Sluicing History*, WHC-SD-WM-TI-302, Westinghouse Hanford Company, Richland, Washington.

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