

Determination of the Need to Assess the Effects of Reactor Compartment Source Term as Part of the SAC. Rev. 0

Summary

A review of the potential release of chromium and Technetium-99 from reactor compartments undergoing disposal at the Hanford site's 218-E-12B burial ground (Trench 94) was performed. The large inventory of three types of stainless steels making up the reactor compartments will corrode over time. The corrosion process in alkaline Hanford soils will convert the large inventory of elemental chromium to immobile Cr^{3+} species (e.g., $\text{Cr}(\text{OH})_3$). The presence of a combined quantity of elemental iron and other iron species (e.g., magnetite, siderite) at a level almost 10-fold that of the chromium inventory will ensure a highly oxygen-depleted environment. The highly reduced environment assures that the chromium will remain in the immobile +3 state and not subject to migration in the vadose zone. A small amount of chromate (+6 state) contained within each reactor vessel (0.005% of total chromium inventory) would not be available for release within the first 1,000 years. When released, the mobile chromate would be reduced to immobile +3 state when it encountered the large inventory of elemental iron and other reduced iron species. In order for the inventory of Tc-99 (2.2 to 6.6 curies as pertechnetate) to be available for release requires corrosive breaching of the external compartment shell and the shell of the internal reactor vessel. Assuming the most conservative corrosion rate, it would take 1,250 years to breach the external reactor compartment shell. In addition, the highly reducing conditions created by the elemental iron will rapidly reduce the mobile pertechnetate to the highly immobile TcO_2 . Both the Tc and the Cr will remain reduced and immobile until the iron has been completely oxidized. This would require a very long time due to the combination of the huge mass of iron to be reduced, a low infiltration rate of water and a relatively low rate of diffusion of oxygen through the vadose zone. With the length of initial assessment simulations set at 1,000 years, it is concluded that simulations of the reactor compartment source terms for chromium and Tc-99 will not be required for SAC Rev. 0.

Assessment

The groundwater /vadose zone integration project assessment design for the systems assessment capability (SAC) lists 8 radionuclides and 2 chemicals to be investigated as part of the SAC Rev. 0 assessment to begin in FY 2001¹. Of those 10 constituents, two (chromium and Tc-99) make up part of the inventory of reactor compartments undergoing disposal at the 218-E-12B burial ground. The current plan is to dispose of 220 reactor compartments by expanding the current trench or constructing a second trench adjacent to the current trench. When completed, the trench(s) will be backfilled with soil and covered with an engineered barrier. At site closure, the steel inventory for all disposed reactor compartments is estimated at 50,380,000 kg. The elemental composition of those steels are summarized in Table 1.

¹ Aaberg, R.L., R.W. Bryce, A.L. Bunn, P.W. Eslinger, C.T. Kincaid, T.B. Miley, W.E. Nichols, M.C. Richmond and S.F. Snyder. "Groundwater/Vadose Zone Integration Project Assessment Design for the Systems Assessment Capability, Rev. 0 Draft, February 2000.

Chromium Inventory, Speciation and Potential for Subsurface Migration

The chemical forms of chromium present are elemental and Cr^{+6} . Elemental chromium is associated with three steels that make up the structure of the disposed reactor compartments and account for the overwhelming majority of the chromium inventory (4,709,100 kg) (see Attachment A). A small amount of chromate solution (1 kg/reactor compartment or 220 kg total) was used on board as a corrosion inhibitor and constitutes the remainder of the inventory. In the post closure site, chromium will be slowly released (over thousands of years) to the surrounding alkaline unsaturated soil in the form of immobile Cr^{+3} (e.g., CrOH_3) species as a result of corrosion of the reactor compartment steels. As a result of the low solubility of CrOH_3 , this release will occur at concentrations that are below the drinking water standard. Also released will be huge quantities of reduced iron (e.g., as magnetite, siderite) from an elemental iron inventory estimated at 31,905,720 kg or almost 10 times the amount of chromium inventory present (Attachment A). The presence of large quantities of reduced iron will result in the presence of a continuous oxygen depleted environment, making it virtually impossible for chromium present to be oxidized over time to the highly mobile +6 state until the iron has been completely oxidized². This would require a very long time due to the combination of the huge mass of iron to be reduced, a low infiltration rate of water and a relatively low rate of diffusion of oxygen through the vadose zone. If oxidizing conditions were to occur at some distant time in the future, Cr would continue to remain reduced and immobile. It is known that $\text{Cr}(\text{OH})_3$ does not generally re-oxidize in the environment. The small inventory of chromate solution would be required to interact with this corrosion layer where it would be rapidly reduced to immobile Cr^{+3} species. Release to the environment would occur beyond a 1,000 year time frame (see below for Tc-99).

Tc-99 Inventory and Potential for Release from Reactor Compartments

It is estimated that from 0.01 to 0.03 curies of Tc-99 are associated with each disposed reactor compartment. For the anticipated disposal of 220 reactor compartments, this would constitute a total Tc-99 inventory in the range of 2.2 to 6.6 curies. Release of the Tc-99 inventory (as pertechnetate) requires first breaching of the reactor compartment external structure (0.75 in thick steel) followed by breaching of the reactor vessel structure (approximately 6 inches of steel). Assuming a progressive corrosion process and a maximum corrosion rate of 0.0006 in/yr, breach of the reactor compartment external shell would occur in 1,250 years and breach through the reactor vessel structure in 11,250 years. Thus, no release of Tc-99 would occur in the first 1,000 years after disposal. It was also estimated that less than 0.2 % (4,400 to 13,200 μCi) of this inventory would be released from the source term in the first 10,000 years of corrosion².

The highly reducing conditions created by the elemental iron will rapidly reduce any released mobile pertechnetate to the highly immobile TcO_2 . Technetium will

² DOE (1996) Final Environmental Impact Statement on the Disposal of Decommissioned, Defueled Cruiser, Ohio Class, and Los Angeles Class Naval Reactor Plants. DOE/EIS-0259, U.S. Department of the Navy, April, 1996.

remain reduced and immobile until the iron has been completely oxidized. This would require a very long time due to the combination of the huge mass of iron to be reduced, a low infiltration rate of water and a relatively low rate of diffusion of oxygen through the vadose zone. If the reactor compartment area did become oxidizing, TcO_2 could reoxidize to the mobile pertechnetate; however, it is likely that the Tc will be dispersed within a cemented zone of $Fe(OH)_3$ and $Cr(OH)_3$ formed through the oxidation of the reactor compartments. This will significantly inhibit the transport of oxygen to the TcO_2 and the subsequent diffusion of pertechnetate from the oxidized reactor compartments. As a result, release of Tc (if it occurs at all) will take place at some very extended time beyond 1,250 years and at a release rate that is expected to occur very slowly through diffusion from the iron and chromium oxide cement.

Table 1: Elemental Composition of Reactor Compartment Steels

Steel Type ¹	Elements (% Composition)					
	Fe ²	Cr	Ni	Mn	Si	C
304 Stainless	69.0	18-20	8-10.5	2.0	1.0	
Inconel 600	7.0	14-17	76.0			
HY-80	92.6	1.5	3.4	1.5		1.0

¹ Elemental data for Cr, Ni, Mn, Si and C from ASM Metals Handbook , Vols. 4 (1991) and 6 (1994). ASM International, Materials Park, OH.

² Determined by difference (100%- % Cr, Ni, Mn, Si, and C).

Attachment A

Estimates of Chromium (elemental and Cr⁺⁶) and Iron Inventories for Reactor Compartments Disposed at 218-E-12B Burial Ground

Rhoads et al¹ estimated the average reactor compartment to contain 121,000 kg of HY-80 stainless steel, 41,000 kg of Type 304 stainless steel, and 67,000 kg of Inconel 600 steel. The plan is for 220 reactor cores to be disposed at the 218-E-12B Burial Ground (DOE 1996). Therefore, the total estimated inventory of elemental chromium and iron-containing steel in the burial ground is 220 times these estimates (i.e., 26,620,000 kg, 9,020,000 kg, and 14,740,000 kg, respectively).

The elemental composition for each of the stainless steels is summarized in Table 1. For estimating chromium inventories, the high weight percent chromium values from Table 1 were used in the calculations below.

Estimated Total Elemental Chromium Inventory

$$26,620,000\text{kg} \times 0.015 + 9,020,000\text{kg} \times 0.2 + 14,740,000 \text{ kg} \times 0.17 = 4,709,100 \text{ kg}$$

Estimated Total Elemental Iron Inventory

$$26,620,000 \text{ kg} \times 0.926 + 9,020,000 \text{ kg} \times 0.69 + 14,740,000 \text{ kg} \times 0.07 = 31,905,720 \text{ kg}$$

Estimated Total Inventory of Cr⁺⁶ in Reactor Compartments

It is estimated that each reactor compartment contains 1kg of chromium (as Cr⁺⁶) (DOE 1996). Therefore, the total inventory of Cr⁺⁶ is estimated as 220 kg (220 reactor compartments X 1kg Cr⁺⁶/reactor compartment).

¹ Rhoads, K., B.N. Bjornstad, R.E. Lewis, S.S. Teel, K.J. Cantrell, R.J. Serne, L.H. Sawyer, J.L. Smoot, J.E. Szecsody, M.S. Wigmosta, and S.K. Wurstner. Estimation of Release and Migration of Nickel Through Soils and Groundwater at the Hanford Site 218-E-12B Burial ground. PNL-9791, Pacific Northwest Laboratory, Richland, WA, May, 1994).