Glass Formulation and Testing for U.S. High-Level Tank Wastes—Project 17210

JD Vienna, Pacific Northwest National Laboratory, Richland, WA, USA
AA Kruger, U.S. Department of Energy, Office of River Protection, Richland, WA, USA
Hanford History, cont.

2 Million m³

Tank Waste Generated (1944-1988)

- Reprocessed 190,000 m³ (10%)
  - Started 1956

- Disposed to Ground* 455,000 m³ (23%)
  - Started 1945

- Leaked to Ground 3800 m³ (<1%)
  - Started 1956

- Evaporated 1.1M m³ (57%)
  - Started 1951

- 200,000 m³ (10%)
Waste Treatment Plant ~60% Complete

PT: 4 Stories
228' Wide x 570' Long

LAW: 4 Stories
240' Wide x 330' Long

HLW: 4 Stories
275' Wide x 440' Long
High Level Waste Glass

- WTP to start in 2018 (hot ops in 2019)
- Processing complete in ~2045
- Produce 10,000 – 15,000 canisters
  - 4.5 m tall, 0.61 m diameter
  - 3 MT glass per canister
  - 7.5 MT glass/day instantaneous (~5.25 MT glass/day on average)
- Roughly 35 wt% waste loading
- Store on-site until repository is available
WTP Issues

- Mixing and transport of concentrated slurries
- Cleaning of tanks to sufficient level for closing
- Efficiency of pretreatment process
- Need for supplemental low activity waste treatment
- “black cells” sealed shut for the life of plant
- Very broad range of waste chemistry/characteristics
Glass Formulation Approach

- Each glass formulation must simultaneously meet a full set of requirements.
- Requirements are described as mathematical functions of glass composition.
- Numerical optimization is used to adjust additive composition and waste loading while meeting all requirements.
Only a small fraction of the entire composition region was tested, modeled, and qualified for production in WTP.

Need to develop glasses with high waste loading across the entire composition region.

Full Mission Hanford HLW Glass Composition Region

WTP Commissioning Portion of the Region

Conceptual glass composition region

Conceptual glass composition region subdivided into subregions
Grouping of Wastes

Six groups or regions of waste and glass composition were identified.

Projected Waste Oxide Mass

- High-Al2O3: 47%
- High-Fe2O3: 12%
- Spinel limited: 19%
- Cr2O3-SO3 limited: 12%
- P2O5-CaO limited: 9%
- High-Na2O: 1%

11,075 MT

Projected Glass Mass (HTWOS revised constraints)

- High-Al2O3: 44%
- Nepheline or Crystals in melter
- Crystals in melter
- Spinel limited: 15%
- High-Fe2O3: 11%
- Cr2O3-SO3 limited: 9%
- High-Na2O: 1%

28,186 MT
Advanced Glass Formulation Efforts

- Develop high waste loaded glass compositions for selected wastes to define the boundaries
- ~20 glasses formulated and tested at crucible and small melter scale with maximum waste loading
  - high SO\(_3\) (up to 1.9 wt%)
  - high Fe\(_2\)O\(_3\) (up to 20 wt%)
  - high Al\(_2\)O\(_3\) (up to 27 wt%)
  - high Cr\(_2\)O\(_3\) (up to 4 wt%, with low SO\(_3\) and transition metals)
  - high transition metal (Fe\(_2\)O\(_3\), NiO, Cr\(_2\)O\(_3\), MnO)

Full Mission Hanford HLW Glass Composition Region

Region divided into subregions
Data used for current study

red dots represent existing data
Advanced Glass Formulation Efforts

- Develop statistical design of glasses to cover each region in-turn

- Start with high $\text{Al}_2\text{O}_3$ region that represents ~ half the waste and glass mass
Three Year Program Plan

- **Year 1:** Evaluate the range of high alumina HLW compositions and the existing data and models for use in glass formulation and plant operation. Develop glass composition test matrices to fill gaps in current glass property data with one matrix each for WTP and DWPF. Fabricate test matrix glasses and begin their characterization.

- **Year 2:** Complete the characterization of test matrix glasses by performing the following tests: viscosity, electrical conductivity, and crystallinity as functions of temperature, product consistency test and toxicity characteristic leach procedure of quenched and slow cooled glass samples, and additional testing found warranted during the glass testing program.

- **Year 3:** Compile data and perform data screening, generate glass property models, and document the results.
Acknowledgements

Research is funded by the U.S. Department of Energy, Office of River Protection

Research is being performed in a team approach between DOE-ORP, PNNL, and Catholic University of America
Backup Slides
Process Flow Diagram

Glass Former Facility
- Silos
- GFCs

Hopper

Pretreatment
- HBV
- HLW

HLW Vitrification
- MFPV
- MFV
- Melter A
- Off-gas
- Canister

MFPV
- MFV
- Melter B
- Off-gas
- Canister
Composition Averaging and Time Delay

- **MFPV i-3**
- **MFPV i-2**
- **MFPV i-1**
- **MFPV i**
- **MFPV i+1**

- **MFV and Melter**

- **3 d/MFPV Batch**
- **3 cans/Batch**
- **1 can/day/melter**
- **20 min/formulation**

- **Can D-12**
- **Can D-6**
- **Can D-5**
- **Can D-4**
- **Can D-3**
- **Can D-2**
- **Can D-1**
- **Can D**
- **Can D+1**
- **Can D+2**

**Time**
Schematic of Processing Window

- HLW
- Liquidus
- Nominal/Target Composition
- Composition Uncertainty
- Minimum Waste Loading
- Viscosity/Conductivity
- Durability
- Prediction Unc.

Additives (GFC Blends)
Composition Uncertainty

- Mixing/Sampling
- Analytical
- Level Measurement
- GFC Mass and Composition
- Melter Volatility
Basis for Existing WTP HLW Qualified Glass Composition Region

- 6 Contract Specifications
- 6 WASRD Specifications
- 4 Delisting Requirements
- 14 WAPS Specifications
- 10 Reporting Requirements
- 12 Processing Constraints
- IHLW Formulation Algorithm

- Over 300 Fully Characterized Simulant Glasses
- >33 DM-100 Melter Simulant Tests
- 48 Large Scale Mixing and Sampling Tests
- 16 Glass Property Models
- 5 Fully Characterized Actual Waste Glasses
- 30 Pilot Melter Simulant Tests
- Over 200 Melter Feed Rheology Tests