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GLOVE BOX VITRIFICATION SYSTEM FOR LANL TRU WASTE

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and

Steve Bates and Jeff Klinger
Idaho National Engineering and Environmental Laboratory

ABSTRACT

A glove box vitrification system is being fabricated to process aqueous evaporator bottom waste generated at the Plutonium Facility (TA-55) at Los Alamos National Laboratory (LANL). The system will be the first within the U.S. Department of Energy Complex to routinely convert Pu$^{239}$-contaminated transuranic (TRU) waste to a glass matrix for eventual disposal at the Waste Isolation Pilot Plant (WIPP). Currently at LANL, aqueous evaporator bottom waste (TRU waste) is solidified in cement. Radionuclide loading in the cementation process is restricted by potential radiolytic degradation (expressed as a wattage limit), which has been imposed to prevent the accumulation of flammable concentrations of H$_2$ within waste packages. Waste matrixes with a higher water content (e.g., cement) are assigned a lower permissible wattage limit to compensate for their potential higher generation of H$_2$. This significantly increases the number of waste packages that must be prepared and shipped, thus driving up the costs of waste handling and disposal.

The glove box vitrification system that is under construction will address this limitation. Because the resultant glass matrix produced by the vitrification process is nonhydrogenous, no H$_2$ can be radiolytically evolved, and drums could be loaded to the maximum allowable limit of 40 watts. In effect, the glass waste form shifts the limiting constraint for loading disposal drums from wattage to the criticality limit of 200 fissile gram equivalents, thus significantly reducing the number of drums generated from this waste stream. It is anticipated that the number of drums generated from treatment of evaporator bottoms will be reduced by a factor of 4 annually when the vitrification system is operational. The system is expected to undergo preliminary startup in the year 2000, and be fully operational in the year 2002.

INTRODUCTION

Nitric acid dissolution processes are typically used at Technical Area-55 (TA-55) for plutonium recovery (see Figure 1). These solutions are passed through various ion exchange media to recover actinides. Effluents from the ion exchange columns are sent to an evaporator for concentration. The bottoms from the evaporator are discharged to 30 L vertical vessels within a glove box, where they are sampled and stored.
The evaporator bottoms (EVB) are analyzed for actinides, and the results are used for accountability purposes and to determine the volume of waste that can be sent to the cementation process for treatment. The volume of waste per 55-gal. drum of cement is limited by the Safety Analysis Report for the TRUPACT-II Shipping Package (1) requirements for wattage and criticality. The volume limits for typical compositions of EVB are based on wattage considerations. The wattage limit established for Portland cement-based waste forms is 0.9068 watts per 55-gal. drum. During routine operations, the volume of EVB processed is in the range of 30 to 50 L per 55-gal. drum of cement.

The vitrification process will increase the volume of EVB that can be treated in a 55-gal. drum by at least a factor of 4, or 120 to 200 L per 55-gal. drum. This is because the glass matrix is a nonhydrogenous waste form, and the potential for H$_2$ gas generation is reduced. A glass waste form has a wattage limit of 40 watts. Typical glass waste drums will not be wattage limited, but will be loaded to the criticality limit of 200 fissile gram equivalents.

Figure 2 visually depicts the volume reduction realized by the vitrification process. In this case the assumption is that the actinide concentrations in the waste are such that the wattage and criticality limits are not exceeded.

**WASTE/GLASS COMPOSITION**

The compositions of the EVB waste streams vary widely from batch to batch, but are in a large part nitric acid, water, and nitrate salts. Table 1 presents the composition of EVB.
Figure 2. Final Volumes for Cementation and Vitrification of Evaporator Bottoms

Table 1. Composition of Evaporator Bottoms (wt%)

<table>
<thead>
<tr>
<th></th>
<th>Min</th>
<th>Mean</th>
<th>Median</th>
<th>Max</th>
<th>10%</th>
<th>90%</th>
<th>Std dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>A₂O₃</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.01</td>
<td>.001</td>
<td>.001</td>
<td>0.00</td>
</tr>
<tr>
<td>Al₂O₃</td>
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<td>3.34</td>
<td>2.62</td>
<td>45.60</td>
<td>1.46</td>
<td>5.10</td>
<td>4.25</td>
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<tr>
<td>Am₂O₃</td>
<td>0.00</td>
<td>0.03</td>
<td>0.02</td>
<td>0.13</td>
<td>0.01</td>
<td>0.05</td>
<td>0.02</td>
</tr>
<tr>
<td>As₂O₃</td>
<td>0.00</td>
<td>0.01</td>
<td>0.00</td>
<td>0.07</td>
<td>.000</td>
<td>.009</td>
<td>0.01</td>
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<tr>
<td>BaO</td>
<td>0.00</td>
<td>0.01</td>
<td>0.01</td>
<td>0.12</td>
<td>0.00</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>BeO</td>
<td>0.00</td>
<td>0.18</td>
<td>0.08</td>
<td>1.40</td>
<td>0.00</td>
<td>0.51</td>
<td>0.29</td>
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<tr>
<td>CaO</td>
<td>0.00</td>
<td>23.28</td>
<td>23.70</td>
<td>62.72</td>
<td>13.4</td>
<td>30.0</td>
<td>7.53</td>
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<tr>
<td>C₆O</td>
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<td>0.00</td>
<td>0.00</td>
<td>0.12</td>
<td>0.00</td>
<td>0.00</td>
<td>0.01</td>
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<tr>
<td>C₇O₃</td>
<td>0.32</td>
<td>2.69</td>
<td>2.24</td>
<td>10.71</td>
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<td>4.60</td>
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<td>Fe₂O₃</td>
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<td>11.31</td>
<td>9.51</td>
<td>41.24</td>
<td>5.63</td>
<td>19.2</td>
<td>6.47</td>
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<tr>
<td>K₂O</td>
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<td>14.21</td>
<td>12.91</td>
<td>44.42</td>
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<td>8.14</td>
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<tr>
<td>MgO</td>
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<td>39.11</td>
<td>92.57</td>
<td>25.9</td>
<td>51.8</td>
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<tr>
<td>Na₂O</td>
<td>0.06</td>
<td>2.99</td>
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<td>59.09</td>
<td>0.50</td>
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<tr>
<td>NiO</td>
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<td>1.10</td>
<td>6.41</td>
<td>0.70</td>
<td>2.22</td>
<td>0.85</td>
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<td>PbO</td>
<td>0.01</td>
<td>0.10</td>
<td>0.07</td>
<td>0.51</td>
<td>0.03</td>
<td>0.19</td>
<td>0.08</td>
</tr>
<tr>
<td>PuO₂</td>
<td>0.00</td>
<td>0.38</td>
<td>0.23</td>
<td>2.52</td>
<td>0.12</td>
<td>0.84</td>
<td>0.38</td>
</tr>
<tr>
<td>T₂O₅</td>
<td>0.00</td>
<td>0.02</td>
<td>0.01</td>
<td>0.25</td>
<td>0.01</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>U₃O₈</td>
<td>0.00</td>
<td>0.02</td>
<td>0.01</td>
<td>0.10</td>
<td>0.01</td>
<td>0.03</td>
<td>0.02</td>
</tr>
</tbody>
</table>

The variability of the waste coming to the vitrification system has been accounted for in the development of the frit to be used in the process. The composition of approximately 160 different EVB batches was used in the frit development activity (2). This activity was performed
at the Pacific Northwest National Laboratory (PNNL). The constraints used to develop the EVB frit were limited to 1050°C melt temperature, 25 percent waste loading, and compliance of the final glass with the EPA-required leach test (Toxicity Characteristic Leaching Procedure [TCLP]). Glass viscosity at melt temperature was also constrained as a measure of "meltability" for a given composition. Table 2 presents the frit composition for the LANL application. This composition was successful in generating glasses that met the performance constraints for almost 80 percent of the EVB compositions provided in the study. PNNL correlated the data and provided predictive models for key glass parameters based on waste composition.

Table 2. EVB Frit Composition (wt%)

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>70.26</td>
</tr>
<tr>
<td>Li₂O</td>
<td>10.08</td>
</tr>
<tr>
<td>B₂O₃</td>
<td>8.00</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>6.00</td>
</tr>
<tr>
<td>Na₂O</td>
<td>5.66</td>
</tr>
</tbody>
</table>

**EQUIPMENT DESCRIPTION**

Primary design for the TA-55 vitrification system was performed at the Idaho National Engineering and Environmental Laboratory and is documented in the *Final Design Report* (3). The system was designed as a batch operation due to facility restrictions. Operations are allowed in the facility 10 hours per day, and 4 days per week. Equipment must be placed in a safe condition during periods when the facility is unoccupied. Figure 3 depicts the vitrification system.

The vitrification system will be installed in a new glove box system, which will be located within the plutonium processing area of TA-55. The glove box will be located inside a stainless steel-lined room, which is approximately 12 x 14 ft with a 12- to 13-ft ceiling.

EVB is typically generated in 25-L batches, and approximately eight batches are required for one vitrification run. The batches will be blended together based on the PNNL predictive models for glass performance. A set of 10, 50-L pencil tanks will be used to store the EVB until an appropriate blending scheme is determined. Approximately 200 L of EVB will then be transferred to two 125-L melter feed tanks. Frit will be delivered in bulk bags and stored outside the facility. A series of augers and hoppers will be used to transfer the frit to the vitrification system.

The melter system consists of a resistance-heated unit, an alloy lid, and an Inner Can Movement Mechanism (ICMM). The melt can is made of stainless steel with an outer diameter of 16 in. and a height of 27 in. The materials of construction will be chosen based on the single use nature of the process, cost, and thermal/corrosion resistance requirements of the operation. The ICMM is a mechanism that raises the melt can vertically into the heating zone of the melter lowers the processed can down into a cooling jacket, and then allows the can to be moved out from under the melter toward the discharge port of the glove box. A crane is available in the
glove box to lift the cooled can of glass from the ICMM and place it in a standard 55-gal. drum for storage and disposal.

Figure 3. 3-D Drawing of TA-55 Vitrification System

Off-gas generated by the processing of EVB consists primarily of water vapor, CO₂, and NOₓ. The off-gas scrubbing system consists of a quench nozzle, scrub column, and caustic scrub solution recirculation system. A set of interim storage tanks is provided to allow sampling and analysis of the scrub solution before discharge to the radioactive liquid waste treatment facility. Figure 4 shows the major components of the system in a simplified form.
PROCESSING SEQUENCE

Operating constraints within TA-55 will require that EVB be processed in batches. The key operating regions will be evaporation, calcination, and melting. These steps will be repeated approximately three times per melt can of glass. The operating procedure for processing EVB will be as follows:

- A melt can will be moved into the glove box via a 55-gal. disposal drum.
- The can will be lifted out of the drum and loaded into the ICMM.
• The ICMM will move the can and cooling jacket into position below the melter.
• The can will be raised into the melter heating zone.
• Frit will be transferred to the melt can.
• The frit will be heated and EVB will be introduced.
• Additional frit will be added to the can.
• The batch temperature will be raised to between 300 and 600°C – evaporation and calcination.
• The batch temperature will be raised to between 1000 and 1100°C.
• The system will be placed in stand by (power reduced) overnight.

This sequence will be repeated three times until the entire batch has been processed.

Following processing of an EVB batch, the glass will be allowed to cool, placed in the 55-gal. disposal drum, bagged out from the glove box, and sent to storage. The scrub solution will be transferred to the interim storage tanks, sampled and analyzed and discharged to the radioactive liquid waste treatment facility.

The vitrification system will be set up outside TA-55 for System Operability testing. A series of tests will be conducted using surrogate materials to demonstrate the process and fine tune the process control system. The system will then be disassembled and installed at TA-55.

STATUS

The primary system components (i.e. melter, glove box, tanks, off-gas system) have been purchased and delivered. RCRA permit modifications for the new treatment system are in preparation and will be submitted to the state of New Mexico during the first quarter of FY2001. Modifications to the processing room within TA-55 are under way. The glove box vitrification system is scheduled to be operational by fourth quarter of FY2002.

CONCLUSION

The TA-55 vitrification system is being implemented in an effort to reduce the number of drums resulting from the treatment of TRU evaporator bottoms, thus requiring fewer shipments to the WIPP for disposal. The vitrification system will be the first in the DOE Complex to process routinely generated TRU waste.

REFERENCES

