

LA-UR-01-2863

Approved for public release;
distribution is unlimited.

Title:

VITRIFICATION SYSTEM FOR TREATMENT OF PLUTONIUM-BEARING WASTE AT LOS ALAMOS NATIONAL LABORATORY

Author(s):

Ron Nakaoka, Gerry Veazey, Don Mullins,
and Chester Smith, Jr.

Submitted to:

<http://lib-www.lanl.gov/la-pubs/00796079.pdf>

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

VITRIFICATION SYSTEM FOR TREATMENT OF PLUTONIUM-BEARING WASTE AT LOS ALAMOS NATIONAL LABORATORY

Ron Nakaoka, Gerry Veazey, Don Mullins, and Chester Smith, Jr.
Nuclear Materials Technology Division, Los Alamos National Laboratory
Los Alamos, New Mexico USA

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} -bearing transuranic (TRU) waste to a glass matrix for eventual disposal at the Waste Isolation Pilot Plant (WIPP). Currently at LANL, this waste is solidified in Portland 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 non-hydrogenous, 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 currently undergoing non-radioactive operability testing, and will be fully operational in the year 2003.

Keywords: vitrification, glass, glove box, plutonium waste

1. 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.

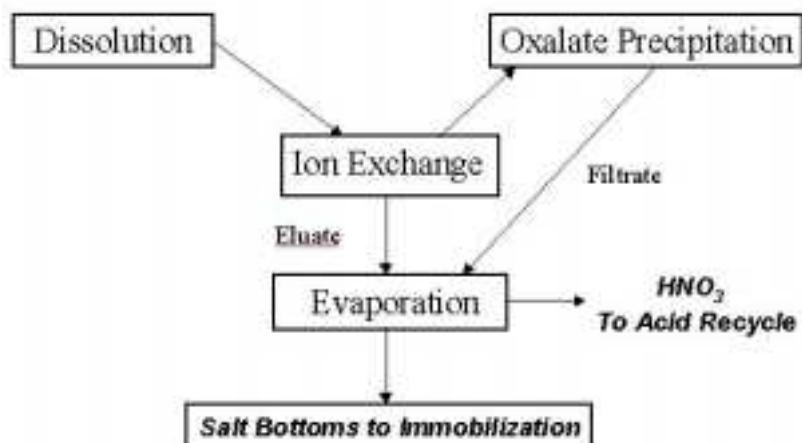


Figure 1. Flowsheet for Nitric Acid Recovery of Plutonium

The evaporator bottoms (EVB) are analyzed for actinides. The results are used for accountability purposes and to determine the volume of waste that can be processed via cementation. 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 non-hydrogenous waste form, and the potential for H₂ 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.

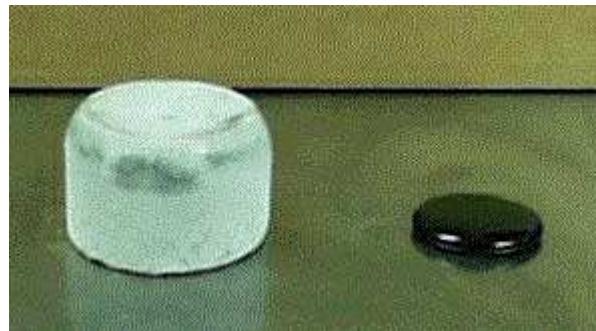


Figure 2. Final Volumes for Cementation and Vitrification of Evaporator Bottoms

2. 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. Data is presented in a waste oxide format as a result of the frit development activity.

Table 1. Composition of Evaporator Bottoms (wt%)

	Min	Mean	Median	Max	10%	90%	std dev
Ag ₂ O	0.00	0.00	0.00	0.01	0.00	0.01	0.00
Al ₂ O ₃	0.00	3.34	2.62	45.60	1.46	31.0	4.23
Am ₂ O ₃	0.00	0.03	0.02	0.13	0.01	0.05	0.02
As ₂ O ₃	0.00	0.01	0.00	0.07	0.00	0.09	0.01
BaO	0.00	0.01	0.01	0.12	0.00	0.03	0.02
BeO	0.00	0.18	0.08	1.40	0.00	0.51	0.29
CaO	0.00	33.28	23.70	62.72	13.4	30.0	7.53
CdO	0.00	0.00	0.00	0.12	0.00	0.00	0.01
Cr ₂ O ₃	0.32	2.69	2.24	10.71	1.35	4.60	1.57
Fe ₂ O ₃	0.00	11.31	9.51	41.24	5.63	19.2	6.47
K ₂ O	0.00	14.21	12.91	44.42	3.30	23.6	8.14
MgO	0.00	40.07	39.11	92.57	23.9	51.8	13.10
Na ₂ O	0.06	2.99	1.11	59.09	0.50	3.33	7.08
Nb ₂ O ₅	0.03	1.36	1.10	6.41	0.70	2.22	0.85
PbO	0.01	0.10	0.07	0.51	0.03	0.19	0.08
PuO ₂	0.00	0.38	0.23	2.52	0.12	0.84	0.38
Ta ₂ O ₅	0.00	0.00	0.01	0.23	0.01	0.03	0.03
U ₃ O ₈	0.00	0.00	0.01	0.10	0.01	0.03	0.02

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 (oxide basis), 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%)

SiO ₂	70.26
Li ₂ O	10.08
B ₂ O ₃	8.00
Al ₂ O ₃	6.00
Na ₂ O	5.66

3. 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 within the plutonium processing area of TA-55. The glove box will be located inside a stainless steel-lined room, which is approximately 12 × 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.

Off-gas generated by the processing of EVB consists primarily of water vapor, CO₂, and NO_x. The off-gas scrubbing system is made up 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.

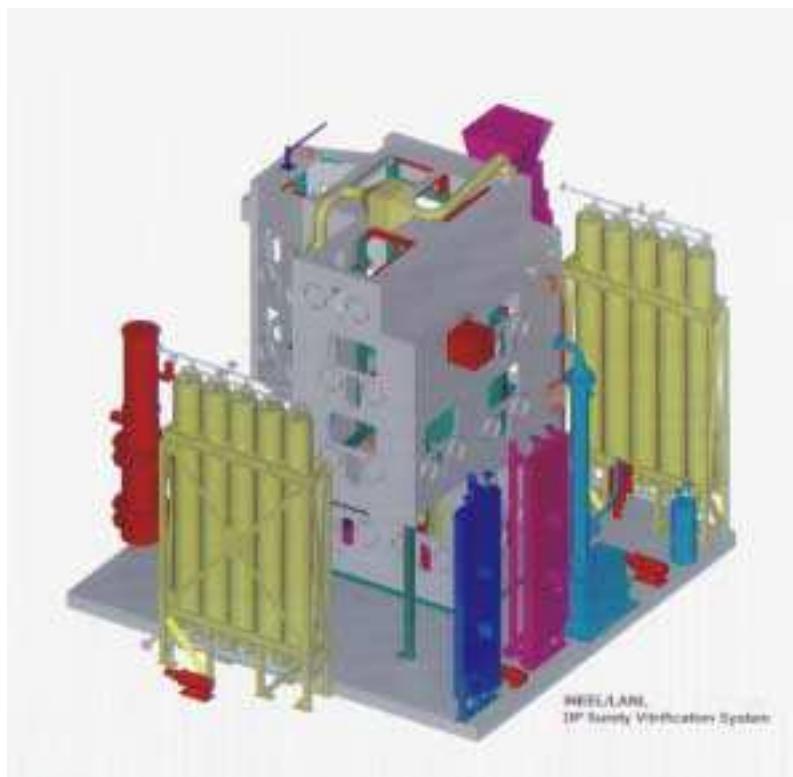


Figure 3. 3-D Drawing of TA-55 Vitrification System



Figure 4. Major Components of TA-55 Vitrification System

4. 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.
- EVB will be introduced, and the frit/EVB mix heated until dry
- Additional frit will be added to the can.
- The batch temperature will be raised to invoke calcination.
- The batch temperature will be raised to between 1000 and 1100°C to melt the mix.
- The system will be placed in stand by (power reduced) overnight.
- This sequence will be repeated two more times until the entire batch has been processed.
- The glass will be held at melt temperature for approximately 4 hrs during the final step to ensure the homogeneity of the final product.

Following processing of an EVB batch:

- The melt can will be lowered into cooling jacket via the ICMM.
- The cooling jacket/melt can will be moved from under the melter.
- The second cooling jacket assembly will be moved into position under the melter.
- The process will be repeated.

The completed melt can 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 has been set up in a non-radioactive area for System Operability testing. A series of tests are being conducted using surrogate materials to demonstrate the process and fine tune the process control system. The system will then be disassembled and installed in the Plutonium Facility.

5. STATUS

The primary system components (i.e. melter, glove box, tanks, off-gas system) have been purchased and delivered. The RCRA permit application for the new treatment system has been submitted to the state of New Mexico. Modifications to the processing room within TA-55 are complete. System Operability testing with the primary processing equipment (melter, off-gas system etc.) is currently under way. The glove box vitrification system is scheduled to be operational by the end of FY2003.

6. CONCLUSION

The TA-55 vitrification system is being implemented in an effort to reduce the number of drums resulting from the treatment of Pu-bearing (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

1. NRC, *Safety Analysis Report for the TRUPACT-II Shipping Package*, Docket No. 9218, Current Revision, Washington, DC, U.S. Nuclear Regulatory Commission, 1994.
2. J.D. Vienna et al., *Glass Development for Treatment of LANL Evaporator Bottoms Waste*, PNNL-11865, Pacific Northwest National Laboratory, 1998.
3. S.O. Bates and J.B. Klinger et al., *Final Design Report*, January 2000.