Use of the Existing Sheilded Cells Melter for CST Vitrification

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INTRODUCTION

Oak Ridge National Laboratory (ORNL) and SRTC are participating in a joint project in which supernate waste from the Melton Valley Storage Tanks at Oak Ridge (OR) will be treated by passage through a crystalline silicotitanate (CST) ion exchange medium[1]. The CST was designed to sorb cesium, the primary radionuclide (Cs-137) in the supernate of the Melton Valley tanks. A smaller amount of strontium will also be sorbed. The loaded sorbent will then be shipped to SRTC where it will be mixed with glass formers and fed as an aqueous slurry to a joule-heated melter within the SRTC Shielded Cells. The molten glass (~ 1150°C) will be poured into 500 mL stainless steel beakers which in turn will be placed in 30 gallon drums for shipment to and disposal at the Nevada Test Site (NTS). This paper focuses on the requirements necessary for disposal of the vitrified CST at NTS. This work is funded by the Tank Focus Area with additional funding from EM-30 at OR.

A reduction in scope is currently under consideration for the vitrification demonstration. This change in scope would reduce the number of drums sent to SRTC from seven to one. The amount of CST that would be vitrified in this case is ~ 38 Kg. If this scope change is realized, then the vitrified CST in the 500 mL beakers will be disposed of at Savannah River Site (SRS). The results presented in this report will also be useful if the vitrified waste remains at SRS.

The Shielded Cells Melter currently contains glass produced during a 1995 DWPF demonstration campaign. That campaign incorporated radioactive Tank 51 sludge into a DWPF borosilicate glass. The Tank 51 campaign in the Shielded Cells Melter was preceded with a flushing of the melter using non-radioactive glass. This flushing was preceded by a different Tank 51 campaign again using borosilicate glass. The 1995 Tank 51 campaign and the melter flushing each used less than one melter volume of material. This implies that the glass currently in the melter is a composite of glass from both Tank 51 campaigns and from melter flushing. If the vitrified CST will be shipped to NTS, the radioactive glass currently in the melter must be removed prior to the CST vitrification demonstration in order to avoid commingling of the waste.

This report (1) estimates the concentrations of the radioactive and RCRA hazardous components of the glass now contained in the Shielded Cells melter, (2) presents the requirements for displacing the existing glass with non-radioactive glass to ensure that radionuclide and RCRA metal levels are below the Waste Acceptance Criteria (WAC) limits set by NTS, (3) identifies the risks associated with a melter flushing strategy and (4) considers the alternative of installing a new melter within the Shielded Cells.

STATUS OF MELTER

The melter within the Shielded Cells contains ~ 10 Kg of molten glass at ~ 1150°C. Between campaigns the normal operating procedure is to maintain the melter at 1050°C with a full load of glass.

Radioactive Tank 51 sludge[2] was used in the last melter campaign, which produced 9.1 Kg of glass. This Tank 51 campaign was preceded with a 6.95 Kg flushing of the melter using nonradioactive glass frit. This flushing was preceded by an earlier Tank 51 campaign, using ~ 31 Kg of borosilicate glass. Since the nonradioactive glass frit did not contain radionuclides or RCRA metals, the only source for introduction of these constituents is Tank 51 sludge.

Material flow through the melter can range from plug flow to a continuously stirred tank flow. For this report the more conservative material flow, continuously stirred tank flow, was used to estimate the current constituents within the melter. This approach implies that the waste is composed of ~80% radioactive waste glass (containing ~25 wt% Tank 51 sludge) and ~20% non-radioactive glass. A conservative approach was taken by assuming that the melter currently contains 100% radioactive waste glass with a 25 wt% loading of Tank 51 sludge.

RADIONUCLIDE INVENTORY

The results of radionuclide analyses are presented in two groups. The first group lists transuranic waste, and the second lists the remaining radionuclides.

Group 1. Transuranics.

The NTS definition of transuranic waste is:

> TRANSURANIC (TRU) WASTE: Radioactive waste containing alpha emitting radionuclides having an atomic number greater than 92, and half-lives greater than 20 years, in concentrations greater than 100 nCi/g.

Table 1 lists the transuranics (including Cm-244, even though it has a half life of 18.1 years) present in washed Tank 51 sludge.[2] This Table also contains the concentrations of transuranics present in the waste glass (assuming 25 wt% Tank 51 sludge).

TABLE 1 - Alpha Emitting Transuranic Concentrations Present in Tank 51 Sludge (washed and dried) and in the Borosilicate Glass

Radionuclide	Half-Life years	Tank 51 nCi/g	Glass nCi/g
Np-237	2.1 E+06	1.4 E+01	3.5 E+00
Pu-238	8.8 E+01	1.4 E+05	3.5 E+04
Pu-239	2.4 E+04	6.9 E+03	1.7 E+03
Pu-240	6.6 E+03	1.8 E+03	4.5 E+02
Pu-242	3.7 E+05	1.6 E+00	4.0 E-01
Am-241	4.3 E+02	6.2 E+03	1.6 E+03
Am-243	7.4 E+03	3.8 E+01	1.0 E+01
Cm-244	1.8.E+01	1.2 E+04	3.0 E+03

The total alpha for transuranics in the glass listed is ~42,000 nCi/g. The NTS requirement limits the TRU component of the waste to less than 100 nCi/g. In order to ensure that this limit is not exceeded, an operational limit of ~ 50 nCi/g is proposed. To achieve the proposed operational limit, the glass in the melter must be displaced by the nonradioactive frit such that the TRU radionuclide concentration is reduced by a factor of 840. That is, 99.88% of the radionuclides must be removed prior to start of the CST vitrification demonstration. A conservative approach is to require a reduction of the TRU components by a factor of 1000.

Group 2. Other Radionuclides.

The concentrations for the remaining radionuclides[2] in the washed Tank 51 sludge and in the glass are presented in Table 2. (The radionuclide content in Ci/Kg was converted to Ci/m^3 using a glass density of 2.7 g/cc.) These radionuclides emit primarily beta-gamma radiation. For purposes of the calculations, it is conservatively estimated 100% of the elements listed here are present in the glass (at 25% waste loading) within the Shielded Cells Melter.

TABLE 2 - Beta-Gamma Emitting Radionuclide Concentrations in Washed Tank 51 Sludge and in DWPF Glass

Radionuclide	Sludge Ci/kg	Glass Ci/kg	Glass Ci/m^3
Ni-59	5.40E-05	1.40E-05	2.70E-03
Ni-63	9.60E-03	2.40E-03	6.50E-00
Se-79	2.20E-05	5.00E-06	1.40E-01
Sr-90	6.40E-01	1.60E-01	4.32E+02
Zr-93	5.40E-05	1.35E-05	3.65E-02
Nb-93m	3.10E-05	7.75E-06	2.10E-02
Tc-99	2.20E-04	5.50E-05	1.50E-01
Pd-107	3.80E-07	9.50E-08	2.60E-04
Sn-126	6.50E-06	1.63E-06	4.40E-03
Cs-135	2.50E-07	6.25E-08	1.70E-04
Cs-137	6.30E-02	1.60E-02	4.32E+01
Sm-151	1.20E-02	3.00E-03	8.10E+00
Th-230	4.10E-08	1.03E-08	2.80E-05
U-234	2.50E-05	6.25E-06	1.70E-02
U-238	1.10E-05	2.75E-06	7.40E-03

The contribution of the beta-gamma emitters present in the Tank 51 glass to the total radioactivity of the CST glass will be small. The removal of 99.9% of the radionuclides (as a result of the 1000 fold reduction for TRU components) would leave ~0.4 Ci/m^3 of Sr-90, the major radionuclide. This value is insignificant relative to the Class C limit of 4600 Ci/m^3 .

RCRA METAL CONCENTRATIONS

The glass within the melter contains RCRA metals from the Tank 51 sludge.[2] Displacement of the current waste glass with a characteristically non-hazardous (from a RCRA point of view) glass frit will reduce the RCRA metals. The estimated RCRA metal concentrations currently in

the melter are listed in Table 3. (As with the radionuclides, an assumption was made that the melter contains waste glass with a 25 wt% loading of Tank 51 sludge). These values were obtained by dissolution and analysis of the vitrified Tank 51 SME product.

TABLE 3 - RCRA Metals Present in the SME Product for the 1995 Washed Tank 51 Sludge Campaign

Metal	Concentration in Glass wt%	ppm	Regulatory Limit (ppm)
Barium	0.012	120	100
Cadmium	0.037	370	1
Chromium	0.196	1960	5
Mercury	0	0	0.2
Silver	0	0	5
Lead	0.05	500	5

The glass in the melter must be displaced with a non-radioactive, RCRA metal-free glass, such that the worst case metal (chromium) is reduced below the regulatory limit of 5 ppm. This will lead to a new glass within the melter which is characteristically non-hazardous (RCRA). To reach the 5 ppm level, the displacement with frit must reduce the level of chromium by a factor of 392. In order to ensure that this limit is not exceeded, an operational limit of 2.5 ppm is proposed. This requires that the factor of 392 be doubled to 784. Therefore, a reduction by a factor of 1000 (path forward for the transuranics) will reduce the RCRA metal levels below the threshold values of characteristically hazardous materials.

OVERALL DISPLACEMENT

The displacement of the glass currently in the melter by a new nonradioactive glass frit must reduce radionuclides and RCRA metals to levels that meet the NTS WAC if the vitrified CST will be shipped to NTS. To achieve this, both the transuranics and RCRA metals must be reduced to concentrations that are approximately 1000 times less than currently in the glass within the melter. The value of 1000, although conservative, will be used in the discussions that follow.

MELTER FLUSHING

Glass frit can be fed to the melter to displace the existing waste. The amount of frit that must be introduced and processed depends upon the mechanism of mixing within the melter. If the two extremes of mixing are plug flow and continuously stirred tank flow, then the continuously stirred tank model is more conservative. If one melter turnover (defined as the melter capacity, which in this case is ~10 kg) displaces 50% of the radionuclides and RCRA metals, then 10 melter turnovers will be required to reduce the radionuclides and RCRA metals to acceptable levels. The actual displacement by one melter volume is greater than 60% for continuously stirred tank mixing. Therefore, the 50% assumption can be considered conservative.

A potential risk with flushing is that regions within the melter where convection and mixing may be slower (including the wall/glass interface) could prolong the release of radionuclides and RCRA metals. If dried slurry/glass adheres either to the refractory above the molten glass line, the melter lid, or the electrodes, there is also the possibility that pieces of this glass or dried slurry could break free from these locations and enter the melt pool.

An estimate has been made for the amount of waste glass (containing 25 wt% Tank 51 sludge) which must be introduced into the melt to cause radionuclides or RCRA metal limits to be exceeded. Using the concentration level of Pu-238 in the glass, ~25 grams of Tank 51 containing glass, which if thoroughly and instantly mixed with the melt pool, would cause the entire pool to reach the TRU limit of 100 nCi/g. Since the density of glass is ~2.5, this corresponds to ~10 mL of glass.

The type of non-radioactive glass used to flush the melter is also important. Certain glasses may more readily remove or displace the current glass in the melter. However, the glass in the melter must be compatible with the feed introduced during the vitrification of the CST sorbent. The preferred way of accomplishing this is to use a frit that is identical to the expected CST waste glass formulation (without radionuclides).

During the process of flushing the melter, samples will be taken from the glass pour stream. These samples will be dissolved and analyzed for RCRA metals and radionuclides to determine the efficiency of the flushing. The actual number of melter turnovers required will not be based on the material flow models, but on the analytical results of the glass samples.

INSTALLATION OF A NEW MELTER

An alternative to the approach of melter flushing is to install a new melter in the Shielded Cells. This is a non-trivial job which would require additional time. The existing melter would still need to be flushed with nonradioactive material and then decoupled from the off-gas system, removed, and dispositioned. A new melter (one new melter has already been fabricated and is available) would then have to be baked to remove the residual water in the refractory. Nonradioactive testing would then be required to ensure that the system functions as intended. This approach would eliminate the concern of exceeding the NTS limits for TRU and RCRA metals due to contamination.

CONCLUSIONS

In order to meet the NTS WAC for the CST vitrification demonstration, the existing melter within the Shielded Cells must first be flushed with non-radioactive, non-hazardous glass. Due to the high levels of TRU and RCRA metals in the existing glass, ~ 10 melter volumes of material need to be processed to ensure acceptable TRU and RCRA metals levels. A strategy must be developed and implemented to provide the best opportunity for realizing this goal. This may include some periods or equilibration between pours, lowering and/or increasing the molten glass level in the melt pot, and use of different non-radioactive glasses. Sampling and analysis of the glass from the pour stream will be required such that the efficiency of the flushing process can be monitored. It is anticipated that the analytical feedback will provide direction to the actual approach.

At a melter throughput of ~1 pound of glass per hour at 50% attainment (24 hours per day), flushing would take ~ 20 days. Additional steps in the protocol, such as equilibration, sample analysis, etc., will increase the overall time for melter flushing. Once an acceptable level of radionuclides and RCRA metals is obtained, there is still the risk, as discussed above, that pieces of glass or dried slurry could release, mix with the glass, and cause the waste glass to exceed NTS limits.

If a reduced scope of this task is realized, then the vitrified CST will be disposed of at SRS. In this case, the degree of flushing required will depend upon (1) SRS requirements for waste disposal and (2) task objectives for obtaining required data on the vitrified CST (i.e., the degree of contamination of the glass samples that can be tolerated). A separate report detailing these items will be issued.

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