Direct Feed Options for Hanford High Level Waste Vitrification-17318

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ABSTRACT

In the high level waste (HLW) direct feed (DF) option that is under consideration for early operations of the Hanford Tank Waste Treatment and Immobilization Plant (WTP), the pretreatment facility would be bypassed in order to support an earlier start-up of the HLW vitrification facility. For HLW, this would mean that the ultrafiltration and caustic leaching operations that would otherwise have been performed in the pretreatment facility would either not be performed or would be replaced by an interim pretreatment function (in-tank washing and settling, for example). These changes could result in higher aluminum contents, higher chromium contents, a higher fraction of supernate in the HLW feed, and lower solids contents. This would likely affect glass formulations and waste loadings and have impacts on the downstream vitrification operations. In the present work, new high waste loading glass formulations have been developed and characterized for the DF HLW option. A series of scaled melter tests was then conducted to assess the impacts of increased supernate content resulting from less effective washing on wastes from candidate source tanks for the direct feed option. The melter tests were conducted at solids contents extending below the current WTP baseline, which are likely for the direct feed option. The effects on glass production rate, melter operations, and off-gas carryover were determined, as was the ability of increased bubbling to compensate for the increased evaporative load. A series of waste compositions were investigated that span the range of in-tank washing efficiencies between the baseline WTP full-wash case and the no-wash case. The tests included wastes from tank AY-102 as well as high plutonium wastes from tank A-104. The results from this work provide the initial basis for assessments of the relative merits of progressively more intensive pretreatment in HLW direct feed options versus the practical benefits of minimal pretreatment. Together, these results illustrate that the optimum washing strategy will depend on the specific tank waste composition and will need to be evaluated on a tank-by-tank basis.

INTRODUCTION

About 56 million gallons of high-level mixed waste is currently stored in underground tanks at the United States Department of Energy's (DOE's) Hanford site in the State of Washington. The WTP will provide DOE's Office of River Protection (ORP) with a means of treating this waste by vitrification for subsequent disposal. In the WTP Pretreatment Facility, the tank waste will be separated into low activity waste (LAW) and high level waste (HLW) fractions, which will then be separately converted to glass by vitrification. The LAW glass will be disposed in an engineered facility on the

Hanford site while the HLW glass is designed for acceptance into a national deep geological disposal facility for high-level nuclear waste.

Technical issues with WTP Pretreatment Facility have led to the adoption of a Direct Feed LAW (DF LAW) approach in order to avoid further delays in the startup of LAW vitrification. In that approach, the pretreatment facility is bypassed and LAW is fed to the vitrification facility with minimal in-tank or near-tank pretreatment, which will likely involve ion-exchange and solids removal. While implementation of the DF LAW is underway, consideration has also been given to the possibility of a parallel direct feed approach for HLW (DF HLW) [1], which is the subject of the present work.

In the HLW direct feed option that is under consideration for early operations of the WTP, the pretreatment facility would be bypassed in order to support an earlier start-up of the HLW vitrification facility. For HLW, this would mean that the ultrafiltration and caustic leaching operations that would otherwise have been performed in the pretreatment facility would either not be performed or would be replaced by an interim pretreatment function (in-tank washing and settling, for example). These changes would likely affect glass formulations and waste loadings and have impacts on the downstream vitrification operations. Modification of the pretreatment process may result in: (i) Higher aluminum contents if caustic leaching is not performed; (ii) Higher chromium contents if oxidative leaching is not performed; (iii) A higher fraction of supernate in the HLW feed resulting from the lower efficiency of in-tank washing; and (iv) A higher water content due to the likely lower effectiveness of in-tank settling compared to ultrafiltration. The HLW direct feed option has also been proposed as a potential route for treating HLW streams that contain the highest concentrations of fast-settling plutonium-containing particles, thereby avoiding some of the potential issues associated with such particles in the WTP Pretreatment Facility [1].

Previous analysis of potential HLW blending and processing strategies concluded that about 50% of the radioactivity in the Hanford tanks can be processed over a three year period at the WTP using the HLW direct feed option [2]. Various optimized blending and washing strategies were developed for those streams and preliminary HLW glass formulations were developed [3]. That work was based on HLW blends and wash options that represent a large proportion of the waste from candidate source tanks that were identified in a tank waste classification study [4]. To further develop this approach new high waste loading glass formulations have been developed and characterized for the DF HLW option [5, 6]. A series of scaled melter tests was then conducted to assess the impacts of increased supernate content resulting from less effective washing on wastes from candidate source tanks for the direct feed option. The melter tests were conducted at solids contents extending below the current WTP baseline, which are likely for the direct feed option. The effects of this on glass production rate, melter operations, and off-gas carryover were determined, as was the ability of increased bubbling to compensate for the increased evaporative load. The tests also evaluated processability and the need for redox control resulting from the higher levels of nitrates from the increased supernate fraction. A series of waste compositions were investigated that span the range of in-tank washing efficiencies between the baseline WTP full-wash case and the no-wash case. The tests included simulated wastes from tank AY-102 as well as high plutonium wastes from tank A-104. This work builds on previous work performed to increase waste loadings in HLW glass formulations and processing rates [7-16] and to support the HLW direct feed option [5, 6].

SYSTEM AND TEST DESCRIPTION

Melter System

Testing was performed on a continuously-fed DM100 joule heated ceramic melter (JHCM) system and off-gas treatment system [5, 6]. The DM100 is a ceramic refractory-lined melter fitted with five Inconel 690 plate electrodes that are used for joule-heating of the glass pool and a bubbler for stirring the melt to increase the glass production rate. Thermocouples installed in a thermowell provide temperature measurements at various locations within the glass pool and in the plenum space. The nominal operating temperature is 1150^{o}C. The glass product is removed from the melter periodically by means of a prototypical air-lift discharge system. The DM100 unit has a melt surface area of 0.108 m^2 and a glass inventory of about 170 kg. The off-gas system maintains the melter under slight negative pressure (typically about 2 inches W.C.) with respect to ambient. The melter feed is introduced in batches into a feed container that is mounted on a load cell for weight monitoring. The feed is stirred with a variable speed mixer and constantly recirculated except for periodic, momentary interruptions during which the weight is recorded. Feed is directed from the recirculation loop that extends to the top of the melter and then diverted through a peristaltic pump, which regulates the flow of feed through a Teflon-lined feed line and water-cooled feed tube into the melter.

The melter is equipped with a dry off-gas treatment system involving gas filtration operations only. Exhaust gases leave the melter plenum through a film cooler device that minimizes the formation of solid deposits. The film-cooler air has constant flow rate and its temperature is thermostatically controlled. Consequently, under steady-state operating conditions, the exhaust gases passing through the transition line (between the melter and the first filtration device) can be sampled at constant temperature and airflow rate. The geometry of the transition line conforms to the requirements of the 40-CFR-60 air sampling techniques. Immediately downstream of the transition line are cyclonic filters followed by conventional pre-filters and high efficiency particulate air (HEPA) filters. The temperature of the cyclonic filters is maintained above 150°C while the temperatures in the HEPAs are kept sufficiently high to prevent moisture condensation. The entire train of gas filtration operations is duplicated and each train is used alternately. An induced draft fan completes the system.

A variety of sampling points are available on the DM100 system that permit sampling of the melter feed, glass product, glass pool, and off-gas. Off-gas characterization included both isokinetic sampling of melter exhaust as well as continuous emissions monitoring using Fourier transform infra-red spectroscopy (FTIR) of a wide variety of gaseous species, including NO, NO_2, N_2O, CO, and SO_2. The glass product from the melter tests was characterized for composition as well as for compatibility with the

WTP processing and product quality requirements and for secondary phases. Analyses of glass and off-gas samples provided the necessary data for mass balances around the melter for key constituents. The conditions prevailing in the vitrification system during these tests were characterized by a variety of temperatures, pressures, flow rates, voltages, currents, and other data as well as visual observations that were recorded throughout the tests.

Waste Compositions and Glass Formulations

Tests were conducted with simulants based on wastes from Hanford tanks AY-102 and A-104. AY-102 was previously planned to provide the commissioning feeds for the WTP. The fully washed AY-102 solids correspond to a waste oxide mass of 332 MT. On an oxide basis, Fe_2O_3 and Al_2O_3 account for > 60 wt% of the waste. The other significant oxides include Na_2O, SiO_2, MnO, and P_2O_5. The soluble fraction in the AY-102 tank waste is primarily a solution of alkali nitrates and nitrites, which, on an oxide basis, is greater than 90% sodium and potassium with the balance consisting of aluminum, phosphate, sulfate, and halides. Tank A-104 was selected from among tanks with the highest concentrations of plutonium (0.12 mg of Pu (all isotopes) per gram of sludge) and appreciable amounts of sludge (oxide mass of the fully washed solids of 76 MT). On an oxide basis, Fe_2O_3 and Al_2O_3 account for > 77 wt% of the waste; other significant oxides include MnO, SiO_2, NiO, Na_2O, and CaO. The soluble fraction in the A-104 tank waste is primarily a solution of alkali hydroxides, nitrites, and nitrates which, on an oxide basis, is greater than 90% sodium with the balance consisting primarily of sulfate, calcium, silicon, and chloride.

In direct-feed scenarios, some of the functions of the WTP ultrafiltration process would be replaced by interim alternatives such as in-tank settling and washing. Since these processes are likely to be less efficient than the WTP ultrafiltration process, the resulting HLW stream would retain larger amounts of the tank supernate and wash water. To evaluate these effects, tests were performed with blends of solids, supernate, and wash water that might be generated from direct-feed processing of wastes from tanks AY-102 and A-104.

For each tank waste, four waste compositions were evaluated in the glass formulation development and melter testing work. These represent various blends of the solids and supernate fractions corresponding to various extents of washing of the solids to remove the soluble fraction. The end-members of this series of compositions are the fully washed solids and the pure supernate. Two intermediate blends are then produced by assuming one or two wash cycles, as described below. These blends are based on the assumptions that the blended tank waste can be settled to achieve a slurry with 15 wt% un-dissolved solids and that each in-tank wash cycle results in a three-fold dilution of the soluble fraction followed by settling to achieve a slurry with 15 wt% un-dissolved solids. Thus, the four waste compositions selected for testing correspond to:

- Blend 1: HLW solids in the supernate produced by dilution of the sludge with water to give 15 wt% un-dissolved solids.

- Blend 2: Blend 1 diluted three-fold with water and settled to 15 wt% un-dissolved solids (i.e., one in-tank wash/settle cycle).
- Blend 3: Blend 2 diluted three-fold with water and settled to 15 wt% un-dissolved solids (i.e., two in-tank wash/settle cycles).
- Solids: Fully washed solids (i.e., washed to the same extent as in the WTP baseline) and settled to 15 wt% un-dissolved solids.

All of the waste blends assume settling to 15 wt% un-dissolved solids, which corresponds to 10.5 wt% and 13.3 wt% HLW oxides for AY-102 and A-104, respectively. The dissolved solids contribute the LAW oxide fraction. The solids content, waste oxide contribution, and chemical composition all change in response to the washing process. The blend representing the unwashed waste (Blend 1) consists of the 15 wt% un-dissolved solids with the remaining 85 wt% being the supernatant solution that is generated by adding water to the sludge. Therefore 61 wt% or 37 wt% of the oxides in the unwashed waste originate from the supernatant for AY-102 and A-104, respectively, resulting in high alkali concentrations similar to LAW streams. The LAW contributions to the solids and oxides, and consequently the alkali content, decrease as the waste is washed; other supernate species also decrease, including nitrate and nitrite, which are the most abundant dissolved volatile constituents in the waste. The fully washed waste is composed of only un-dissolved HLW solids with no LAW solids and therefore has a composition generally similar to baseline WTP HLW streams.

A primary glass formulation objective was to develop and evaluate glass compositions with high waste loadings and processing rates and acceptable durability and processing properties. Glass formulations were developed using an active design strategy, in which characterization data from a set of crucible melts were fed back to design the next set of formulations, in combination with predictions from glass property-composition models. The glasses were characterized with respect to crystallization upon heat treatment, melt viscosity and electrical conductivity, leach resistance on the Product Consistency Test (PCT) and Toxicity Characteristic Leaching Procedure (TCLP), sulfate solubility, melt rate, and K-3 refractory corrosion. The results were used to identify glass formulations, for each of the four blends for each of the two tank wastes (i.e., eight formulations), that have high waste loadings and processing rates and that are fully compliant with the WTP requirements.

DM100 melter tests were conducted with each of these eight formulations. Each of the four AY-102 formulations was processed at two different melt pool bubbling rates (nominal and optimized) and the Blend 4 formulation was also processed at two different feed solids contents (corresponding to a settled solids content of 10 wt% in addition to the 15 wt% value assumed for all other cases), for a total of ten melter runs. Each of the four A-104 formulations was processed at the nominal melt pool bubbling rate and the Blend 1 and 3 formulations were also processed at the optimized bubbling rate, for a total of six melter runs.

Melter feed for each test was produced by combining each waste simulant with the requisite blend of prototypical WTP glass forming chemicals to produce the respective target glass composition. In view of the increased nitrate content for feeds with higher supernate fractions, sugar was added at the ratio of 0.75 moles of carbon per mole of nitrogen oxide present in the waste to control melt pool foaming, as per the WTP LAW baseline. For AY-102, the feed solids content ranged from 0.15 to 0.5 kg glass per kg feed depending on the concentration of HLW solids in the waste, the amount of dissolved solids derived from the LAW supernate, and the amounts and types of glass forming additives that are used. For A-104, the feed solids content ranged only from 0.316 to 0.346 kg glass per kg feed as result of the concentration of HLW solids in the waste being the same for each feed.

RESULTS AND DISCUSSION

Glass Formulations

The selected glass formulations developed for the four AY-102 waste blends spanned a range of waste loadings as the number of wash cycles increases and the contribution of LAW to the overall waste composition decreases, as shown in Fig. 1.

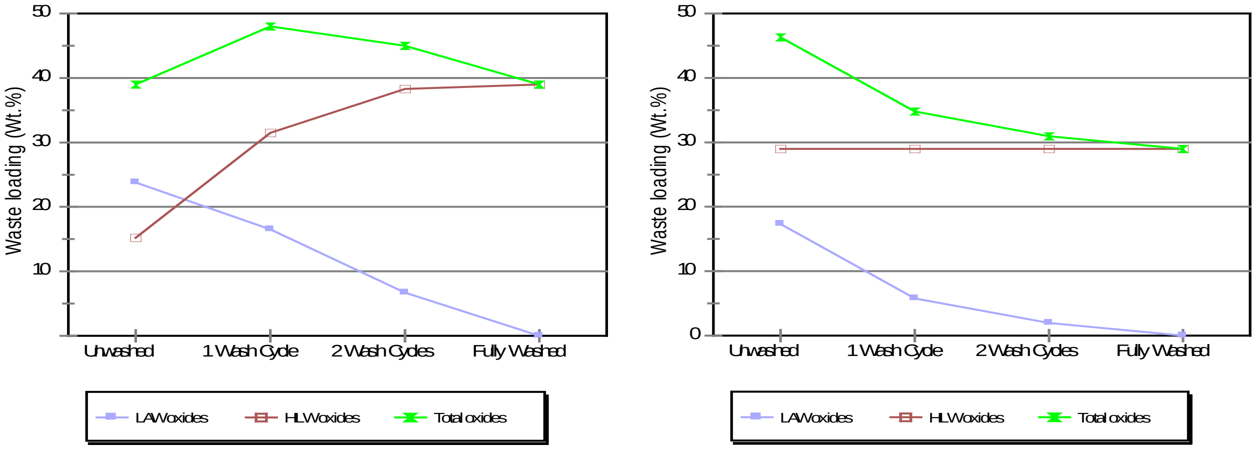

Figure 1. Waste loadings for glasses formulated for AY-102 waste (left) and A-104 waste (right) for various washing scenarios.

Since AY-102 Blend 1 waste with the highest LAW contribution contains high concentrations of alkali oxides, the waste loading was limited by K-3 refractory corrosion. The glass composition selected to treat Blend 1 has a waste loading of 39.0 wt% with LAW contribution of 23.8 wt% and HLW contribution of 15.2 wt%. Blend 2 waste, with a lower LAW contribution, has lower alkali oxide and higher Al_2O_3 concentrations making nepheline formation the waste loading limiting constraint. The glass formulation that was developed for Blend 2 has a waste loading of 48.0 wt% with 16.5 wt% from LAW and 31.5 wt% from HLW. The waste loading for Blend 3 waste and the fully washed HLW solids were limited by spinel crystallization on heat treatment of the glasses. Accordingly, glass formulation efforts were directed at limiting spinel crystallization by adding components such as Na_2O and Li_2O. The glass formulation developed for Blend 3 waste has a waste loading of 45.0 wt% with 6.7 wt% from LAW and 38.3 wt% from HLW. The glass formulation developed to treat the fully washed AY-102 HLW solids has a waste loading of 39.0 wt%, all from HLW. These glasses meet all of the processing and product quality requirements for the WTP. The loading of HLW in the glass increases sharply in going from no wash (Blend 1) to one wash cycle (Blend 2), more moderately as the number of washing cycles is increased from one (Blend 2) to two (Blend 3), and very little in going from two wash cycles to fully washed HLW solids. In terms of HLW waste loading in the glass, there is clearly no advantage in conducting more than two wash cycles for AY-102 because the additional sodium that is removed from the waste is put back as glass former additive in order to limit spinel crystallization in the glass formulation for the fully washed HLW solids.

Similarly, waste loadings for the A-104 blends are also shown in Fig. 1. The glass composition selected to treat A-104 Blend 1 with the highest LAW contribution has a waste loading of 46.32 wt% with an LAW contribution of 17.32 wt% and an HLW contribution of 29.00 wt%. Even for this Blend 1 waste with no washing, spinel crystallization due to the high transition metal content was the waste loading limiting constraint. Thus, the HLW components of the waste (Fe, Mn, Ni, Cr), rather than the LAW components (Na, S, Cl) were waste loading limiting. This is in contrast to the AY-102 waste where LAW components limited waste loading of the unwashed waste. Blend 2 A-104 waste, with lower LAW contribution, has lower Na_2O and higher Fe_2O_3 concentrations but spinel crystallization was still the waste loading limiting factor. This was the case also for the Blend 3 waste and the fully washed HLW solids. Therefore, the same glass formulation could be used to treat the fully washed HLW solids and Blend 2 and Blend 3 wastes with one and two washings, respectively. The only significant difference between the melter feeds for these waste streams was the source of sodium, whether from LAW, as a glass former additive, or a combination of the two. The glass formulation for Blend 2 has a waste loading of 34.78 wt% with 5.77 wt% from LAW and 29.01 wt% from HLW. The glass formulation for Blend 3 waste has a waste loading of 30.95 wt% with 1.93 wt% from LAW and 29.03 wt% from HLW. The glass formulation for the fully washed HLW solids has a waste loading of 29.0 wt%, all from HLW. The above glasses meet all of the processing and product quality requirements for the WTP. The loading of HLW in the glass remains constant, whether the waste is fully washed, subjected to one or two washings, or unwashed. In terms of HLW loading in the glass, there is clearly no advantage in conducting any washing for A-104 because the sodium that is removed from the waste is put back as an alkali glass former additive in order to limit spinel crystallization in the glass formulation.

Melter Tests

The 16 melter tests conducted in this work produced 6000 kg of glass from 8300 kg of feed. Samples of the product glass were collected throughout each test and analyzed for composition. The feeds were processed without difficulties and cold cap conditions while processing feeds containing more HLW than LAW oxides were largely similar to the range of conditions observed in previous tests with HLW feeds. Some shelves of reacting feed formed along the walls of the melter, although not to any rate-limiting extent. Most of these deposits were observed after discharging glass, which lowered the glass level in the melter leaving deposits adhering to the walls out of contact with the molten glass. For the A-104 formulations, the amount of deposits was observed to increase with the proportion of the LAW contribution to the feed. The opposite trend was observed for the AY-102 formulations, which featured a far greater range of water contents, waste loadings, and target glass compositions, as well as far greater contributions of nitrogen oxides and added reductants (sugar) from the LAW supernate.

The glass production rates for AY-102 are shown in Fig. 2 and ranged from 500 kg/m^2/day for dilute fully washed HLW solids to 1250 kg/m^2/day for unwashed waste at nominal bubbling. This increase in glass production rate coincides with an increase in feed solids content from 0.15 to 0.5 kg glass per kg feed (decrease in feed water content from 82 to 39%). Glass production rates increased from 36 to 100% (900 vs. 1225 kg/m^2/day to 1250 vs. 2500 kg/m^2/day) with optimized bubbling. While processing feed containing 82% water, glass production rates increased 55% with optimized bubbling. HLW oxide processing rates ranged from 190 kg/m^2/day for unwashed waste to 297 kg/m^2/day for waste that had undergone two wash cycles at nominal bubbling. HLW oxide processing rates were dependent on the HLW oxide waste loading in the glass as well as the overall glass production rate. LAW oxide processing rates ranged from zero for fully washed waste to 297 kg/m^2/day for unwashed waste at nominal bubbling. LAW oxide processing rates were dependent on the LAW oxide waste loading in the glass as well as the overall glass production rate.

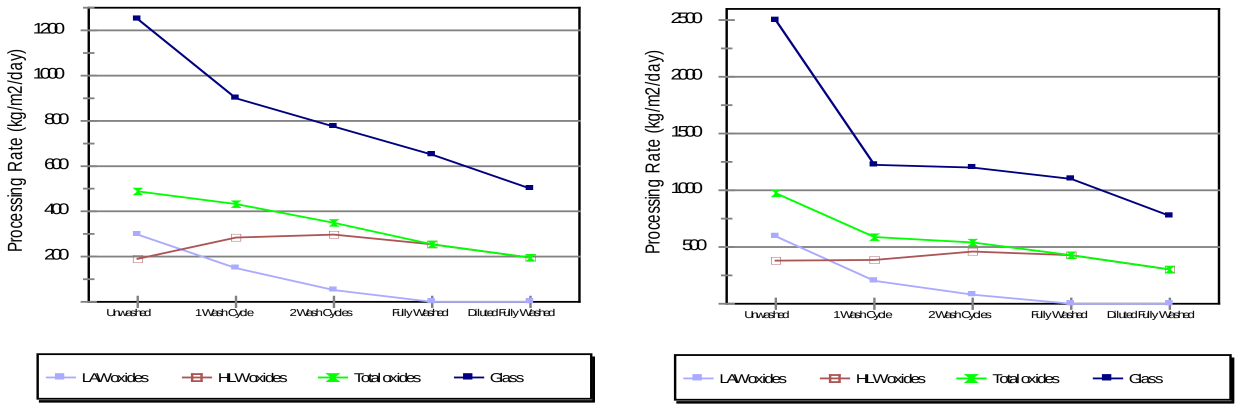

Figure 2. Glass and AY-102 waste processing rates with nominal (left) and optimized (right) bubbling.

Melter exhaust was sampled as each feed composition was processed at the nominal bubbling rate to determine the effect of changing feed composition on particulate and gaseous emissions; the results for total carryover are summarized in Fig. 3.

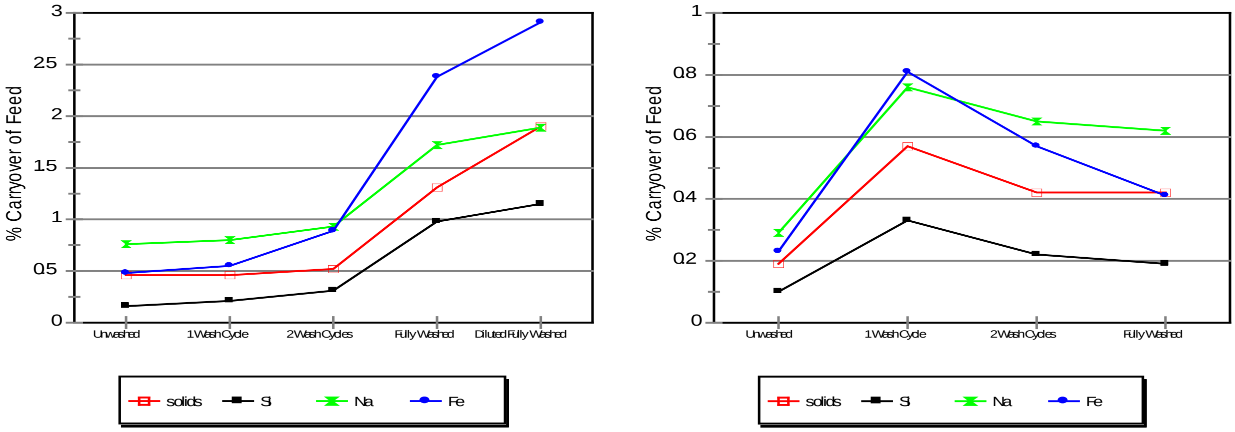

Figure 3. Carryover of feed constituents into the melter off-gas stream for AY-102 feeds (left) and A-104 feeds (right) for various washing scenarios.

Particulate emissions for AY-102 feeds constituted from 0.46 to 1.90 percent of feed solids and increased with the number of wash cycles and feed water content. Solids carryover while processing feed containing fully washed HLW solids at 15 and 10 weight percent solids (71 and 82% water) was 1.3 and 1.9%, respectively, in contrast to 0.46 and 0.54% solids carryover while processing feed containing wastes that have undergone fewer wash cycles and less water. High carryover of solids and iron have been previously observed with high iron, diluted HLW streams, confirming the increased carryover of iron and overall particulate with increasing feed water content in high iron HLW feeds [17]. The level of carryover for the other waste streams tested is within the range of solids carryover observed while processing other HLW and high alkali LAW waste streams containing similar amounts of water. Melter decontamination factors (DFs) were determined for most elements in the feed. The most volatile species were chlorine, fluorine, and sulfur, which is typical. Other elements exhibiting volatile behavior in some of the tests include boron, chromium, potassium, and lead. Gaseous emissions of nitrogen oxides and byproducts of incomplete combustion, such as carbon monoxide and ammonia, ranged from virtually none while processing the fully washed HLW solids to high concentrations of nitrogen oxides (particularly NO) and significant amounts of carbon monoxide and ammonia while processing the unwashed waste. This was expected given the lack of nitrates and organic carbon in the fully washed HLW stream and the high concentration of nitrates in the AY-102 supernate. The extent of the nitrogen oxide emissions was partially mitigated by the addition of sugar to the feed.

As shown in Fig. 3, melter exhaust sampling for A-104 tests showed that particulate emissions constituted from 0.42 to 0.57 percent of feed solids for feeds containing fully and partially washed solids processed with nominal bubbling, consistent with the level of solids carryover measured while processing various feeds containing high iron HLW simulants. Solids carryover while processing unwashed waste solids constituted only 0.19 percent of feed solids while processing with nominal bubbling. Melter DFs showed that the most volatile species were chlorine and sulfur. Other elements exhibiting volatile behavior in some of the tests include boron, chromium, potassium, and lead. As was the case for the AY-102 waste, gaseous emissions of nitrogen oxides and byproducts of incomplete combustion, such as carbon monoxide and ammonia, ranged from virtually none while processing the fully washed HLW solids, to significant concentrations of nitrogen oxides (particularly NO) and carbon monoxide and ammonia while processing the unwashed waste.

The glass production rates for A-104 are shown in Fig. 4 and ranged from 1900 kg/m^2/day for the fully washed HLW solids to 1200 kg/m^2/day for the unwashed waste at nominal bubbling. The feed solids content is relatively constant for the four feeds and is thus not a factor in differences in production rate. Decreases in production rate appear to be related to increases in waste sodium hydroxide at the expense of lithium and sodium carbonate as additives. The slower melt rate for the unwashed waste was consistent with the results from melt rate screening tests. HLW oxide processing rates ranged from 348 kg/m^2/day for unwashed waste to 551 kg/m^2/day for the fully washed waste at nominal bubbling. HLW oxide processing rates were dependent on the overall glass production rate since the HLW oxide loading was the same for all glasses tested. LAW oxide processing rates ranged from zero for fully washed waste to 208 kg/m^2/day for unwashed waste at nominal bubbling. LAW oxide processing rates were dependent on the LAW oxide loading in the glass as well as the overall glass production rate. The total waste oxide processing rate was relatively constant across the four feed compositions tested at around 550 kg/m^2/day.

Also shown in Fig. 4 is a comparison between the glass production rates for the A-104 wastes and the AY-102 wastes. For A-104, glass production rates increased with solids washing and thus decreasing LAW solids loading. The opposite trend was observed with AY-102 wastes due to the increases in HLW oxide loading and consequent increases in water content with washing of the AY-102 wastes. However, the net result is that the HLW oxide processing rate is relatively constant for the various washing scenarios for both A-104 and AY-102.

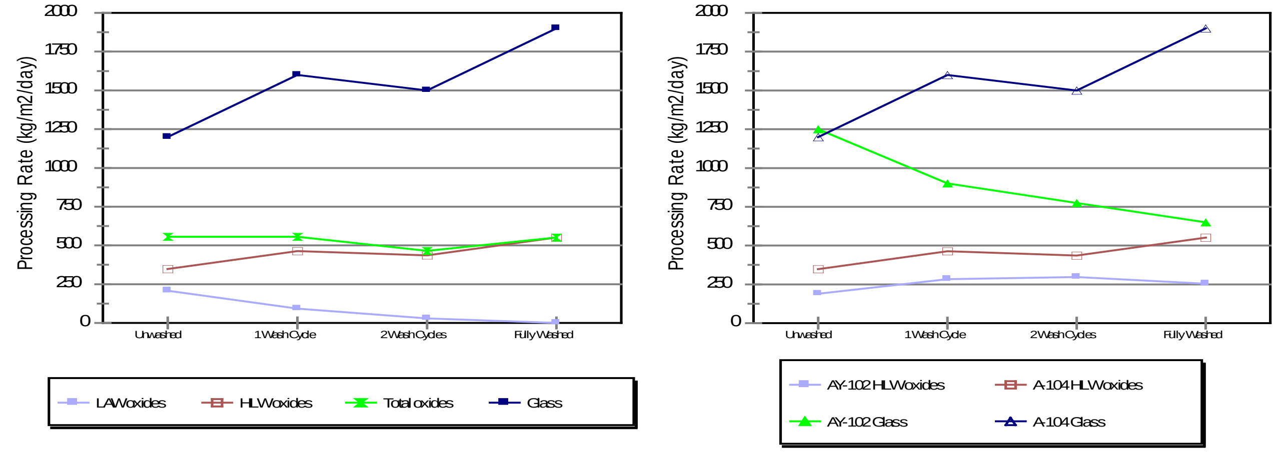

Figure 4. Glass and A-104 waste processing rates (left) and comparison between A-104 and AY-102 rates (right) with nominal bubbling

CONCLUSIONS

The results from the glass formulation and melter testing demonstrate the viability of the HLW direct feed approach and illustrate the relative merits for each waste pretreatment strategy. The amount of time required to vitrify the 332 MT of HLW oxides in Hanford tank AY-102 using a single HLW melter with a surface area of 3.75 m^2 operated at 70% total operating efficiency (TOE) is depicted in Fig. 5. Also shown is the number of HLW canisters produced from AY-102 waste, each assumed to contain 3020 kg of glass.

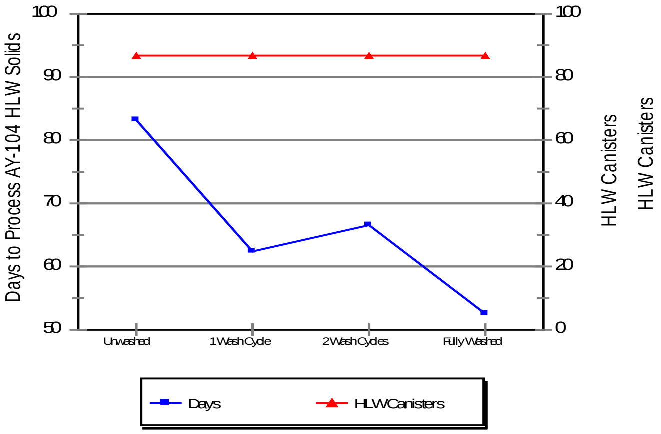

Figure 5. Time (at 70% TOE) and HLW canisters required to process 332 MT of AY-102 HLW oxides (left) and 78 MT of A-104 oxides (right).

Processing AY-102 waste without washing would result in two to three times as many HLW canisters (about 720) than washed waste and would require about 660 days at nominal bubbling conditions to process the HLW contents of tank AY-102. This is primarily attributable to the low HLW waste oxide loading (15.2%) imposed by the high concentration of alkali in the supernate that is not washed from the HLW solids. The addition of a single wash cycle reduces the total canister count by about a factor of two (to about 350) and reduces the number of processing days at nominal conditions to about 450. Adding a second wash cycle prior to vitrification further reduces the required number of canisters to less than 300 and results in the shortest amount of time (about 420 days) required to treat all the HLW solids in the tank. Fully washing the waste results in the fewest number of HLW canisters (about 290) but longer time is required (about 500 days) to vitrify the tank waste due primarily to the increased water content of the feed, which decreases the glass production rate. The fully washed waste also has the added disadvantage of higher solids carryover, also attributable to the high water content of the fully washed feed. The use of bubbling optimization reduces the time required to vitrify the HLW solids by 30 to 50%, to about 300 - 340 days, depending on the extent of washing. Finally, the important effect of the solids content that is achievable by settling is illustrated in the results for tests with the diluted fully washed feed, which corresponds to a settled solids content of 10 wt% instead of the 15 wt% value assumed for all other cases. While this change has no effect on the waste loading, and therefore the number of canisters produced, it results in a significant reduction in glass production rate and an increase in the processing time from about 500 days to about 650 days.

Similarly, the amount of time required to vitrify the 76 MT of HLW oxides in Hanford tank A-104 using a single HLW melter with a surface area of 3.75 m^2 operated at 70% TOE is also depicted in Fig. 5. Also shown is the number of HLW canisters produced from A-104 waste. Only 87 HLW canisters are produced from the fully washed, partially washed, or unwashed waste since the waste loading of HLW oxides is the same for all waste pretreatment strategies tested. This is in contrast to AY-102 for which the HLW oxide loading decreased without washing due to the high concentration of alkali in the supernate, resulting in a more than two-fold increase in HLW canisters produced. Since the HLW loading for each of the formulations for the A-104 waste is the same, the number of days required to process the HLW oxides in each feed is dependent on the glass production rate. Based on the glass production rates obtained from the present tests, the time required to process the A-104 HLW oxides ranged from 53 days for the fully washed waste to 83 days for the unwashed waste.

It should be noted that other factors that could constrain throughput, such as feed preparation, off-gas system constraints, or canister handling limitations, were not considered in this analysis.

The results from this work provide the initial basis for assessments of the relative merits of progressively more intensive pretreatment in HLW direct feed options. Although a simple in-tank settle/decant washing process was assumed in the analysis, similar considerations arise in the evaluation of various possible alternative direct feed interim pretreatment facilities and operations. The principal conclusions for AY-102 are the rapidly diminishing benefits of multiple wash cycles, and, consequently, also of more complex and intensive washing facilities, and the importance of maintaining sufficiently high solids content in the HLW feed to the vitrification facility. Thus, of the pretreatment strategies for direct HLW feed of AY-102 evaluated in this work, the first wash cycle provides the vast majority of the overall benefit of washing in terms of HLW loading and HLW processing time; two wash cycles appears to be optimal in those respects since the second wash cycle provide further, though smaller, gains but that must be weighed against the operational costs of each successive wash cycle. In particular, in the in-tank scenario, settling times to achieve reasonable solids contents can be very long.

Similarly, the principal conclusion for A-104 is the lack of significant benefit of washing this waste as a form of pretreatment. Although the fully washed waste processed faster than the unwashed waste, the time and secondary waste generation associated with washing off-sets this modest benefit in processing duration. Furthermore, there is no benefit in the number of canisters produced since the same HLW oxide loading was achieved for all A-104 waste blending scenarios evaluated. Collectively, these studies demonstrate that the optimum washing strategy, in terms of minimizing the number of HLW canisters, processing duration, and secondary waste generation varies depending on the tank waste being treated.

It should be noted that the AY-102 and A-104 supernates evaluated in these studies are relatively low in sulfate and halides and therefore the primary benefit of washing on waste loading is via removal of sodium. Consequently, excessive washing is counter-productive since sodium is a required additive for HLW vitrification. This is particularly true for the A-104 supernate which has relatively low dissolved solids content. For supernates with high levels of sulfur or halides, more extensive washing may be required [2], particularly in view of the fact that, unlike the WTP LAW melter systems, the WTP HLW melter systems are not designed to tolerate high levels of these species. Together, these results illustrate that the washing strategy will depend on the specific tank waste composition and will need to be evaluated on a tank-by-tank basis.

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