Technology Development to Reduce Mission Life, Life Cycle Costs, and Glass Volumes for US High Level Waste Vitrification Facilities - 11461

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ABSTRACT

Resolution of the nation's High Level Waste (HLW) legacy requires the design, construction and operation of large and technically complex, one-of-a-kind processing facilities coupled to equally complex waste treatment and vitrification facilities. Vitrification technology was chosen to treat HLW at the Hanford site and the Savannah River Site, low-activity waste (LAW) at Hanford site, and may potentially be applied to other defense waste streams such as Idaho National Laboratory (INL) tank waste or calcine material. Joule-heated melters (JHMs) are being used at the Defense Waste Processing Facility (DWPF) and will be used at the Hanford Tank Waste Treatment and Immobilization Plant (WTP) to vitrify tank waste fractions. The loading of waste into glass and the glass production rates at DWPF are limited by either the current melter technology or process control models, which relate glass compositions to various properties. Significant reductions in glass volumes for disposal and mission life are possible with refinement of current property models (or development of new models) and advancements in melter technology and glass formulations. The Savannah River National Laboratory (SRNL) is developing various technologies and strategies to improve both waste loading and melt rate, which ultimately dictate the waste throughput for the DWPF. Specific areas focused upon in this manuscript include development of alternative processing strategies and the assessment of the applicability of current process control models to broader compositional regions as defined by higher waste loadings.

INTRODUCTION

High-level waste (HLW) throughput (i.e., the amount of waste processed per unit time) is a function of two critical parameters: waste loading (WL) and melt rate. For the Waste Treatment and Immobilization Plant (WTP) at the Hanford Site and the Defense Waste Processing Facility (DWPF) at the Savannah River Site (SRS), increasing HLW throughput would significantly reduce the overall mission life cycle costs for the Department of Energy (DOE). Significant reductions in glass volumes for disposal and mission life are possible with advancements in melter technology and glass formulations coupled with refinement of current glass-composition property models (or development of new process control models).

Significant increases in waste throughput were achieved at DWPF for Sludge Batch 3 (SB3) and SB4 through key technical and operational initiatives that included, (i) improving or maximizing facility attainment, (ii) improving the Chemical Processing Cell (CPC) flowsheet [1], (iii) improving critical process control models or implementation approaches [2, 3], and (iv) strategic glass formulation efforts. With respect to strategic glass formulation efforts, frit development has shifted from a global "one frit fits all" concept to a focused effort on optimizing or compositionally tailoring frits for specific sludge batches [1]. In addition, a new liquidus temperature (T_L) model and a revised strategy for approaching the durability limits were developed and implemented [2, 3]. These strategy shifts and model upgrades have allowed DWPF to target higher WLs, while maintaining or improving melt rate, which in turn has been a significant contributor to the improved waste throughputs obtained in the facility.

As a result of these key initiatives, DWPF increased WLs from a nominal 28% (SB2) to approximately 38% WL, while maintaining or improving canister fill times for SB3 and SB4. Although significant improvements in WL and waste throughput were obtained, even higher WLs (> 40%) could have been targeted based on the Product Composition Control System (PCCS) models [4]. More specifically, the models that predict the properties of a glass based on its composition indicated that WLs greater than 40% could have been targeted for these glass systems, while continuing to satisfy both melter processing and product performance constraints. Higher WLs were not targeted during the processing of these previous sludge batches due to experimental [5, 6] and actual facility data demonstrating that melt rate is significantly reduced as higher WLs are targeted, which in turn adversely impacts waste throughput. Therefore, during processing of a specific sludge batch, DWPF typically evaluates melt rate as a function of WL to determine the WL that yields the maximum waste throughput. Optimum waste throughput has historically been demonstrated at a WL significantly lower than the maximum allowed by the current process control models. Narrowing or eliminating this WL gap is of primary interest for continual improvements in the DWPF process.

Revolutionary changes in WL, melt rate, and ultimately waste throughput, are still possible through the development of advanced silicate glasses, implementation of alternative melter technologies, or continued improvement in the process control system that dictates what glass systems can be processed through the melter. For example, in September 2010, DWPF implemented a bubbler technology into the melter to enhance melt rate, which could potentially minimize (if not eliminate) the historical trends between melt rate and WL. If this occurs, there will be a fundamental shift in the technical or process control criteria that will limit DWPF's ability to target higher WLs for future operations. More specifically, the process control models (or the implementation strategy of those models) underpinning the Slurry Mix Evaporator (SME) acceptability process could become the critical limitation, rather than the previous limitation of reduced melt rate as WL is increased [4].

The impact of implementing a new melter technology on glass production rates and the downstream impacts or restrictions to target even higher WLs is demonstrated in Figs. 1 and 2. For previous sludge batches processed at the DWPF, as WL generally increased, melt rate decreased (conceptually shown in Fig. 1). This trend was observed during processing of Frit 418 and SB3, so this system will be used in the following discussion. Also shown in Fig. 1 is the projected operating window (i.e., the WL interval over which the glass is classified as acceptable based on model predictions) for the Frit 418 – SB3 system, which in this example is 25 – 45% WL. That is, if one were to compute the overall glass composition based on the compositions of Frit 418 and SB3 at a WL within this range and compare the predicted properties (T_L, viscosity, durability, etc) of that glass to the current constraints, the glass would be deemed acceptable for melter processing. At 46% WL, model predictions associated with a specific process or product performance constraint would be classified as unacceptable and thus access to WLs higher than 45% would not be allowed.

Savannah River National Laboratory (SRNL) laboratory testing and subsequent radioactive operations at DWPF evaluated melt rate as a function of WL for this system and found a gradual decrease in melt rate with increased WL approximated by curve in Fig. 1 [5, 6, 7]. The maximum waste throughput (the amount of waste process per unit time) was determined to be at approximately 38% WL for the Frit 418 – SB3 system (represented by the "peak" in the blue line of Fig. 1). Although the process control models allowed WLs of up to 45% to be targeted, the considerably negative impact on melt rate at higher WLs resulted in a reduction of the targeted WL to 38% in order to maximize waste throughput. Therefore, during SB3 processing, a seven percentage point WL interval (39 to 45% WL – see shaded area in Fig. 1) was not targeted due to significant reductions in melt rate. Although WLs could have been higher, targeting higher WLs would have lead to a prolonged processing time for the SB3 system and a negative impact on the overall mission life. It is this WL gap (and beyond) that is being targeted through strategic technology development efforts within the Department of Energy's (DOE) Environmental Management Technology Development program.

It should be noted that if one were only concerned with minimizing the number of canisters produced, glasses targeting the highest WL allowed by the process control models would achieve that goal (e.g., for the Frit 418 – SB3 system, WLs of 45% would have met this objective). Based on this strategy and historical melt rate trends, canister fill times would increase, leading to a longer mission life. On the other hand, targeting maximum waste throughput should allow both Tank Farm and DWPF operations to be terminated sooner; however, this latter strategy does not minimize the canister count. This dilemma forces the DOE and/or the operating facility to make business decisions regarding minimizing canister count or reducing mission life – both having significant impacts to the overall life cycle costs.

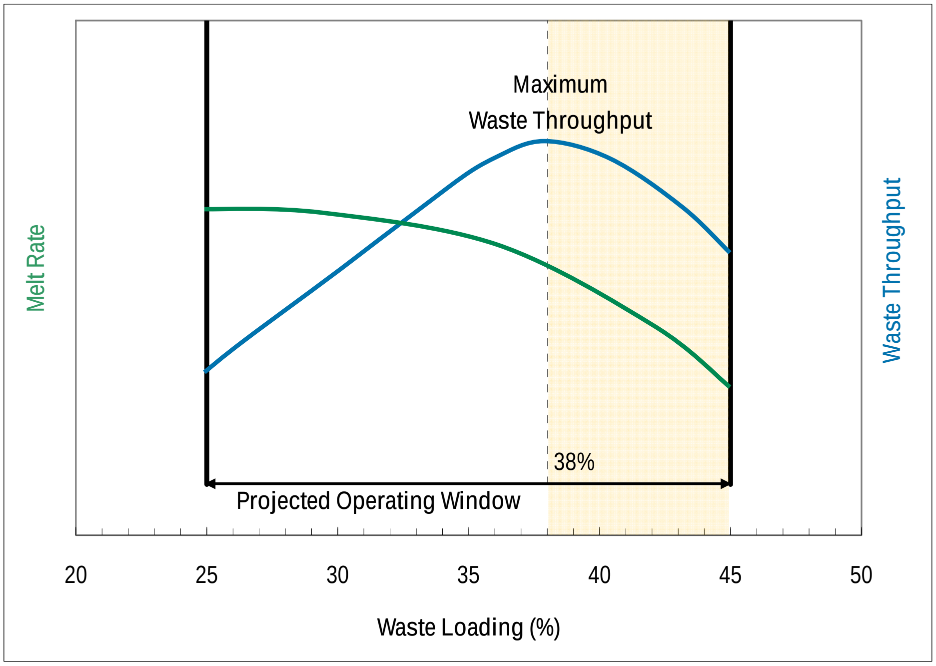

Fig. 1. Historical melt rate and waste throughput as a function of WL trends. The shaded region indicates WLs where acceptable glasses are predicted, but decreased melt rate would hinder waste throughput.

As previously mentioned, DWPF has implemented a bubbler technology to improve convection within the glass melter, which should eventually lead to enhanced melt rates and glass production rates. In terms of strategic planning, the potential for the bubbler technology to eliminate the historical dependence of melt rate on WL (including any impact of feed rheology) must be considered and accounted for. Assuming that waste retrieval and melter feed preparation unit operations do not become the limiting factor of the flowsheet, Fig. 2 represents a potential scenario if the historical dependence of melt rate and WL are eliminated. The horizontal dashed line in Fig. 2 signifies that melt rate is constant as a function of WL. If true and using the Frit 418 – SB3 system strictly an example, DWPF could conceptually (and ideally) target 45% WL for this system to maximize waste throughput. By targeting 45% WL, DWPF would not only be processing the maximum amount of waste per unit time, but would also be minimizing the number of canisters produced under the limitations of the product control models leading to significant reductions in the overall life cycle costs. Although significant improvements in waste throughput could be demonstrated by targeting 45% WL, the driver for targeting even higher WLs would shift from being melt rate or waste throughput limited to restrictions based on the process control models or the criteria implemented for specific glass properties, such as liquidus temperature (T_L), viscosity, durability, or nepheline formation). In this scenario, options to gain further improvement in waste throughput would fall into at least three categories: (a) reducing conservatism or uncertainties in the existing models, (b) developing and implementing new models, and (c) developing new process or product performance criteria or an alternative implementation or control strategy.

Therefore, SRNL has focused on three key initiatives to continue to enhance WL for the DWPF: (1) nepheline formation, (2) model applicability for advanced silicate glasses and (3) developing alternative model implementation strategies. It is this latter initiative that is the focus on this manuscript.

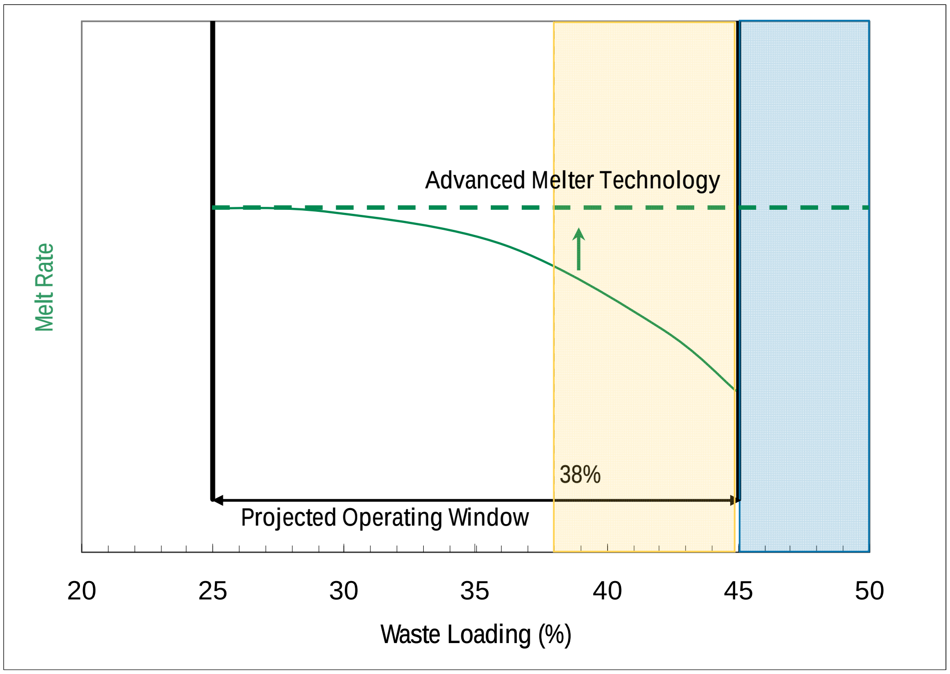

Fig. 2. Potential melt rate improvements as a result of alternative melter technologies.

Possible Impact of T_L Implementation Strategy Change

One of the options being evaluated by SRNL to gain access to higher WLs is the potential to change the implementation strategy of the current T_L model [2]. For example, assume in Fig. 2 that access to higher WLs (≥ 46%) is limited by predictions of T_L relative to the current DWPF acceptance criterion (Measurement Acceptance Region (MAR) limit). In this case, T_L predictions at 46% WL exceed the T_L MAR criterion, which is based on a nominal 1050°C Property Acceptability Region (PAR) value onto which model and measurement uncertainties are applied. Application of the 1050°C PAR value provides a 100°C offset with the nominal melter temperature (1150°C) to prevent massive devitrification within the melt pool. Once uncertainties are applied, the resulting MAR criterion for acceptance may be on the order of 1015°C (assumed for this example). Therefore, if the predicted T_L of the glass at 46% WL is greater than 1015°C, the glass composition would be deemed unacceptable from a process perspective even though the (assumed) predicted T_L for this system is approximately 135°C below the nominal melt pool temperature.

If faced with this situation, options to gain access to higher WLs (i.e., the blue shaded region in Fig 2) would include developing new process or product performance criteria or models or defining a new implementation strategy. One example of an alternative implementation strategy would be to evaluate the potential impacts of reducing the 100°C offset between the nominal melt pool temperature (1150°C) and the 1050°C T_L PAR criterion (without uncertainties added) on the ability to increase the operating windows for T_L-limited glass systems. If significant increases in WL could be gained through the use of a 25°C or 50°C offset (1075 or 1100°C, respectively, instead of the 1050°C currently used), then decisions to implement this approach would need to balance the positive impacts of higher WLs on the overall mission life against the risk of a reduction in conservatism associated with melt pool crystallization. The magnitude of this impact would also be influenced by the WL at which the next product or performance constraint becomes a limiting factor. For example, the Frit 418 – SB3 system is T_L limited at 46% WL, but predictions of either low viscosity (< 20 Poise at 1150°C without uncertainties applied) or predictions of nepheline formation become a limiting factor at 47% WL. Implementation of a smaller T_L offset would have very little benefit in this case.

SRNL completed an evaluation of the potential impacts of implementing a new T_L PAR criterion of 1100°C as compared to the current PAR constraint of 1050°C [8]. The evaluation used the MAR assessment approach developed by Peeler and Edwards [9] in which the current PCCS algorithms (including the 1050°C T_L constraint) were used to assess the projected operating windows of future sludge batches (SB8 through SB17) projected in Revision 15 of the DWPF High Level Waste Systems Plan [10]. Use of the current constraints not only identified specific glass forming systems whose projected operating windows are T_L-limited at higher WLs, but also provided a baseline from which comparisons were made with the use of the 1100°C T_L criterion (keeping all other constraints as currently implemented).

Multiple approaches were used to screen glass forming systems in an effort to determine the potential impact of reducing the T_L offset on future operations. Glass forming systems which provided the maximum increase in upper WL with the implementation of the 1100°C T_L PAR were then identified for each sludge batch. This information is summarized in Table I. It should be noted that there may have been more than one frit that provided the maximum increase for a specific sludge, but only one frit for each sludge batch is shown in Table I. Other feasible systems are not shown to conserve space.

First consider the information associated with SB9 (first row in Table I). These results indicate that with the current 1050°C T_L PAR, the projected operating window (for a specific frit ($8B_2O_3$-$7Li_2O$-$85SiO_2$ wt%)) is 36 to 44% WL). However, implementation of the 1100°C T_L PAR yields a projected operating window of at least (as low as) 38% with an upper WL limit of 52% WL ("Max (%WL) 1100°C T_L"). This is an eight percentage point increase in the operating window and with the shift in the acceptance criterion for the operating window, a significant increase in WL could be attained (e.g., at least (as low as) 38% WL up to 52% WL for the SB9 system).

Table I. Impact of a Shifting "Acceptable" Operating Window and the 1100°C T_L PAR Constraint on Various Glass Forming Systems.

Sludge Batch	Frit Composition (wt%)	1050°C T_L PAR		1100°C T_L PAR	
		Min (%WL)	Max (%WL)	Min (%WL)*	Max (%WL)
SB9	$8B_2O_3$-$7Li_2O$-$85SiO_2$	36	44	38	52
SB10	$8B_2O_3$-$2Fe_2O_3$-$5Li_2O$-$1Na_2O$-$84SiO_2$	38	41	38	52
SB11	$8B_2O_3$-$5Li_2O$-$4Na_2O$-$83SiO_2$	36	44	38	52
SB12	$9B_2O_3$-$9Li_2O$-$82SiO_2$	34	45	38	54
SB13	$8B_2O_3$-$2Fe_2O_3$-$10Li_2O$-$80SiO_2$	28	44	38	53
SB14	$8B_2O_3$-$6Li_2O$-$4Na_2O$-$82SiO_2$	37	46	38	54
SB15	$14B_2O_3$-$2Fe_2O_3$-$4Li_2O$-$1Na_2O$-$79SiO_2$	38	41	38	52

* A lower WL of 38% is shown for all operating windows with the 1100°C T_L PAR. This indicates that the lower WL is at least as low as 38% but could be lower.

Table II provides the detailed MAR assessment results for the SB10 system when coupled with $8B_2O_3$-$2Fe_2O_3$-$5Li_2O$-$1Na_2O$-$84SiO_2$ frit. The first column (%WL) represents the WL interval from 25 to 60%. The second column (MAR w 1050 T_L PAR) summarizes the MAR results with the current 1050°C T_L PAR. The information shown in this column indicates the property (or properties) that fail their respective MAR criteria at each WL. For example, at 25 and 26% WL, predictions of high viscosity (highv) and inadequate Al_2O_3 content fail their respective MAR criteria. At 27%, only predictions of high viscosity fail, which is the case through a WL of 37%. The dashes ("-") from 38% WL to 41% WL indicate that all of the properties for those glasses pass the MAR criteria and are acceptable for DWPF processing. At 42% WL, predictions of T_L exceed the 1050°C constraint (after uncertainties are applied), and thus the system is limited only by T_L predictions up through 52% WL, where low viscosity and T_L become the co-limiting constraints at 53% WL. This glass forming system illustrates the impact of reducing the 100°C offset to 50°C to gain access to higher WLs. Given the WL interval from 42% to 52% is only restricted by T_L

predictions based on the use of the 1050°C PAR, it is not surprising that once the 1050°C T_L constraint is relaxed to 1100°C, the projected operating window size increases to 38 – 52% WL. This is a gain in access to 11 percentage points in WL space for this system, and could allow DWPF to target WLs in the upper 40s or low 50s (assuming applicability of all PCCS models at this higher WL which is discussed below).

Table III shows other examples of the impact that relaxing the T_L PAR criterion has on gaining access to significantly higher WLs for SB9, SB11, and SB12. In these examples, specific frits have been coupled with each nominal sludge batch composition and evaluated using both the 1050°C and 1100°C T_L PAR criteria. Each of the systems is T_L limited when the 1050°C T_L PAR is applied. When the T_L PAR is relaxed, access to higher WLs (from 8 to 11 WL points) occurs for all systems with upper WLs in the low 50s being acceptable. These results demonstrate the potential impact of relaxing the T_L PAR criterion from 1050°C to 1100°C for future sludge batch operations.

As previously mentioned, given the potential for the bubbler technology to eliminate the historical dependence of melt rate on WL (including any impact of feed rheology) and assuming that the tank retrieval and CPC unit operations were not rate limiting, strategic planning is critical to identify the next rate limited factors to gain access to higher WLs. The reduction of the T_L offset is one option that could be considered if access to higher WLs is limited by current T_L model predictions. Although the results of this study have shown significant increases are possible for future sludge batches, prior to implementation of this alternative processing strategy, an assessment of the potential gains in WL space against the reduction in conservatism (depending on the magnitude of the off-set) associated with this process related constraint should be made. In addition, applicability of the current T_L model to higher WL glass compositional regions must be assessed.

Table II. Assessments for SB10 and a Candidate Frit with a 1050 and 1100°C T_L Constraint. ($8B_2O_3$-$2Fe_2O_3$-$5Li_2O$-$1Na_2O$-84 SiO_2, wt%)

% WL	**MAR with 1050°C T_L PAR**	**MAR with 1100°C T_L PAR**
25	highv Al_2O_3	highv Al_2O_3
26	highv Al_2O_3	highv Al_2O_3
27	highv	highv
28	highv	highv
29	highv	highv
30	highv	highv
31	highv	highv
32	highv	highv
33	highv	highv
34	highv	highv
35	highv	highv
36	highv	highv
37	highv	highv
38	-	-
39	-	-
40	-	-
41	-	-
42	T_L	-
43	T_L	-
44	T_L	-
45	T_L	-
46	T_L	-
47	T_L	-
48	T_L	-
49	T_L	-
50	T_L	-
51	T_L	-
52	T_L	-
53	T_L, lowv	T_L, lowv
54	T_L, lowv	T_L, lowv
55	T_L, lowv	T_L, lowv
56	T_L, lowv	T_L, lowv
57	T_L lowv, Neph	T_L lowv, Neph
58	T_L lowv, Neph	T_L lowv, Neph
59	T_L lowv, Neph	T_L lowv, Neph
60	T_L lowv, Neph	T_L lowv, Neph

Table III. Assessments for the SB9, SB11, and SB12 and a Candidate Frit with a 1050 and 1100°C T_L Constraint.

	SB9		**SB11**		**SB12**	
% WL	**1050°C T_L PAR**	**1100°C T_L PAR**	**1050°C T_L PAR**	**1100°C T_L PAR**	**1050°C T_L PAR**	**1100°C T_L PAR**
25	highv	highv	highv	highv	highv	highv
26	highv	highv	highv	highv	highv	highv
27	highv	highv	highv	highv	highv	highv
28	highv	highv	highv	highv	highv	highv
29	highv	highv	highv	highv	highv	highv
30	highv	highv	highv	highv	highv	highv
31	highv	highv	highv	highv	highv	highv
32	highv	highv	highv	highv	highv	highv
33	highv	highv	highv	highv	highv	highv
34	highv	highv	highv	highv	-	-
35	highv	highv	highv	highv	-	-
36	-	-	-	-	-	-
37	-	-	-	-	-	-
38	-	-	-	-	-	-
39	-	-	-	-	-	-
40	-	-	-	-	-	-
41	-	-	-	-	-	-
42	-	-	-	-	-	-
43	-	-	-	-	-	-
44	-	-	-	-	-	-
45	T_L	-	T_L	-	-	-
46	T_L	-	T_L	-	T_L	-
47	T_L	-	T_L	-	T_L	-
48	T_L	-	T_L	-	T_L	-
49	T_L	-	T_L	-	T_L	-
50	T_L	-	T_L	-	T_L	-
51	T_L	-	T_L	-	T_L	-
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54	T_L, lowv	T_L lowv	T_L, lowv	T_L, lowv	T_L	-
55	T_L, lowv	T_L lowv	T_L, lowv	T_L, lowv	T_L Neph	T_L Neph
56	T_L lowv	T_L lowv	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph
57	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph
58	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph
59	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph
60	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph	T_L lowv Neph

Impact of Higher WLs on Current Process Control Models

With the development of advanced glass formulations, implementation of alternative melter technologies, and/or changes in the implementation strategy of current models, revolutionary improvements in WL, melt rate, and waste throughput are expected. Anticipating a significant transition in WL, assessments of the applicability of the current models to broader compositional regions are needed. More specifically, glass composition-property prediction models are currently utilized during the DWPF SME acceptability process to demonstrate that these constraints are successfully and confidently being met for each process batch. The models that provide structure to this feed forward process control strategy were developed and validated over specific compositional regions. While the reliable performance of these models to date is immediately apparent in the successful operation of DWPF, future operations targeting higher WLs may result in significant shifts to glass compositional regions that have not been previously evaluated. With this in mind, questions associated with reliable performance of the models in these broader compositional regions surface. In order to answer these questions, data are needed in these broader compositional regions to ensure that the current models are applicable or that adequate data are available to refine the models or develop alternative models. Preemptive assessments of the process control models in the projected compositional regions of interest must be performed in order to ensure access to higher WL regions which may be required to meet contractual agreements. This assessment is currently being performed through DOE's Environmental Management (EM) program through a joint effort at SRNL and Pacific Northwest National Laboratory (PNNL) and will be the subject of a subsequent manuscript.

SUMMARY

High-level waste throughput (i.e., the amount of waste processed per unit time) is a function of two critical parameters: WL and melt rate. For the WTP at the Hanford Site and the DWPF) at the Savannah River Site, increasing HLW throughput would significantly reduce the overall mission life cycle costs for the DOE. Significant reductions in glass volumes for disposal and mission life are possible with advancements in melter technology and glass formulations coupled with refinement of current glass-composition property models (or development of new process control models). Although significant increases in waste throughput have been achieved at DWPF for previous sludge batches, recent implementation of a new melter technology (i.e., bubblers) should enhance waste throughput even further. In fact, this new technology (coupled with other process enhancements) could transition or shift restrictions in waste throughput from melt rate limited to limitations associated with the current process control models and/or how they are implementing. Options to gain further improvement in waste throughput would fall into at least three categories: (a) reducing conservatism or uncertainties in the existing models, (b) developing and implementing new models, and (c) developing new process or product performance criteria or an alternative implementation or control strategy.

Assessments have been made on the impact of redefining how the current T_L model is implemented on gaining access to higher WL glasses. The results of that study indicate that a reduction of the T_L offset (i.e., the 100°C "safety" factor between the nominal melt pool temperature and the 1050°C T_L model PAR constraint) can provide a significant increase in WL for systems that are T_L-limited. In fact, for certain systems, a 10 point increase in the upper WL is potentially achievable with a 50°C offset. Although the results of this study have shown significant increases are possible for future sludge batches, prior to implementation of this alternative processing strategy, an assessment of the potential gains in WL space against the reduction in conservatism (depending on the magnitude of the off-set) associated with this process related constraint should be made. In addition, applicability of the current T_L model to higher WL glass compositional regions must be assessed.

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