## Performance Enhancements to the Hanford Waste Treatment and Immobilization Plant Low-Activity Waste Vitrification System

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## ABSTRACT

The U.S Department of Energy Office of River Protection (DOE-ORP) is constructing a Waste Treatment and Immobilization Plant (WTP) for the treatment and vitrification of underground tank wastes stored at the Hanford Site in Washington State. The WTP comprises four major facilities: a pretreatment facility to separate the tank waste into high level waste (HLW) and low-activity waste (LAW) process streams, a HLW vitrification facility to immobilize the HLW fraction; a LAW vitrification facility to immobilize the LAW fraction, and an analytical laboratory to support the operations of all four treatment facilities.

DOE has established strategic objectives to optimize the performance of the WTP facilities and the LAW and HLW waste forms to reduce the overall schedule and cost for treatment and vitrification of the Hanford tank wastes. This strategy has been implemented by establishing performance expectations in the WTP contract for the facilities and waste forms. In addition, DOE, as owner-operator of the WTP facilities, continues to evaluate 1) the design, to determine the potential for performance above the requirements specified in the WTP contract; and 2) improvements in production of the LAW and HLW waste forms. This paper reports recent progress directed at improving production of the LAW waste form.

DOE's initial assessment, which is based on the work reported in this paper, is that the treatment rate of the WTP LAW vitrification facility can be increased by a factor of 2 to 4 with a combination of revised glass formulations, modest increases in melter glass operating temperatures, and a second-generation

LAW melter with a larger surface area. Implementing these improvements in the LAW waste immobilization capability can benefit the LAW treatment mission by reducing the cost of waste treatment.

# INTRODUCTION

The WTP contract [1] requires that the LAW vitrification facility be designed to include two LAW melter lines to convert pretreated LAW waste to a glass waste form. Each melter line was specified to produce a name-plate design capacity of 15 metric tons of glass per day (MTG/d). The LAW melters are designed with a surface area of 10 m<sup>2</sup> and a glass production rate of 1.5 MTG/m<sup>2</sup>/d.

The waste feeds to the LAW melters comprise primarily sodium and aluminum, nitrates, nitrites, sulfates, and hydroxides. Radioactive materials, primarily Cs-137, Sr-90, plutonium, and americium, are removed from the LAW waste stream by processing in the pretreatment facility so that the immobilized LAW glass produced is a nontransuranic, low-radiation dose waste form.

The WTP contract [1] divides the LAW waste feeds into composition envelopes denoted as Envelopes A, B, and C based on their chemical and radiochemical compositions. DOE-specified minimum waste loading requirements in the LAW glass, based on Na<sub>2</sub>O concentrations in the waste envelopes, were 14, 3, and 10 wt% Na<sub>2</sub>O for Envelopes A, B, and C, respectively. These waste loading requirements were derived from glass-testing information available at the time of the WTP contract award, and they account for sodium that would need to be added to the WTP process to support waste treatment operations.

DOE-ORP, as owner-operator of the WTP, which is currently under construction, is evaluating options to improve the waste treatment rate. One area of evaluation is possible enhancement of the treatment capability of the LAW vitrification facility. This is being done to reduce the life-cycle cost and schedule for LAW treatment. It will also provide a basis for establishing performance expectations for the future WTP operating contractor.

DOE's initial assessments indicated that the LAW vitification facility can support a higher glass production rate [2] and the waste loading of the LAW waste form can be increased [3]. These assessments of the LAW vitrification system design indicate that the most effective, lowest-risk alternatives for improving the performance of the LAW vitrification plant are:

- Increasing the LAW glass waste loading
- Increasing the LAW melter glass production rate by operating the LAW vitrification process at higher temperatures
- Increasing the LAW melter glass production rate by increasing the LAW melter glass-pool surface area within the existing melter space external envelope.

The following sections summarize recent results of the continued assessments.

## TARGETED IMPROVEMENTS TO INCREASE LAW VITRIFICATION CAPABILITY

### LAW Glass Waste Loading Improvements

Extensive glass formulation development work sponsored by the WTP contractor has led to the selection of a range of working "baseline" glass formulations for the anticipated LAW streams. These glass formulations have been developed to support melter test programs over a range of scales up to one-third-scale nonradioactive testing. The results from these tests have been used to successfully support design confirmation and flow-sheet analyses for the LAW vitification facility. However, despite the maturity of

these LAW baseline formulations, the extent of optimization that was possible was limited by the aggressive schedule and limited project budget for completion of the LAW vitrification facility design.

DOE's review of the LAW system indicated that a technical basis existed for improving the LAW loadings over the baseline solution [3]. This was based primarily on work completed by other DOE-sponsored programs and on evaluation of the margins that existed in the waste form performance of the WTP baseline glasses. These studies showed that sodium and sulfate are the primary influences on LAW glass waste loading. Because of the importance of waste loading in determining overall waste treatment costs, DOE supported additional glass development testing by Duratek, Inc. and the Vitreous State Laboratory of The Catholic University of America to evaluate alternative glass formulations.

The strategy prescribed by DOE in this alternative glass development testing program was to clearly establish acceptable composition boundaries for LAW glass waste forms by testing glasses that have a high confidence for acceptability and glasses anticipated to be unacceptable. In this effort it was expected that a high fraction (> 30 to 40%) of the tested glasses would fail based on durability requirements or processing requirements, thereby establishing a better understanding of true glass capability. This compares with the traditional waste glass development approach (see Fig. 1) in which the composition boundaries of acceptable glasses are conservatively expanded to establish the acceptable composition region. The alternative waste glass development strategy results in earlier establishment of the acceptable glass composition region, which reduces both glass development and future operating costs. In the alternative glass development work DOE specified minimum LAW glass waste loadings based on:

- WTP target waste compositions that had been previously qualified and tested, and
- Minimum waste loading requirements for Na<sub>2</sub>O and SO<sub>3</sub>, the two primary chemical constituents that limit LAW glass waste loadings.

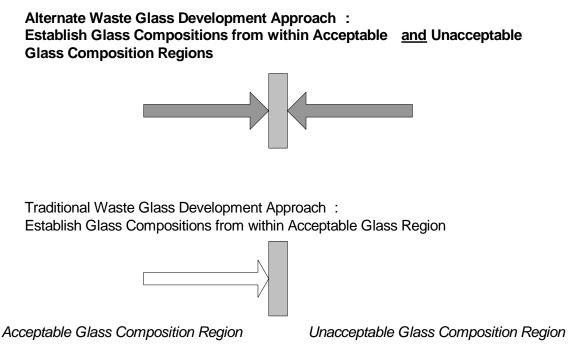


Fig. 1. Comparison of traditional and alternative waste glass development approaches

Figure 2 shows the relationship in terms of Na<sub>2</sub>O and SO<sub>3</sub> compositions in the LAW glasses for the WTP baseline glasses tested (LAW A44, LAW B96, and LAW C35), the WTP baseline glass correlation, the DOE LAW glass model, the revised DOE glass model, and recent glasses tested (LAW A187, LAW B99, and LAW C100). The WTP glass model was based on the WTP contractor's design solution to the requirements specified by DOE in the WTP contract. The model was based on DOE's review of the capability of LAW glasses. The revised DOE model is based on recent experimental work and represents a new LAW waste loading performance target. The transition between these representations of LAW glass loading represents DOE's changing expectations for LAW glass waste loadings.

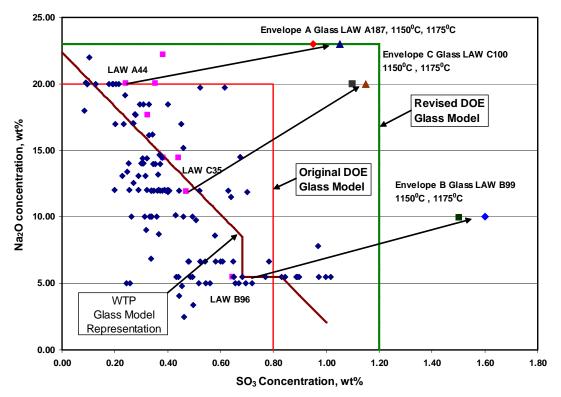


Fig. 2. Comparison of LAW glass models for the WTP and enhanced glass compositions

The recent glass formulation development and melter testing was conducted to improve waste loadings in prototypic LAW waste feeds denoted as Envelopes A, B, and C waste streams. The WTP baseline and revised LAW glass compositions are summarized in Table I. The results from the recent LAW glass testing program have been presented in several reports [4-7].

Glass development and testing work entailed several types of crucible-scale tests to identify glass compositions that lead to increased sulfate solubility and sulfate incorporation rates, screening tests on a small scale melter (denoted as the DM-10) and confirmation testing in a larger-scale melter (denoted as the DM-100) capable of glass production rates of 200 to 400 kg/day.

Glass formulation development was based on specific waste compositions in the Hanford tanks. LAW Envelopes A, B, and C were based on the composition of wastes from tanks AN-105, AZ-102, and AN-102, respectively. The objective of the glass development work was to develop glass formulations containing greater than 23 wt% Na<sub>2</sub>O for Envelope A, greater than 10 wt% Na<sub>2</sub>O for Envelope B, and greater than 20 wt% Na<sub>2</sub>O for Envelope C, with increased concentrations of sulfate having target minimum values of 1.2 wt% SO<sub>3</sub>.

	Envelope A Gla	SS	Envelope B Glas	38	Envelope C Glas	SS
Component	WTP Baseline Glass (LAW A44)	Revised Glass Formulation LAWA187 (for AN-105)	WTP Baseline Glass (LAW B96)	Revised Glass Formulation LAWB99 (for AZ-102)	WTP Baseline Glass (LAW C35)	Revised Glass Formulation LAWC100 (for AN-102)
$Al_2O_3$	6.10	10.57	6.16	10.10	6.07	10.16
$B_2O_3$	8.84	12.77	10.01	10.95	9.42	13.68
CaO	1.96	6.47	6.76	10.15	7.35	8.02
$Cr_2O_3$	0.02	0.52	0.03	0.11	0.01	0.02
Cs <sub>2</sub> O	0.15	-	0.15	-	0.15	-
Fe <sub>2</sub> O <sub>3</sub>	6.87	0.90	5.28	1.14	3.60	1.00
K <sub>2</sub> O	0.44	0.51	0.12	0.41	0.09	0.15
Li <sub>2</sub> O	-	-	4.29	3.53	3.25	-
MgO	1.96	0.90	2.97	1.14	1.49	1.00
Na <sub>2</sub> O	20.66	23.00	5.47	10.00	11.97	20.00
NiO	-	-	-	-	-	0.03
PbO	-	-	-	-	0.01	0.01
SiO <sub>2</sub>	43.82	34.80	48.66	42.84	47.24	36.62
SnO <sub>2</sub>	-	1.00	-	42.90	-	-
TiO <sub>2</sub>	1.96	-	1.39	-	1.08	-
$V_2O_5$	-	0.97	-	1.23	-	1.00
ZnO	2.92	2.99	4.85	3.52	3.99	3.00
ZrO <sub>2</sub>	2.94	2.99	3.17	3.52	3.00	3.00
Cl	1.17	0.64	0.01	0.01	0.39	0.65
F	-	0.00	0.02	0.07	0.11	0.19
$P_2O_5$	-	0.00	0.01	0.03	0.16	0.27
SO <sub>3</sub>	0.19	0.95	0.65	1.20	0.63	1.20
SUM	100.00	100.00	100.00	100.00	100.00	100.00

Table I. Oxide Compositions of Target Glasses Evaluated in LAW Waste Loading Enhancement Studies

Glass development included crucible melts prepared with varying concentrations of additives such as CaO,  $Li_2O$ ,  $Al_2O_3$ ,  $B_2O_3$ , and  $SiO_2$ .  $V_2O_5$  was included as an additive to increase sulfate incorporation rates.  $SnO_2$  was added to improve the Vapor Hydration Test performance results of the glass based on previous test results.  $Cr_2O_3$  was added to reduce refractory corrosion with the high-alkali compositions. The crucible melts were also tested for processing characteristics (viscosity, electrical conductivity, crystallization, phase separation), and product quality requirements (Product Consistency Test, Vapor Hydration Test). Based on these tests, a glass composition was selected for each waste envelope for melter testing.

Sulfate solubilities in the glass melts were determined by batch solubility tests in which the glass samples were remelted with excess sulfate; the glass product was then analyzed for sulfate content after removing any separate sulfate phase. The kinetics of sulfate incorporation as well as sulfate solubility was determined by a second method that involved bubbling a mixture of  $SO_2/O_2$  through the glass melt.

Several tests were conducted on a small-scale DuraMelter<sup>TM</sup> (DM-10) system to select glass compositions to be tested with the larger DM-100 melter. Each test segment was conducted with a unique combination of waste feed simulant, sulfur feed content, and glass pool temperature (1150° and 1175°C). Bubbling of the glass melt pool was adjusted to achieve the target glass production rate, 2.25 MTG/m<sup>2</sup>/d. Evaluation of the melter glass pool and discharge samples provided a clear indication of the tolerance of the glass formulations to sulfur at both glass pool temperatures. Analysis was performed on samples of the glass product taken throughout the tests as well as on the melter exhaust to verify composition and to provide data for elemental mass balance determination. The bubbling rate from the steady-state portion of the test segment conducted at a glass temperature of 1150°C was held constant in the following test segment conducted at 1175°C to determine the effect of glass temperature on production rate.

Table II provides results of DM-100 melter testing with the modified glass formulation. Compared in the table are the expected melter glass processing rate,  $Na_2O$  and  $SO_3$  waste loadings, and the sodium immobilization rate at the LAW vitrification facility for the WTP baseline glass and the modified LAW glass compositions. These results show a 15 to 130% increase in waste loading and a 22% increase in the glass melting rate based on vitrification of the waste feeds at 1175°C, which, combined, could provide a 40 to 180% increase in overall treatment rate.

# LAW Melter Temperature Changes Improve Production Rate

The WTP LAW melter was designed to operate with a nominal melt pool temperature of 1150°C. It is generally known that waste processing rates increase with higher melter operating temperatures. Hanford LAW feeds were also expected to process faster at higher temperatures, but the extent of this effect for relatively modest temperature increases (within the operational limits of the materials of construction) was unknown. Furthermore, the solubility of sulfate, its rate of incorporation, and its partitioning between the glass and the off-gas stream are all strongly temperature dependent. The effects generally favor increased sulfate tolerance with increasing temperature.

The effect of slightly higher melter operating temperatures  $(1175^{\circ} \text{ to } 1225^{\circ}\text{C})$  on the processing rate was investigated using the DM-3300 LAW pilot melter [8]. The DM-3300 was designed as a one-third model of the full-scale design for the WTP LAW melter. It was designed with a melt pool surface area of  $3.3 \text{ m}^2$  compared with 10 m<sup>2</sup> for the full-scale WTP melter. The pilot melter design simulated the LAW melter's key dimensions between opposing electrodes while maintaining LAW melter design concepts and process parameters. The LAW pilot melter operated with a molten glass pool that was electrically heated to  $1150^{\circ}\text{C}$  using submerged metallic electrodes. The glass was mixed with a patented Duratek, Inc. proprietary agitation system designed to increase the waste processing rate.

Prior to the shutdown of the pilot melter, five batches of 8-molar sodium feed were processed to determine the effect of increased glass temperatures on glass production rates. The objectives of this test were as follows:

- Collect data to determine the effects of increased temperature (1175°, 1200°, and 1225°C) on melter throughput.
- Observe the glass surface to ensure that processing at elevated temperatures does not result in formation of a deleterious separate sulfate layer in the melter.
- Collect and analyze discharged glass to determine sulfur retention in the glass.

During this testing, all melter parameters (e.g., bubbler configuration, plenum temperature, no lid heaters) were held constant except for the glass pool temperature. The first batch processed replaced the glass in the melter pool with the desired composition. Each of the next four batches was used to determine the glass production rate at various temperatures (1150°, 1175°, 1200°, and 1225°C). Each batch lasted approximately 3 to 4 days, produced approximately 20,000 kg of glass, and was sufficient to turn over the melter inventory more than three times. Melter feed rates were adjusted to maintain a plenum temperature of  $400 \pm 50^{\circ}$ C. The bubbling rates were held constant at nominal rates for the test period. Glass production rates were determined by direct glass weight measurements using a calibrated scale. By using the total time the melter was fed and the total glass produced, a normalized average glass production rate was determined for each batch.

	WTP Baseline LAV	V Glass			Modified LAW Gla	ss Composition		
Envelope	Processing Rate	Concentration of	Concentration of	Na Treatment	Processing Rate	Concentration of	Concentration	Na Treatment Rate at
Glass Composition	Expected in WTP	Na <sub>2</sub> O	$SO_3$	Rate at 100%	Expected in	Na <sub>2</sub> O	of SO <sub>3</sub>	100% Availability
_	Melter <sup>a</sup>	(wt%)	(wt%)	Availability	WTP Melter <sup>b</sup>	(wt%)	<sub>(</sub> wt%)	2 Melters
	(MTG/d)			(MT Na/yr)	(MTG/d)			(MT Na/yr)
Envelope A	21.2/melter	20.02	0.24	3098	21.2 @ 1150°C	23.0	0.95 @ 1150°C	3559 @ 1150°C
Subenvelope A1								
-					25.9 @ 1175°C		1.05 @ 1175°C	4349 @ 1175℃
Envelope B,	18.8/melter	5.47	0.65	402	18.8 @ 1150°C	10.0	1.5 @ 1150°C	947 @ 1150°C
Subenvelope B2		(3 wt% waste Na				(6.9 wt% waste		
•		in glass)			22.9 @ 1175°C	Na in glass)	1.6 @ 1175°C	1153 @ 1175°C
Envelope C	21.0/melter	11.97	0.63	1835	21.0 @ 1150°C	20.0	1.1 @ 1150°C	3066 @ 1150°C
Subenvelope C2								
-					25.6 @ 1175°C <sup>°</sup>		1.15 @ 1175°C	3738 @ 1175°C

Table II. Comparison of Estimated WTP Baseline and Modified LAW Glass Composition Performance

<sup>a</sup> Processing rate based on LAW pilot melter processing data from [9].

<sup>b</sup> The 1175°C processing data on DM100 did not reach steady state; therefore, the processing rate is based on an average rate increase from Envelopes A and B (~22% increase in rate from 1150° to 1175°C)

<sup>c</sup> The 1150°C rates are based on the LAW Pilot Melter processing rates. The 1175°C rates are based on the percent increase in processing rates seen on the DM100 and applied to the 1150°C processing rates.

In addition to glass production rate information, the melter was tested after each batch was complete to determine whether a separate sulfate phase had accumulated on the glass melt surface. The formation of a separate sulfate phase within the melter was determined by dip sampling of the glass pool immediately after the cold cap had dissipated.

The LAW waste composition chosen for this test was Subenvelope A1, the composition used for the Envelope A glass development discussed above. The feed composition was based on the WTP baseline Subenvelope A1 glass formulation modified slightly for the high-temperature operating conditions (Subenvelope A1 formulation LAWA140). Table III presents the composition of the melter feed. After the feed was prepared but before it was fed to the melter, sugar was added as a reducing agent to manage foaming within the melter. Sufficient reducing agent was added to the LAW feed to provide a stoichiometric ratio of 0.5 carbon with respect to feed nitrates and nitrites.

Oxide	Weight %	Oxide	Weight %
$Al_2O_3$	6.20%	SiO <sub>2</sub>	48.02%
$B_2O_3$	8.98%	TiO <sub>2</sub>	1.49%
CaO	1.99%	ZnO	2.97%
$Cr_2O_3$	0.02%	$ZrO_2$	2.98%
$Fe_2O_3$	4.48%	Cl	0.56%
K <sub>2</sub> O	0.44%	F	0.02%
MgO	1.49%	$P_2O_5$	0.07%
Na <sub>2</sub> O	20.04%	$SO_3$	0.24%
NiO	0.003%	Sum	100%

Table III.	LAWA140 Glass Composition
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The results from this test revealed that the LAW melter could safely produce glass at rates of at least  $3,390 \text{ kg/m}^2$ -day without difficulty by increasing glass temperature to  $1225^{\circ}$ C. The testing also revealed that the sulfate retention in the glass would decrease from 90 to 60% when the glass pool temperature was increased from  $1150^{\circ}$  to  $1225^{\circ}$ C. A summary of the production rate information is provided in Table IV. The DM-3300 tests were performed prior to the glass formulation work to optimize the waste loadings described earlier. However, because essentially the same melt rate increases were observed in DM-100 tests with the LAWA140 formulation, the DM-3300 results provide confidence that the DM-100 rates on the new formulations would also be realized at larger scales.

Table IV. Summary of Glass Production Rate Results from High-Temperature Testing
on the LAW Pilot Melter

Glass Pool Melter Temperature	Glass Produced	Glass Production Rate
(°C)	(MT)	$(MTG/m^2-day)$
1150	19.3	1.94 <u>+</u> 0.17
1175	20.8	2.20 <u>+</u> 0.14
1200	19.2	2.58 <u>+</u> 0.26
1225	21.3	3.01 to 3.39 <u>+</u> 0.59

### **Increased LAW Glass Melter Surface Area**

Knowledge of the rate at which a glass furnace produces glass is required during melter design. This parameter is normally expressed in terms of mass of glass produced per unit area of glass pool surface per unit of time. For the WTP waste glass melters, the production rate is expressed in metric tons of glass produced per square meter of glass surface area per day (i.e., MTG/m<sup>2</sup>-day). The specific glass production rate used in the design of the LAW melter was measured during testing with the LAW pilot

melter (DM-3300). The tests revealed that the specific glass production rate was at least 1.5  $MTG/m^2$ -day. Because the LAW melter was specified to produce glass at a rate of 15 MTG/day, it was designed to have a glass pool surface area of 10 m<sup>2</sup>.

Assuming the same specific production rate, the glass production rate of the WTP melter can only be increased by providing the largest glass surface area that can be reasonably obtained. Evaluation of the WTP LAW melter design indicated that the glass pool surface area could be increased by three melter design changes:

- Removing the backup 5-inch K-3 refractory layer and using the resulting space
- Using the surplus space between the glass-pool refractory cooling panels and the gas-barrier
- Using the surplus space between the gas barrier and the shield wall.

A 5-inch layer of K-3 refractory was provided within the WTP LAW melter behind the primary 12-inch glass contact layer as a backup. This layer was included because it had been in previous melter designs to address concerns of potentially high glass corrosion rates. However, extensive laboratory and field corrosion testing have been performed that have ruled out the possibility of such high corrosion rates. Therefore, the 5-inch backup K-3 layer could be removed from the second generation LAW melter.

The WTP LAW melter design also includes some surplus space in the area between the outside surface of movable refractory cooling panels and the inside surface, a gas barrier used to provide confinement for the melter. This space was an artifact of the nearly 5-year melter design evolution. Using this surplus area will allow the refractory sidewalls to be expanded outward and will increase the glass surface area.

The LAW melter also contained surplus area between the outside surface of the gas barrier and the structure supporting the inside of the shield walls for the melter shielded enclosure. The melter glass pool can be further expanded by moving the refractory sidewalls, cooling panels, and gas barrier outward to fill this unused space.

The glass pool dimensions and total surface area of the WTP LAW melter and the proposed secondgeneration LAW melter resulting from these changes is provided in Table V. These changes result in a 47% increase in glass pool surface area with a corresponding increase in the melter waste processing rate.

Dimension Comparison		
Parameter	WTP LAW Melter	Proposed Second-Generation
1 arameter	WII LAW Metter	LAW Melter
Depth (inch/cm)	30/76	35/89
Length (inch/cm)	194/493	232/590
Width (inch/cm)	80/203	99/250
Surface Area (m <sup>2</sup> )	10	14.7

Table V. WTP Baseline and Potential Second-Generation LAW Melter Glass Pool Dimension Comparison

## SUMMARY

The cumulative benefits of the three alternatives considered in this paper for improving the design capacity of the LAW vitrification plant are summarized in Table VI. These benefits are compared with the projected WTP baseline sodium vitification rates for Envelopes A, B, and C of 3,098, 402, and 1,835 MT Na/yr, respectively. This comparison in increased production rate is based on 100% availability of the LAW vitrification facility sodium waste loading and assuming the experimental data and specific LAW compositions identified earlier.

	Alternative Meth			
	Melter Temperature			Total Potential
Waste	Waste Loading	Increase	Melter Surface	Increase,
Envelope	Increase	1150° to 1175°C	Area Increase	(Percent/Factor)
А	15%	22%	47%	110%/2.1
В	130%	22%	47%	310%/4.1
С	67%	22%	47%	200%/3.0

Table VI. Cumulative Benefits of Alternative Methods to Increase LAW Production Rate	Table VI.
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This assessment shows that improvements in waste loading range from 15 to 130% based on the composition. A significant increase in waste treatment rate ( $\sim$ 22%) can be obtained by a modest increase in glass pool operating temperature. Increasing the melter surface area, as could occur in a second-generation LAW melter, also has a significant effect (47%) on waste treatment rate.

The majority of the Hanford LAW has a composition similar to the Envelope A waste described in this paper. However, some of these streams are sodium limited while others are sulfate limited. For sodium-limited Envelope A streams, a doubling of the waste treatment rate is projected based on the suggested LAW system modifications. However, this estimate is conservative because the sulfate capacity of the new Envelope A formulation is also significantly more than that of the baseline (nearly a factor of 3). Consequently, the fraction of Envelope A inventory that will be sodium (rather than sulfate) limited is also increased using the new formulations, which provide yet further increases in waste treatment rates.

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