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Evaluation of Immobilizing Secondary Waste from a Proposed Treatment Process for Hanford WTP LAW Melter Condensate

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EXECUTIVE SUMMARY

The off-gas system in the Low Activity Waste (LAW) vitrification facility at the Hanford Waste Treatment and Immobilization Plant (WTP) will generate an aqueous condensate recycle stream. Two unit operations generate this condensate, a Submerged Bed Scrubber and a Wet Electrostatic Precipitator (LAW SBS-WESP condensate). The expected disposition of this stream during baseline operations is to send it to the WTP Pretreatment Facility, where it will be blended with LAW, concentrated by evaporation and recycled to the LAW vitrification facility again. The primary reason to recycle this stream is to immobilize the semi-volatile ^{99}Tc isotope into the glass. During the Direct Feed Low Activity Waste (DFLAW) portion of the mission, the condensate is sent to the Effluent Management Facility (EMF) where it is evaporated then recycled to the LAW facility where the evaporated condensate is combined with the LAW feed from the pretreatment system.

Because the condensate stream also contains non-radioactive salt components that are problematic in the melter, diversion of this stream to another process would eliminate recycling of these salts and would enable simplified operation of the LAW melter and the Pretreatment Facilities. Diversion from recycling this stream within WTP would have the effect of decreasing the LAW vitrification mission duration and quantity of Immobilized LAW. To enable diversion, a process is being developed by Savannah River National Laboratory to remove the ^{99}Tc so that the decontaminated aqueous stream, with the problematic salts, can be disposed elsewhere.

Previous lab-scale testing has shown $SnCl_2$ to be an effective agent for the ^{99}Tc removal through reductive precipitation. The removal is believed to work by reducing the Tc(VII) ion in the soluble pertechnetate (TcO_4^-) to Tc(IV), leading to precipitation of technetium dioxide (TcO_2). The technetium dioxide is a minor constituent in the slurry, which is predominantly tin and chromium-containing solids. Other minor constituents will also be present in this stream, such as Hg, Zn, and silica. In total, the precipitation process adds ~1 gram/liter of insoluble solids to the condensate which also has a small amount of entrained solids from the melter, which would presumably be concentrated by clarification and/or filtration. This report documents the results of a preliminary evaluation of the disposal path for the Tc-containing stream generated by this process.

Based on the results of this assessment, it appears that the solid in the slurry from the ^{99}Tc-precipitation can be recycled back to the front end of the LAW vitrification process. Even if three times the projected steady state volume of condensate is processed per Concentrate Receipt Vessel (CRV) batch, the resulting LAW glass contains 0.42 weight percent (wt.%) SnO_2 at an Na_2O loading of 30 wt.%. This concentration of SnO_2 has been shown to have an inconsequential impact on the predictions for viscosity and the Product Consistency Test (PCT) response. Based on the amount of precipitated solids being recycled, there is expected to be little impact on melter feed rheology and melting behavior. Since the projected concentration of added SnO_2 is within the validity constraints of the 2016 glass property models, no experimental testing is recommended.

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LIST OF ABBREVIATIONS

CRV	Concentrate Receipt Vessel
DFLAW	Direct Feed Low Activity Waste
DOE	Department of Energy
EMF	Effluent Management Facility
ETF	Effluent Treatment Facility
HLW	high-level waste
IDF	Integrated Disposal Facility
LAW	low-activity waste
LERF	Liquid Effluent Retention Facility
PA	Performance Assessment
PCT	Product Consistency Test
SBS	Submerged Bed Scrubber
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
VHT	Vapor Hydration Test
WESP	Wet Electrostatic Precipitator
wt.%	weight percent
WTP	Waste Treatment and Immobilization Plant

1.0 Introduction

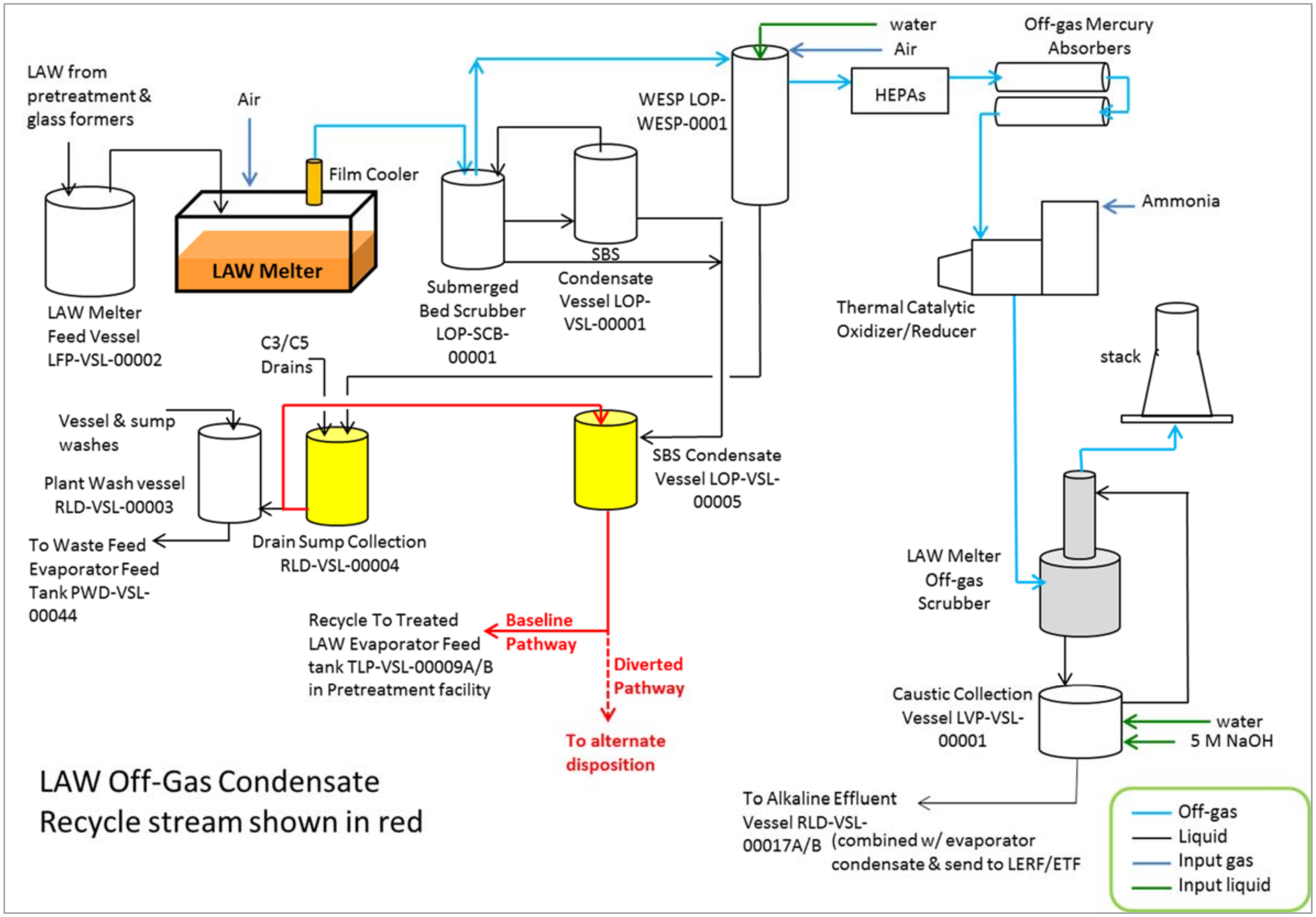
The Hanford low-activity waste (LAW) off-gas condensate stream will be generated in the Waste Treatment and Immobilization Plant (WTP) during Direct Feed LAW (DFLAW) operations by condensation and scrubbing of the LAW melter off-gas by a submerged bed scrubber (SBS) and wet electrostatic precipitator (WESP), as shown in Figure 1-1.[1] Pilot simulant tests[2] indicate that this SBS-WESP condensate stream is expected to be a dilute salt solution with near neutral pH, and will contain some insoluble solids from melter carryover. The soluble salts are expected to be mostly sodium and ammonium salts of nitrate, chloride, fluoride, and sulfate. The expected disposition of this stream during baseline operations is to send it to the WTP Pretreatment Facility, where it will be blended with LAW, concentrated by evaporation and recycled to the LAW vitrification facility again During the DFLAW portion of the mission, the condensate is sent to the Effluent Management Facility (EMF) where it is evaporated then recycled to the LAW facility where the evaporated condensate is combined with the LAW feed from the pretreatment system.[1]

The primary reason to recycle this stream is to allow higher retention of the semi-volatile ^{99}Tc isotope into the glass than would occur in a single pass system. Although other radionuclides are expected to be present at low concentrations in the LAW off-gas condensate, such as ^{129}I, ^{90}Sr, ^{137}Cs, ^{241}Pu, and ^{241}Am, it is the long-lived and environmentally mobile ^{99}Tc that is the primary constituent of concern. Because ^{99}Tc has a very long half-life and is highly mobile, it is the largest dose contributor to the Performance Assessment (PA) of the Integrated Disposal Facility (IDF).[3]

Because this stream contains non-radioactive salt components (halides and sulfate) that are problematic in the melter and reduce Na_2O loading, diversion of this stream to another process would:

- Eliminate recycling of these salts
- Enable simplified operation of the LAW melter and the Pretreatment Facilities
- Decrease the LAW vitrification mission duration, cost and quantity of glass waste

To enable diversion, a process[4] is being developed by the Savannah River National Laboratory (SRNL) to remove the ^{99}Tc so that the decontaminated aqueous stream, with the problematic halide and sulfate salts, can be disposed elsewhere (Figure 1-2). Previous lab-scale testing[5-10] has shown $SnCl_2$ to be an effective agent for the ^{99}Tc removal through reductive precipitation. The removal is believed to work by reducing the Tc(VII) ion in the soluble pertechnetate (TcO_4^-) to Tc(IV), leading to precipitation of technetium dioxide (TcO_2). The TcO_2 is a minor constituent in the slurry, which is predominantly tin and chromium-containing solids. The chromium is present in the SBS-WESP stream due to partial volatility from the melter and is initially soluble. The chromium precipitates because it gets reduced to Cr(III) as a consequence of reaction with the stannous ion. Other minor constituents will also be present in this stream, such as Hg, Zn, and silica. In total, the precipitation process adds ~1 gram/liter of insoluble solids to the condensate which has a small amount of entrained solids from the melter, which would presumably be concentrated by clarification and/or filtration.



Adapted from 24590-WTP-RPT-PT-02-005, Rev. 6[11]
Yellow indicates SBS/WESP LAW off-gas condensate collection tanks, red lines indicate the collected off-gas condensate pathway

Figure 1-1. Simplified LAW off-gas system (baseline WTP operations).

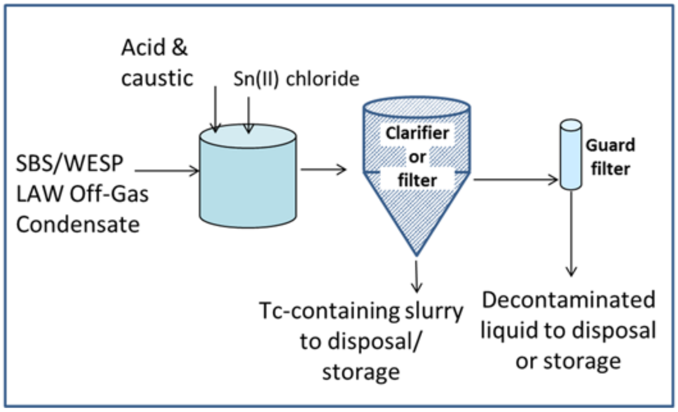


Figure 1-2. Schematic of a Proposed Decontamination Process for LAW Off-Gas Condensate

The scope of this task was a preliminary evaluation to determine whether the solids in the slurry from the ^{99}Tc-precipitation could be recycled back to the front end of the LAW vitrification process. Although other disposal options also exist, this method seems the most practical in the short term. Disposal in the HLW melter is another potential path, but the HLW melter will begin operation after the LAW melter, so the Tc removed by this process would have to be returned to the tank farms or otherwise stored in a slurry phase for the interim period. Storage in a slurry phase is less desirable because it is expected that the ^{99}Tc would re-dissolve as TcO_4^- due to air oxidation, and increase its environmental risk. Another option is to generate a Tc-specific glass waste form for this slurry as was previously evaluated by Los Alamos National Laboratory.[12] While this approach is potentially optimal in the long term because the waste form could be tailored to retain Tc and minimize volume, development of such a waste form is likely years away and very costly. Other alternative disposal paths, including tank farm storage options, will be pursued in the future if LAW glass disposal is not practical.

2.0 Quality Assurance

This test program was directed by a Task Technical and Quality Assurance Plan.[13] Requirements for performing reviews of technical reports and the extent of review are established in Manual E7, Procedure 2.60.[14] SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.[15]

3.0 Results

The Integrated Flowsheet[1] was used as the basis for this evaluation, which utilizes the baseline process shown in Figure 3-1. The SBS-WESP condensate during DFLAW operations is sent to the EMF where it is evaporated and recycled to LAW Concentrate Receipt Vessel (CRV).

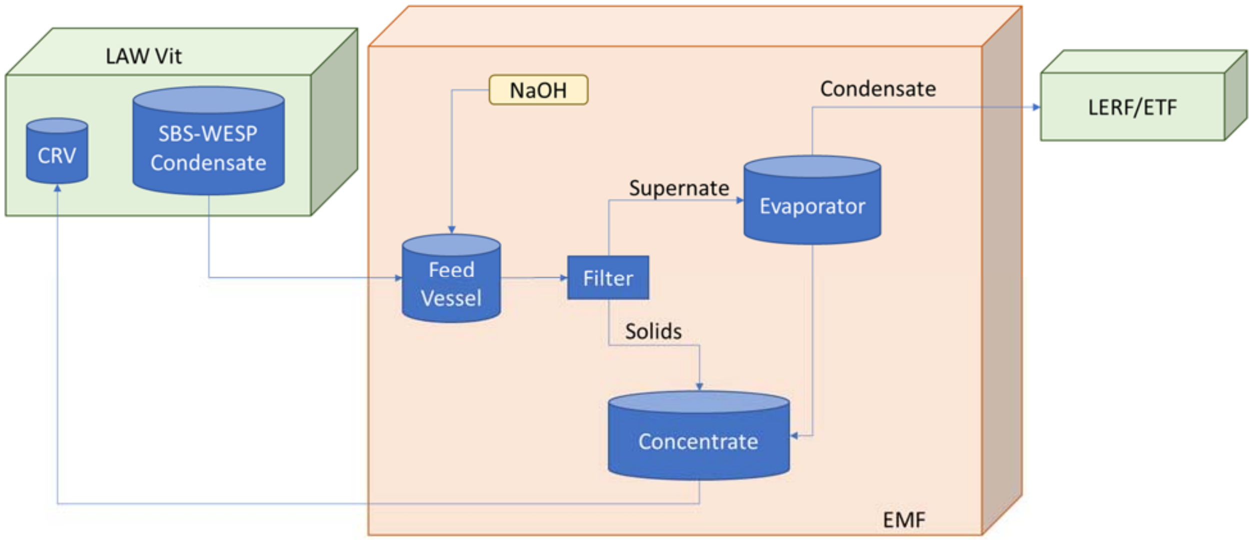


Figure 3-1. Simplified EMF flow diagram assumed for this evaluation.

Several assumptions were needed to perform the evaluation as listed below:

- Average LAW feed and condensate compositions from the Integrated Flowsheet[1] were used to determine the amount of Sn required for the precipitation.
- Only the solids portion of the stream was recycled to the front end of the LAW Vitrification process (into the CRV).
  - Amount of supernate needed to transfer the material has not been finalized.
  - Solids/liquid separation technique not defined, but could be performed by the following:
    - Settle and decant
    - Filtration
    - Cyclone separator
    - Centrifuge
- Solids in the condensate are already included in the Integrated Flowsheet compositions other than the Sn added to precipitate ^{99}Tc since the entire condensate stream is currently recycled to LAW.
- The liquid portion of the recycle stream containing the halide and sulfate salts is sent to the Liquid Effluent Retention Facility (LERF)/ Effluent Treatment Facility (ETF) or alternative treatment and is not recycled.
- Compositions in the Integrated Flowsheet were not adjusted to account for the lack of recycle for soluble species.

A simplified diagram of the modified process flows for treating the condensate from the LAW melters to remove ^{99}Tc based on the bench-scale studies is shown in Figure 3-2.[10] Condensate from the SBS-WESP system is collected and sent to a reactor where $SnCl_2$ is added. Technetium and chromium are precipitated from the condensate. Solids formed from this reductive precipitation include SnO_2, TcO_2 and Cr_2O_3.[8] Liquid effluent is sent to the LERF and treated at the ETF while the precipitated solids are returned to the CRV in the LAW vitrification facility. It has been shown that the solids in this slurry will dissolve when mixed with LAW,[8] so the impact on LAW melter feed rheology would be expected to be insignificant.

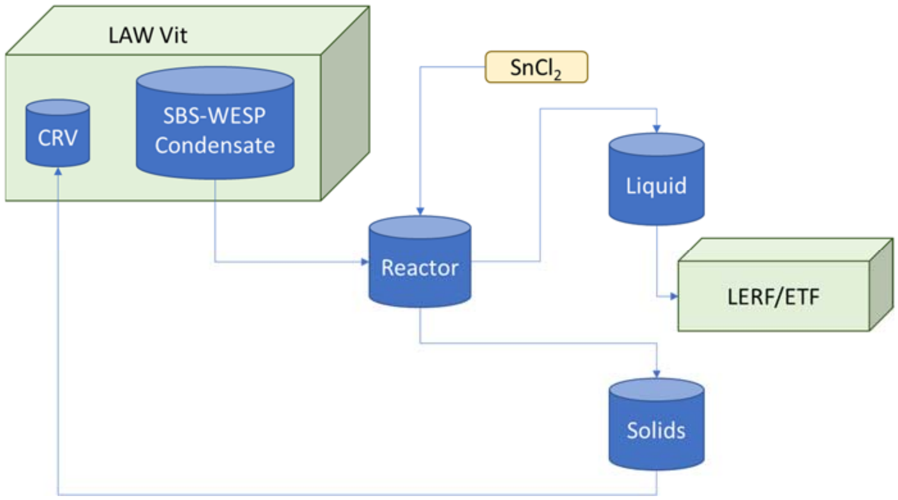


Figure 3-2. Simplified flow diagram for ^{99}Tc removal using the $SnCl_2$ precipitation process.

Calculations based on the Integrated Flowsheet indicate that the 0.808 grams of $SnCl_2$ per liter of condensate determined from experimental testing at SRNL[8,10] is a bounding value for the $SnCl_2$ needed in the ^{99}Tc precipitation process assuming a 1.5 stoichiometric excess based on the number of electrons needed to reduce Cr(VI) and Tc(VII). Further calculations show that the amount of Sn added to the condensate and subsequently recycled to the front end of the LAW vitrification process would result in less than 0.5 weight percent (wt.%) SnO_2 in glass for Na_2O loading up to 30%, even if three times the projected steady state volume of condensate is processed per CRV batch (Table 3-1). While the current WTP baseline maximum Na_2O loading is 21 wt.%,[16] the SnO_2 concentration was determined up to an Na_2O loading of 30 wt.% to maximize the impact of SnO_2 on the LAW glass properties.

Table 3-1. SnO_2 Concentrations in Glass as a Function of Na_2O Loading

Na_2O Loading (wt.%)	10	15	20	25	30
Steady State Condensate Volume	0.05	0.07	0.09	0.12	0.14
3X Condensate Volume	0.14	0.21	0.28	0.35	0.42

It should be expected that the minimization of sulfate, fluorine, and chlorine in the recycle stream would allow higher Na_2O loading in the LAW glass compared to the baseline flowsheet, which recycles ~100% of these components. However, calculations to show how the concentration of soluble semi-volatiles in the final LAW glass would be decreased by minimization of the soluble species in the recycle stream to LAW were outside the scope of this preliminary evaluation.

To determine the potential impact of the additional Sn from the precipitate on the LAW glass waste form, it was desired to predict the properties of LAW glass compositions with and without Sn. While the current WTP baseline models[17,18] do not account for SnO_2, this term has been included in more recent glass property models that were developed in 2013[19] and updated in 2016[16] to further increase waste loading and reduce life-cycle glass mass estimates. These models are based on previous testing[20-24], which has shown that the Vapor Hydration Test (VHT) performance is improved when SnO_2 is added as a glass former additive, especially at increased Na_2O loading. Generally, the models have been validated for up to ~ 5 wt.% SnO_2. Based on the current quality assurance level for these models, they are only intended for use in mission planning activities,[16] which is sufficient for this preliminary evaluation.

Sample LAW glass compositions were selected from previous glass formulation studies performed for Envelopes A (AN-105)[20], B (AZ-101)[20], and C (AN-102).[25] The target concentrations are provided in

Table 3-2 along with the renormalized compositions that have been spiked with maximum SnO_2 concentration shown in Table 3-1 (0.42 wt.%).

Table 3-2. Sample LAW Glass Compositions (wt.%) and Predicted Properties

Component	LAWA171 Target	LAWA171 with SnO_2	LAWB101 Target	LAWB101 with SnO_2	LAWC102 Target	LAWC102 with SnO_2
Al_2O_3	10.16	10.12	10.15	10.11	8.64	8.60
B_2O_3	13.68	13.62	10.01	9.97	13.68	13.62
CaO	5.65	5.63	11.21	11.16	8.02	7.99
Cl	0.65	0.65	0.01	0.01	0.65	0.65
Cr_2O_3	0.02	0.02	0.11	0.11	0.02	0.02
F	0.00	0.00	0.07	0.07	0.19	0.19
Fe_2O_3	1.00	1.00	1.15	1.15	1.00	1.00
K_2O	0.51	0.51	0.41	0.41	0.15	0.15
Li_2O	0.00	0.00	3.54	3.53	0.00	0.00
MgO	1.00	1.00	1.15	1.15	1.00	1.00
Na_2O	23.00	22.90	10.00	9.96	20.00	19.92
NiO	0.00	0.00	0.00	0.00	0.03	0.03
P_2O_5	0.00	0.00	0.03	0.03	0.27	0.27
PbO	0.00	0.00	0.00	0.00	0.01	0.01
SiO_2	36.58	36.42	43.08	42.90	36.62	36.46
SnO_2	0.00	0.42	0.00	0.42	0.00	0.42
SO_3	0.75	0.75	0.75	0.75	1.20	1.19
V_2O_5	1.00	1.00	1.24	1.23	1.00	1.00
ZnO	3.00	2.99	3.54	3.53	3.00	2.99
ZrO_2	3.00	2.99	3.54	3.53	4.52	4.50
Property Predictions						
PCT (g/L)	3.40	3.17	0.24	0.23	1.79	1.68
Difference (g/L)	-0.23		-0.01		-0.10	
Viscosity (Pa·s)	2.37	2.41	3.80	3.86	2.58	2.63
Difference (Pa·s)	0.04		0.06		0.04	

Each of the LAW glass compositions was evaluated using the 2016 enhanced glass-property models[16] for viscosity and the Product Consistency Test (PCT) response.[a] The predicted properties are shown in

[a] To ensure that the viscosity and durability (PCT) models were coded correctly prior to the evaluation, the calculation examples in Section 3.8 of PNNL-25835 were performed. Differences of less than 0.02 g/L and 0.02 Pa·s were noted for the PCT and viscosity models, respectively. These small differences were likely attributed to rounding and are insignificant.

Table 3-2 along with the differences between the predictions for compositions with and without the SnO_2. The differences for the PCT and viscosity predictions are less than 0.3 g/L and 0.1 Pa·s, respectively. Thus, the additional SnO_2 from the precipitation process is not expected to have a significant impact on the glass properties. Based on the previous work[20-24] and the results of these model predictions, the presence of additional SnO_2 in the LAW glass should have a beneficial impact on durability.

Previous DM10 melter testing successfully utilized SnO_2 as a glass former additive in the melter feed up to ~2.8 wt.%,[22] which exceeds the maximum SnO_2 concentration determined in this evaluation (0.42 wt.%). Thus, LAW melter feed containing the solids from the precipitation process would be expected to have similar melting behavior to those melter feeds already evaluated in the DM10 melter testing.

4.0 Conclusions

Based on the results of this preliminary evaluation, it appears that the solids slurry from the ^{99}Tc-precipitation can be recycled back to the front end of the LAW vitrification process. Even if three times the projected steady state volume of condensate is processed per CRV batch, 0.42 wt.% SnO_2 in glass at an Na_2O loading of 30 wt.% would be subsequently recycled. This concentration of SnO_2 has been shown to have an inconsequential impact on the predictions for viscosity and the PCT response. Based on the amount of precipitated solids being recycled, there is expected to be little impact on melter feed rheology and melting behavior. Therefore, no experimental testing is recommended.

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