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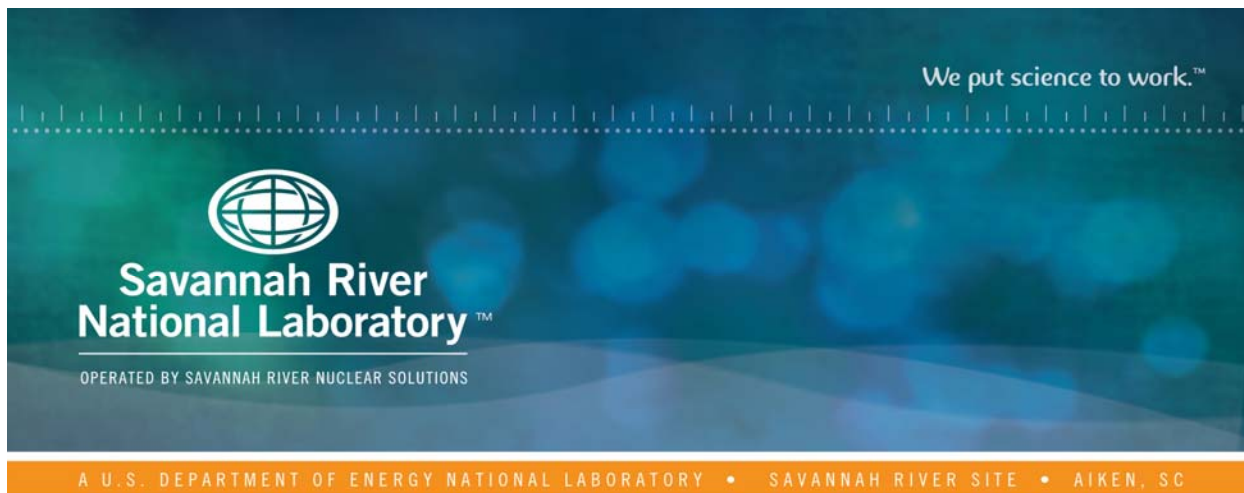
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# Bench Scale Experiments for the Remediation of Hanford Waste Treatment Plant Low Activity Waste Melter Off-Gas Condensate

Kathryn M. L. Taylor-Pashow

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August 2017

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## EXECUTIVE SUMMARY

The Low Activity Waste (LAW) vitrification facility at the Hanford Waste Treatment and Immobilization Plant (WTP) will generate an aqueous condensate recycle stream (LAW Off-Gas Condensate) from the off-gas system. The plan for 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 so that the semi-volatile  $^{99}\text{Tc}$  isotope eventually becomes incorporated into the glass. This stream also contains non-radioactive salt components that are problematic in the melter, so 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. This diversion from recycling this stream within WTP would have the effect of decreasing the LAW vitrification mission duration and quantity of glass waste. The concept being tested here involves removing the  $^{99}\text{Tc}$  so that the decontaminated aqueous stream, with the problematic salts, can be disposed elsewhere.

Technetium will not be removed from the aqueous tank waste during pretreatment in the Hanford WTP and will be sent to the LAW melter. It is intended that  $^{99}\text{Tc}$  will be immobilized in the LAW glass. Because it is semi-volatile at melter temperatures and roughly 70% vaporizes, the only way to get it to stay in the glass is by repeated recycle into the LAW melter. Although other radionuclides are expected to be present in low concentration in the LAW Off-Gas Condensate, such as  $^{129}\text{I}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Pu}$ , and  $^{241}\text{Am}$ , it is the long-lived and environmentally mobile  $^{99}\text{Tc}$  that is the primary component of concern.

The LAW Off-Gas Condensate stream originates from the Submerged Bed Scrubber (SBS) and the Wet Electrostatic Precipitator (WESP) from the LAW melter off-gas system. Pilot simulant tests indicate that this stream is expected to be a dilute salt solution with near neutral pH, and will likely contain some insoluble solids from melter carryover. The soluble salt components are expected to be mostly sodium and ammonium salts of nitrate, chloride, fluoride, and sulfate. Although this stream has not yet been generated and will not be available until the WTP begins operation, a simulant has been produced based on models, calculations, and comparison with non-radioactive pilot-scale tests using simulants of the actual LAW waste.

The recycled components in the LAW Off-Gas Condensate that are problematic for the glass waste form are halides and sulfate, which are volatile at melter temperatures. Recycling in order to incorporate the  $^{99}\text{Tc}$  in the glass causes these components to accumulate in the Condensate stream, exacerbating their impact on the number of LAW glass containers that must be produced. Diverting the stream reduces the halides and sulfate in the melter and is a key outcome of this work. Additionally, under the Direct Feed LAW (DFLAW) scenario, where the LAW vitrification facility commences operation prior to the WTP Pretreatment facility, identifying a disposition path becomes vitally important because the evaporator in the Pretreatment facility will not be operational. The current plan is to construct the Effluent Management Facility (EMF) to concentrate the stream for return to the melter. This task examines the potential treatment of this stream to precipitate radioactive  $^{99}\text{Tc}$  and subsequently disposition the decontaminated aqueous stream elsewhere, perhaps at the Effluent Treatment Facility (ETF) or through an altered EMF where the Tc-decontaminated and concentrated stream is immobilized as a low temperature waste form. The treatment process envisioned focuses on using mature radionuclide removal technologies that are also compatible with long-term tank storage and immobilization methods. For this new process, testing is needed to demonstrate acceptable precipitation agents and measure decontamination factors for Tc removal from this unique waste stream.

Previous work has shown  $\text{SnCl}_2$  to be an effective agent for the  $^{99}\text{Tc}$  removal from this stream through reductive precipitation. The removal is believed to work by reducing the Tc(VII) ion in the soluble

pertechnetate ( $\text{TcO}_4^-$ ) to  $\text{Tc(IV)}$ , leading to precipitation of a technetium dioxide ( $\text{TcO}_2$ ). The present work focused on experiments needed to begin to mature the technology readiness of this process. A key component of that readiness is the scale-up of the reaction and the solid-liquid separation method. These two are related because the mixing of the chemicals during the reaction affects the particle size of the solids, thereby impacting the solid-liquid separation method. Prior kinetics testing has been performed to determine the speed of the Tc removal reaction so that appropriate equipment for mixing scale-up tests could be selected. It was found previously that at small scale, the Tc is removed to below the detection limit within 5 minutes of the addition of stannous chloride. The chromium is similarly removed very quickly, although there may be some small delay in reaching equilibrium. In order to perform the bench scale experiment as a non-radioactive test, the precipitation of chromium was studied. Based on previous small scale testing, the kinetics of the chromium reduction will serve as a reasonable and conservative surrogate for the Tc reduction kinetics. Results from the bench scale mixing experiment indicated that the Cr precipitation is complete within 5 minutes of the addition of stannous chloride, as was seen at smaller scale.

An initial look at solid-liquid separation was also performed by determining the particle size distribution of the precipitate from the bench scale experiment, and also measuring the rate at which the solids settle. The solids generated in these experiments are predominantly Cr and Sn oxide and hydroxyoxide species, which will make up the majority of the precipitate in the actual process. The Tc is present at such a relatively small concentration that the amount of technetium oxide precipitate will not affect the bulk characteristics of the precipitate. The objective was to examine the mixing and bulk properties of the slurry, so  $^{99}\text{Tc}$  was omitted from the testing to avoid generating a radioactive waste unnecessarily. Particle size distribution measurements showed a median particle size of 13.5 microns (volume distribution), with a distribution of sizes from 1.7 microns at the 10<sup>th</sup> percentile to 55.5 microns at the 95<sup>th</sup> percentile. The particle settling rate was measured at the conclusion of the bench scale mixing experiment using the same vessel and was found to have an initial rate of settling of approximately 60 inches per hour. This rate decreased with time and was completely settled after approximately one hour. At that time the solids were in a settled layer, approximately 10% of the liquid height.

Additional tasks needed to further develop this technology include slurry rheology measurements, corrosion and erosion studies, and slurry storage and immobilization.

## TABLE OF CONTENTS

LIST OF TABLES .....	viii
LIST OF FIGURES .....	viii
LIST OF ABBREVIATIONS .....	ix
1.0 Introduction .....	1
1.1 Simulant Formulation Basis .....	3
1.2 Decontamination Process .....	3
2.0 Experimental Procedure .....	4
2.1 Simulant Preparation .....	4
2.2 Mixing Calculations .....	5
2.3 Bench Scale Precipitation Experiment .....	6
2.4 Solids Settling Experiment .....	7
2.5 Quality Assurance .....	7
3.0 Results and Discussion .....	7
3.1 Simulant Composition .....	7
3.2 Bench Scale Precipitation Experiment .....	8
3.3 Settling Experiments .....	9
3.4 Sample Calculation for Full-Scale Reaction Vessel .....	12
3.5 Solid-Liquid Separation .....	13
4.0 Conclusions .....	14
5.0 Future Work .....	14
6.0 References .....	14
Appendix A . Detailed Results .....	A-1



## LIST OF TABLES

Table 2-1. Non-Radioactive Simulant Formulation Targets.....	5
Table 3-1. Neutralized SBS/WESP Simulant Filtrate Composition .....	8
Table 3-2. Cr Decontamination Factors for Bench Scale Experiment.....	9
Table 3-3. Settling Rates.....	9

## LIST OF FIGURES

Figure 1-1. Simplified LAW Off-gas System.....	2
Figure 1-2. Schematic of a Proposed Decontamination Process and Disposition Path of LAW Off-Gas Condensate.....	3
Figure 3-1. Photographs of bench scale mixing experiment taken before (left) and approximately 5 minutes after (right) the addition of stannous chloride.....	8
Figure 3-2. Settling rate versus time. ....	10
Figure 3-3. Photographs taken at various time points during solids settling. ....	11
Figure 3-4. Particle size distribution (volume) of precipitate from stannous chloride reductive precipitation reaction. ....	12

## LIST OF ABBREVIATIONS

ARP	Actinide Removal Process
Avg	Average
DF	Decontamination Factor
DOE	Department of Energy
eq	Equivalents
ETF	Effluent Treatment Facility
g	grams
GFC	Glass Forming Chemical
h	Hour
HLW	High-Level Waste
HTWOS	Hanford Tank Waste Operations Simulator
IC	Ion Chromatography
ICP-ES	Inductively Coupled Plasma – Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma – Mass Spectrometry
IDF	Integrated Disposal Facility
L	Liter
LAW	Low-Activity Waste
LERF	Liquid Effluent Retention Facility
mg	milligram
mL	milliliter
MST	Monosodium Titanate
PA	Performance Assessment
SBS	Submerged Bed Scrubber
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
µg	Micrograms
WESP	Wet Electrostatic Precipitator
WRPS	Washington River Protection Solutions
WTP	Waste Treatment and Immobilization Plant

## 1.0 Introduction

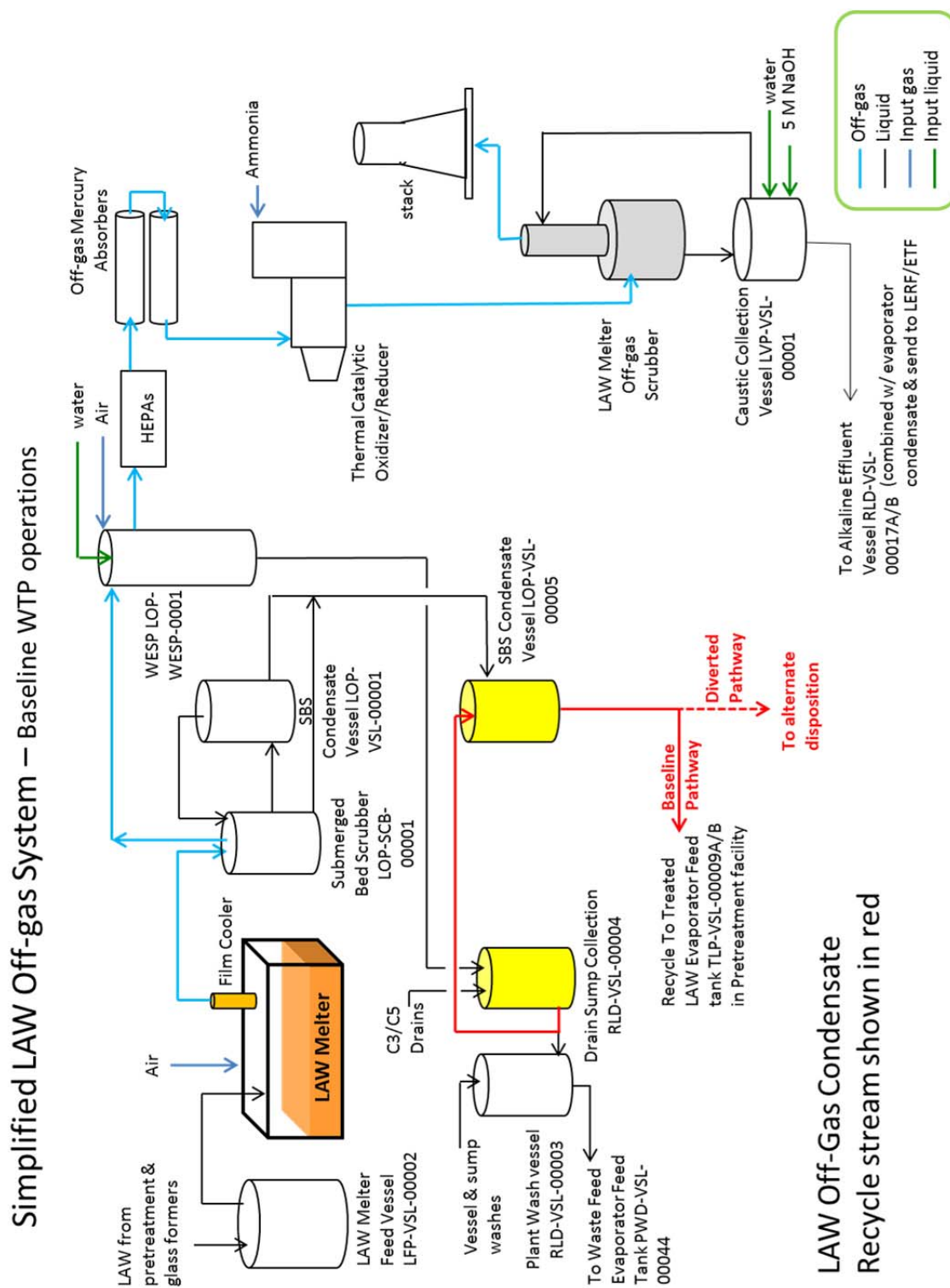
The Hanford LAW Off-Gas Condensate stream will be generated in the WTP by condensation and scrubbing of the LAW melter off-gas system by a SBS and WESP, as shown in Figure 1-1. This stream, which will contain substantial amounts of chloride, fluoride, ammonia, and sulfate ions, will get recycled within the WTP process by return to the Pretreatment Facility where it will be combined with LAW and evaporated. Although the SBS and WESP streams *can* be separately routed to different points in the WTP, they are combined for purposes of this study since they ultimately re-combine during normal operations within the process shortly after generation and they each contain a substantial portion of the  $^{99}\text{Tc}$ . The halide and sulfate components are only marginally soluble in glass, and often dictate glass waste loading and thereby impact LAW waste glass volume. Additionally, long-lived  $^{99}\text{Tc}$  and  $^{129}\text{I}$  are volatile radionuclides that accumulate in the LAW system, and are challenging to incorporate in glass under the Hanford LAW melter operating conditions. Because  $^{99}\text{Tc}$  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)<sup>1</sup>, although the glass waste form has been shown to meet the leaching requirements of the IDF waste acceptance criteria. Diverting this LAW Off-Gas Condensate stream to an alternate disposal path would have substantial beneficial impacts on the cost, life cycle, and operational complexity of WTP because it would reduce the halides and sulfate in the melter feed.

The only chemical form of  $^{99}\text{Tc}$  expected in the off-gas condensate stream is pertechnetate anion ( $\text{TcO}_4^-$ ) with a +7 Tc oxidation state because the high melter temperature should decompose any other form, although this has not been definitively proven. The volatility of Tc under oxidizing melter conditions is well known, where the most likely volatile species is the heptoxide ( $\text{Tc}_2\text{O}_7$ ) because of its boiling point of 311 °C, although it could also sublime as an alkali metal pertechnetate, ammonium pertechnetate, or perhaps  $\text{TcO}_2$ .<sup>2</sup> There is no direct evidence that  $\text{Tc}_2\text{O}_7$  is the actual volatile species because the characterization is based on examining the condensed product and not directly on the vapor. Once the  $\text{Tc}_2\text{O}_7$  contacts water, it would disproportionate to the pertechnetate.<sup>3</sup>

The objective of this development task is to evaluate decontamination of this stream using sorbents and/or precipitation agents so that it can be diverted elsewhere (Figure 1-2). The equipment needed for this process would be comparable to the ARP<sup>a</sup> at SRS that has been operating successfully for years. Although that process treats tank waste (comparable to “LAW” at Hanford), it demonstrates successful deployment of filtration processes for radionuclide removal using a porous stainless steel filter. The concept for this new process utilizes common industrial chemicals and equipment. This task specifically examined removal of  $^{99}\text{Tc}$  using reducing agents, but other sorbents may be needed if removal of other radionuclides is required. Use of these inorganic materials is expected to simplify down-stream issues, such as storage and immobilization. Implementation of this process at WTP would make available both a short-term disposition path if the LAW facility commences operation prior to operation of the Pretreatment Facility and in the long term to divert the stream from recycling. Although Figure 1-2 indicates sending the decontaminated liquid to the ETF, other paths may also be viable options. The ETF is used here as an example of a potential path and is used for an estimation of decontamination requirements.

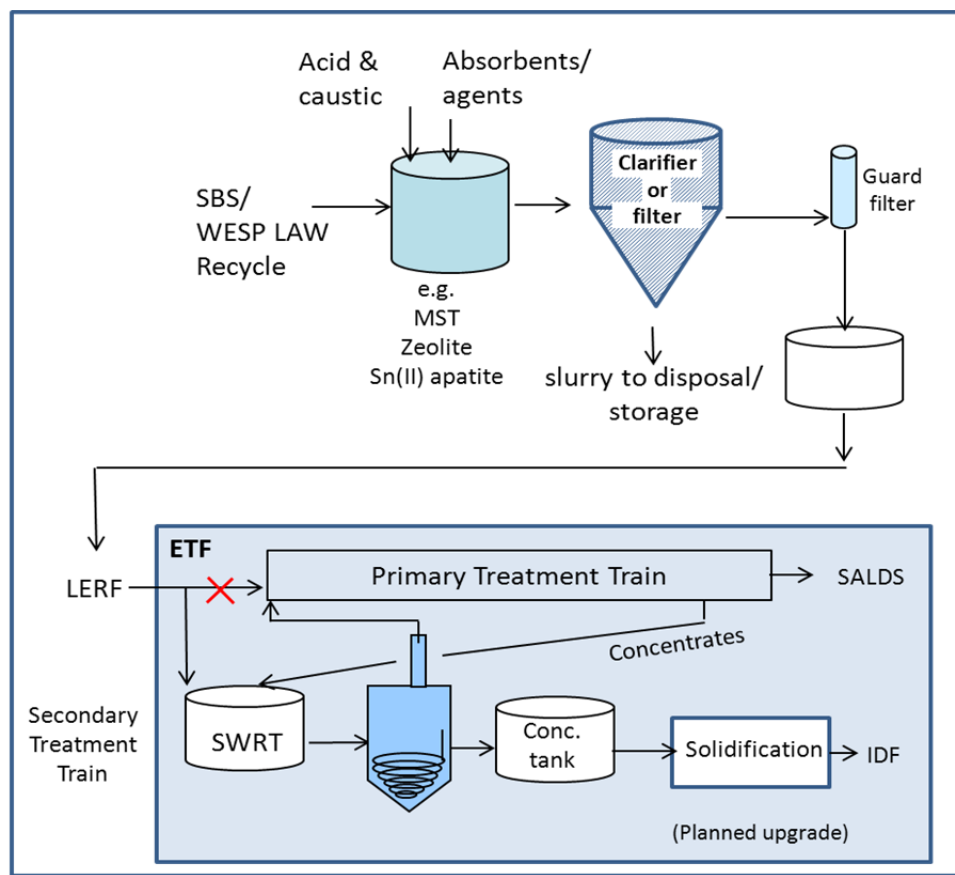
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<sup>a</sup> The Actinide Removal Process (ARP) at SRS decontaminates  $^{90}\text{Sr}$  and actinides from aqueous tank waste before it is further treated for  $^{137}\text{Cs}$  removal by solvent extraction. In ARP, a small amount of Monosodium Titanate (MST) is added to a batch of decanted tank waste supernate and mixed for 6-12 hours, then filtered with a cross-flow stainless steel filter. The spent MST that is loaded with  $^{90}\text{Sr}$  and actinides is washed with water, and sent for vitrification as HLW glass in the DWPF.



(adapted from 24590-WTP-RPT-PT-02-005, Rev. 6); (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**



**Figure 1-2. Schematic of a Proposed Decontamination Process and Disposition Path of LAW Off-Gas Condensate**

The overall plan for technology development of this process, along with options for disposal has been documented.<sup>4</sup> The preliminary testing of this process has also been documented.<sup>5,6</sup> Other alternative disposal paths for the Tc-containing slurry could be considered as well, including tank farm storage options.

### 1.1 Simulant Formulation Basis

Because this stream is not yet available for characterization, the simulant formulation was based on input from two sources. The projected solution chemistry was based on version 7.4 of the Hanford Tank Waste Operations Simulator (HTWOS) modeling of the flow sheet<sup>7</sup> performed by WRPS.<sup>8</sup> This model run was for the average composition of this stream for the entire WTP mission (all 177 tanks) and with full integration of all WTP pretreatment processes, such as caustic leaching, oxidative leaching, and cesium ion exchange. Additional information on composition was obtained from analysis of samples obtained from pilot-scale melter testing using simulated LAW feed. More detail on the basis for and synthesis of the simulant has been documented.<sup>5,6,9</sup>

### 1.2 Decontamination Process

One option that has been previously evaluated is disposal of the LAW Off-Gas Condensate stream directly to the ETF, however, this option has a number of consequences to ETF including increases in waste volume, halide levels, and radioactivity.<sup>10,11</sup> These have not been evaluated any further in this study, but it is recognized that this would have significant impacts that must be addressed by the facility.

The LAW Off-Gas Condensate stream is expected to contain  $^{99}\text{Tc}$  due to its volatility at melter temperatures. The only chemical form of  $^{99}\text{Tc}$  expected in the stream is pertechnetate anion ( $\text{TcO}_4^-$ ) with a +7 technetium oxidation state. Although some fraction of the  $^{99}\text{Tc}$  is present in the initial LAW stream as a soluble “non-pertechnetate” species, the LAW melter is expected to convert it to the same volatile species as the pertechnetate form during calcination reactions that occur during vitrification. The volatile species then becomes pertechnetate ion again when it contacts the water in the SBS and WESP. (Note that this has not been demonstrated.)

The current WTP baseline assumption is that technetium will not be removed from the aqueous waste in the WTP, and will primarily end up immobilized in the LAW glass waste form after several recycle passes to improve retention.<sup>12</sup> The LAW glass will be disposed in the IDF. Because  $^{99}\text{Tc}$  has a very long half-life and is highly mobile,<sup>13,14</sup> it is the major dose contributor to the Performance Assessment (PA) of the IDF,<sup>1</sup> even though it is largely retained by the glass. Due to the high water solubility, high volatility during vitrification, and potential for impact to the PA, effective management of  $^{99}\text{Tc}$  is important to the overall success of the River Protection Project mission. If a process was implemented that allowed disposal of the radionuclides offsite (e.g. by incorporation into HLW glass instead, for example), the amount of  $^{99}\text{Tc}$  disposed in LAW glass at the IDF would decrease substantially.

For this proposed alternative treatment process, separation of the  $^{99}\text{Tc}$  is accomplished by precipitation with chemical reagents, and settling and/or filtration. For the Condensate stream, emphasis was on using entirely inorganic materials to enable easier storage and disposal as immobilized waste. For technetium removal, these materials included reducing agents (e.g. Sn(II) or Fe(II) compounds). Sn(II) with hydroxyapatite and oxalate has previously been found effective for precipitating Tc from water samples;<sup>15</sup> however, previous work by SRNL has shown Sn(II) alone without an absorbent is sufficient for precipitation of the  $^{99}\text{Tc}$  and remains insoluble for at least 72 hours in air.<sup>6,16,17</sup>

For this proposed alternative treatment process, disposal of the aqueous decontaminated Condensate stream at ETF is used as an example pathway. The basis for the target Decontamination Factor (DF) for the radionuclides was described previously.<sup>4</sup> The target DF for  $^{99}\text{Tc}$  based on the current established LERF/ETF limits is only 2, but a DF of 100 was arbitrarily selected to minimize the impact of the final disposed waste form from ETF, which is disposed in IDF. The DF is defined as the initial concentration ( $C_0$ ) divided by the concentration at time  $t$  ( $C_t$ ) (Equation 1).

$$DF = \frac{C_0}{C_t} \quad (1)$$

Other factors that influence particle size of the precipitated solids, such as temperature and method/rate of addition of reagents, will be studied in future work. Immobilization and potential disposition pathways will be evaluated in a subsequent phase of this program, once the slurry composition and quantities are defined.

## 2.0 Experimental Procedure

### 2.1 Simulant Preparation

Detail on the basis and synthesis of the simulant has been documented elsewhere, and is repeated here for completeness.<sup>9</sup> The target concentrations of chemicals were derived from the output from the HTWOS calculation, documented in SVF-2732.<sup>8</sup> A total of 5.0 L of non-radioactive simulant was prepared in three batches (two 2-L batches and one 1-L batch) for this bench scale testing and was also combined

with approximately 500 mL of simulant of the same composition prepared previously.<sup>18</sup> The simulant was prepared from dissolution of laboratory chemicals in deionized water, in the order shown in Table 2-1. Because the HTWOS model is not constrained to generate a charge-balanced composition, no formulation can match all component concentrations simultaneously, and the chemical formulation must balance between cations and anions to create a mixture that can actually be synthesized. Previous simulant preparations have included the addition of glass forming chemicals (GFCs), which were allowed to come to equilibrium with the aqueous phase before the insoluble portion was removed by filtration. Based upon previous simulant analyses, the completely insoluble GFCs were excluded from this preparation, and only the soluble GFCs were added. That included borax, boric acid, lithium carbonate, and sodium carbonate; which were completely soluble at the amounts added. The amount of silica and zinc oxide added is the portion shown to be soluble in previous simulant preparations. After preparing the three batches of simulant, the solutions were mixed for eight to nine days at ambient temperature. The pH of the resulting solutions was measured to be 8.01. The pH of each batch was then adjusted to 7.5 – 7.6 with the addition of 3.1 – 4.6 g of 1 M nitric acid per liter of simulant. After pH adjustment, all three batches of simulant were filtered through a 0.45-micron Nylon filter to remove any insoluble material and were then combined. The remaining ~500 mL of simulant from a previous preparation<sup>18</sup> was also added to the freshly prepared 5 L prior to removing samples for analysis. Duplicate samples were analyzed for elemental composition by Inductively Coupled Plasma – Emission Spectroscopy (ICP-ES), anions and ammonium by Ion Chromatography (IC).

**Table 2-1. Non-Radioactive Simulant Formulation Targets**

Chemical	Formula	Target Mass (g)/L simulant	Target Molarity
Sodium fluoride	NaF	3.209	0.0764
Potassium chloride	KCl	0.219	0.0029
Sodium chloride	NaCl	1.395	0.0239
Sodium chromate	Na <sub>2</sub> CrO <sub>4</sub>	0.283	0.0017
Sodium nitrite	NaNO <sub>2</sub>	0.016	0.0002
Ammonium sulfate	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	3.220	0.0244
Ammonium nitrate	NH <sub>4</sub> NO <sub>3</sub>	2.820	0.0352
Borax	Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub> ·10H <sub>2</sub> O	0.0123	0.00003
Boric acid	H <sub>3</sub> BO <sub>3</sub>	1.430	0.0231
lithium carbonate	Li <sub>2</sub> CO <sub>3</sub>	0.392	0.0053
sodium carbonate monohydrate	Na <sub>2</sub> CO <sub>3</sub> ·H <sub>2</sub> O	0.0035	0.00003
silica	SiO <sub>2</sub>	0.12	0.0020
zinc oxide	ZnO	0.018	0.0002
Sodium nitrate	NaNO <sub>3</sub>	0	0*

\*note that additional nitrate ion is added later as nitric acid during pH adjustment

## 2.2 Mixing Calculations

The rate of the precipitation reaction is a function of the reaction kinetics and mass transfer (i.e., mixing or miscible liquid blending). Wetting and dissolution of the solid stannous chloride would contribute to the overall reaction time, but the stannous chloride was observed to dissolve almost instantaneously, so the dissolution time was assumed to have a small impact on the overall precipitation reaction time. The reaction kinetics will not change with scale, but the mass transfer rate will. The miscible liquid blending time typically increases with scale. Since the objective of the test was to examine the precipitation reaction rather than miscible liquid blending, the test was designed to be kinetically limited.

For the precipitation reaction to be kinetically limited, the tests should be designed to have a blend time much less than 5 minutes, so a blend time of 1 minute was targeted.

The blend time can be predicted with the Grenville correlation which is described by equation [1]

$$N\theta_{95} = 5.2 \frac{T^{1.5}H^{0.5}}{D^2N_p^{1/3}} = 5.2 \frac{(19)^{1.5}(19)^{0.5}}{(5.08)^2(1.27)^{1/3}} = 67 \quad [1]$$

where N is the impeller speed,  $\theta_{95}$  is the time for the fluid to be 95% blended, T is the vessel diameter, H is the height of the vessel contents, D is the impeller diameter, and  $N_p$  is the impeller power number (1.27 for a pitch blade impeller in the turbulent regime).<sup>19</sup> The 95% blend time is the time at which the concentration of the target species is within 5% of its bulk concentration in the vessel. If the impeller rotates at 100 rpm, the calculated blend time is 0.67 minutes. Knowing the 95% blend time, the 99% blend time can be calculated with equation [2].<sup>19</sup>

$$\theta_{99} = 1.537 \theta_{95} \quad [2]$$

From equation [2], the time for the liquid to be 99% blended is 1.03 minutes, which is much less than the 5 minutes for the sampling time. [Since the first sample was collected after 5 minutes, the precipitation reaction may have been complete in much less than 5 minutes. If both the miscible liquid blending and the precipitation occur very quickly (~ one minute), designing a process to perform the precipitation at large scale should not be difficult, so using a 5 minute reaction time duration would be conservative and would bound the maximum tank sizing calculations.]

The impeller Reynolds number can be calculated with equation [3]

$$Re = N D^2 \rho / \mu = (1.667 \text{ sec}^{-1}) (5.08 \text{ cm})^2 (1 \text{ g/cm}^3) / (0.01 \text{ g/cm sec}) = 4,300 \quad [3]$$

where  $\rho$  is the fluid density and  $\mu$  is the fluid viscosity. Since the stream is dilute, the density is assumed to be 1 g/mL, and the viscosity is assumed to be 1 cP. Equation [3] shows the flow in the vessel is in the transition regime. Operating in the transition regime should be sufficient for this small scale test and will provide some conservatism when scaling up the process. The full-scale process should be designed to operate in the turbulent regime (see discussion below).

### 2.3 Bench Scale Precipitation Experiment

A glass reaction vessel was prepared by the SRNL glass shop with an internal diameter of 190 mm and approximate height of 350 mm. To target a 1:1 diameter : height ratio, the vessel was filled to a height of 190 mm with 5.39 L of simulant. An overhead stirrer fitted with a 2-inch pitch blade impeller was used to mix the contents of the tank. This gives an impeller to tank diameter ratio of 0.27. Typical impeller diameter to vessel diameter for pitched blade impellers are 0.25 – 0.5.<sup>20,21</sup> A target of 0.25 was selected to make the blending more challenging in this test. The impeller was positioned in approximately the lower one quarter to one third of the simulant volume. The impeller was rotated at 100 rpm during the experiment. [This impeller type, size and operating speed was selected to provide the mixing needed for this test. Other impeller types, sizes and rotational speeds could be selected to achieve adequate mixing in the bench-scale testing or in the full-scale process.]

The experiment was performed by adding solid stannous chloride dihydrate at a ratio of 1.5:1, with respect to the electrons needed to reduce the Cr present in the simulant. This ratio has been previously shown to be the minimum amount needed to quantitatively reduce the Cr and Tc from the radioactive simulant. This ratio corresponded to a stannous chloride dihydrate concentration of 0.808 g/L in this



simulant. The stannous chloride dihydrate was added as the solid to the top of the vessel, while mixing. The solid was poured directly into the vessel all at once, with the addition complete in less than 10 seconds. The simulant was at ambient laboratory temperature during the experiment, no temperature control was implemented. Samples were then removed after 5, 15, 30, and 60 minutes. Samples were removed by pulling an aliquot of the mixture out and filtering through a 0.1- $\mu$ m syringe filter. Half of the filtrate sample was filtered a second time 24 hours later to confirm there was no post-precipitation after the initial filtration at the sample time. Both sets of samples were submitted for Inductively Coupled Plasma – Mass Spectrometry (ICP-MS) analysis to determine the Cr and Sn concentrations.

#### 2.4 Solids Settling Experiment

At the conclusion of the bench scale mixing experiment (60 minutes of mixing) the mixer was stopped and the initial height of the suspended solids was noted using the graduated markings on the vessel. The solids were then allowed to settle, and the level representing the top of the settled solids was noted periodically while recording the time. These measurements were then used to calculate the settling rate of the solids. The settling experiment was stopped after approximately 28 hours. A sample of the precipitate was then submitted for particle size analysis.

#### 2.5 Quality Assurance

This test program is described in the Task Technical and Quality Assurance Plan for Developing a Flowsheet for Off-Gas Process Liquids from the Hanford Low Activity Waste Vitrification Process.<sup>22</sup> Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. Results are recorded in Electronic Laboratory Notebook #E7518-00211.

### **3.0 Results and Discussion**

#### 3.1 Simulant Composition

Results of the average and standard deviation of the duplicate chemical analysis of the neutralized, filtered SBS/WESP simulant are shown in Table 3-1. These match the target compositions reasonably well, with the exception of nitrate which was low. The small variations are not expected to impact results obtained here. Note that the HTWOS model output is not charge balanced, so it is not possible to create an identical solution. Previous preparations of this simulant have shown the aluminum nitrate and disodium phosphate are insoluble in this simulant, and therefore, they were omitted from this preparation.

**Table 3-1. Neutralized SBS/WESP Simulant Filtrate Composition**

Component	Avg. Concentration (mg/L)	Std. Dev.	HTWOS projection (avg. SVF-2732) (mg/ L)	Avg. Concentration (M)
B	251	0.71	GFC <sup>#</sup>	0.023
Cr	82.8	0.49	91	0.0016
K	114	0	115	0.0029
Li	74.0	0.28	GFC <sup>#</sup>	0.011
Na	2.35E3	7.07	2.29E3	0.10
S	854	0.71	780 (as SO <sub>4</sub> <sup>2-</sup> )	0.025
Si	12.0*	0.14	GFC <sup>#</sup>	4.3E-4
Zn	8.08*	0.035	GFC <sup>#</sup>	1.2E-4
F <sup>-</sup>	1.56E3	4.95	1.45E3	0.082
Cl <sup>-</sup>	982	9.19	950	0.028
NO <sub>2</sub> <sup>-</sup>	11.0	0.071	10.7	2.4E-4
NO <sub>3</sub> <sup>-</sup>	2.55E3	19	5.53E3	0.041
SO <sub>4</sub> <sup>2-</sup>	2.53E3	14	2.34E3	0.026
NH <sub>4</sub> <sup>+</sup>	1.42E3	14	1.51E3	0.079

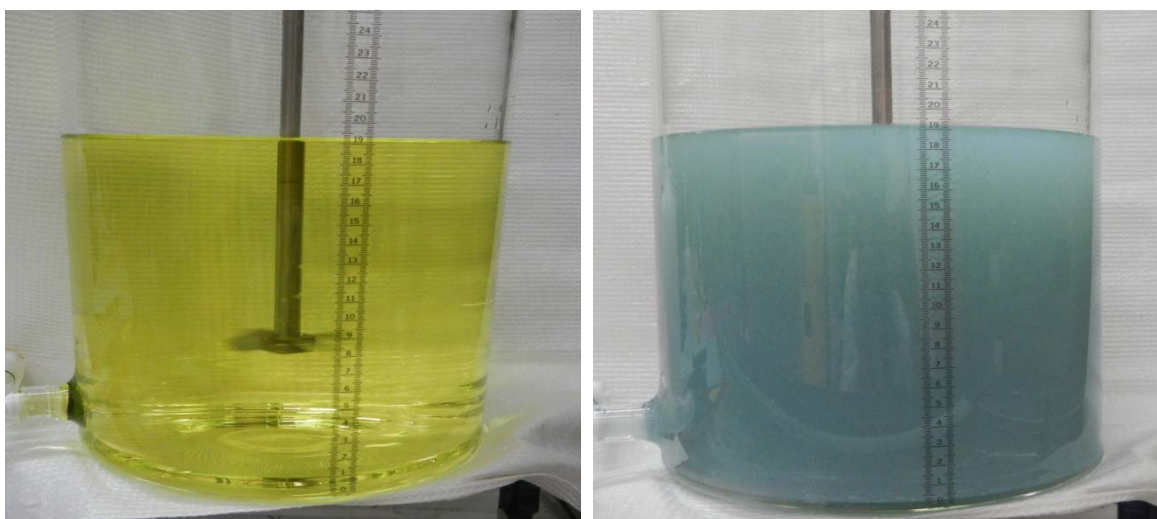
\*Added in reduced amounts compared to previous preparations. Only the expected soluble amount was added.

<sup>#</sup>Glass Forming chemical; minimal HTWOS projected concentration

### 3.2 Bench Scale Precipitation Experiment

The bench scale precipitation experiment used 5.39 L of the SBS/WESP simulant described in the previous section. Stannous chloride was added to the test at the baseline ratio of 1.5 eq. SnCl<sub>2</sub> relative to the equivalents of electrons needed to reduce the Cr (2.25 moles/mole). This corresponded to SnCl<sub>2</sub> concentrations of 0.679 g/L (0.0036 M). Samples were removed after 5, 15, 30, and 60 minutes and were analyzed for Cr and Sn by ICP-MS analysis. Table 3-2 provides the results of these experiments and detailed concentration data can be found in Appendix A.

The color of the simulant began to change almost immediately upon addition of the stannous chloride, indicating reduction of the Cr(VI) (yellow) to Cr(III) (blue/green color). The photographs in Figure 3-1 show the bench scale vessel just before (left) and approximately 5 minutes after (right) the addition of the stannous chloride.



**Figure 3-1. Photographs of bench scale mixing experiment taken before (left) and approximately 5 minutes after (right) the addition of stannous chloride.**

As can be seen from the results presented in Table 3-2, the reductive precipitation of the chromate is complete within 5 minutes. Visual observations of the color change indicated the reaction was likely complete prior to 5 minutes. The color began to change immediately upon the addition of the stannous chloride, and the entire volume appeared to have changed color in less than 2 minutes. There also does not appear to be any post precipitation of Cr after the initial filtration at the time the sample was pulled, as evidenced by the Cr concentrations being equal, within the measurement uncertainty, in the original and post-filtered samples. There is some evidence of Cr redissolving as indicated by the lower DF at 60 minutes compared to the initial sample time; however, the amount is minor compared to the starting concentration of Cr. The DF at 5 minutes corresponds to removal of 99.80% of the Cr versus 99.62% at 60 minutes. The Sn results of the post-filtered samples did indicate a small amount of Sn precipitated after the original sample was filtered (see Appendix A); however, the amounts are minor compared to the amount of Sn added. The amount of soluble Sn was less than 0.5% of the amount added in all samples.

**Table 3-2. Cr Decontamination Factors for Bench Scale Experiment**

	<b>Cr DF, original sample</b>	<b>Cr DF, post-filtered at 24 h</b>
<b>5 min.</b>	493 (98.6)	380 (76.1)
<b>15 min.</b>	373 (74.5)	350 (70.1)
<b>30 min.</b>	349 (69.8)	406 (81.3)
<b>60 min.</b>	265 (53.1)	235 (47.1)

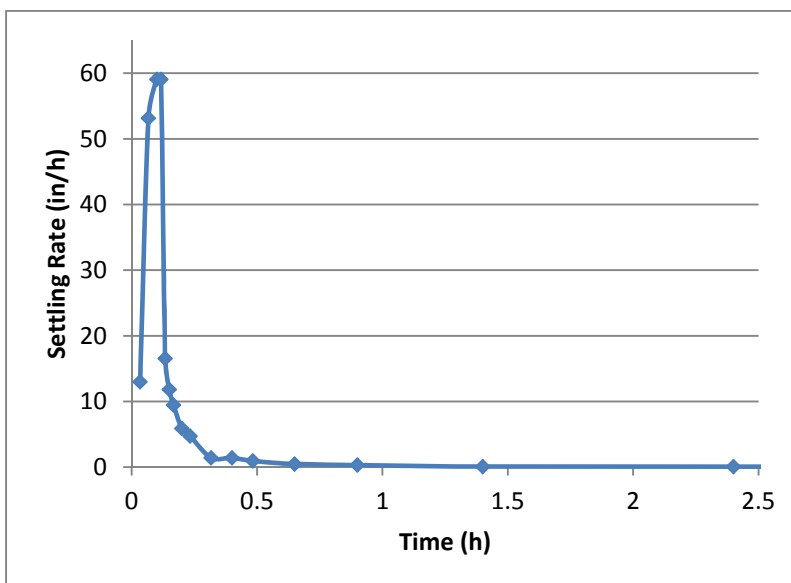
Value in parentheses reflects the reported one sigma uncertainty in the ICP-MS measurement.

### 3.3 Settling Experiments

At the conclusion of the bench scale mixing experiment, the mixer was stopped and the solids were allowed to settle. The height of the suspended solids was recorded periodically, every 1-2 minutes initially, then becoming less frequent with time. This detailed data can be found in Appendix A. The measurements were then used to calculate the solids settling rates reported in Table 3-3. As can be seen from the data in Table 3-3, as well as Figure 3-2, the settling rate peaked within the first few minutes at near 60 inches per hour and then quickly dropped off, presumably due to transitioning from free settling to hindered settling and compaction within the first hour.

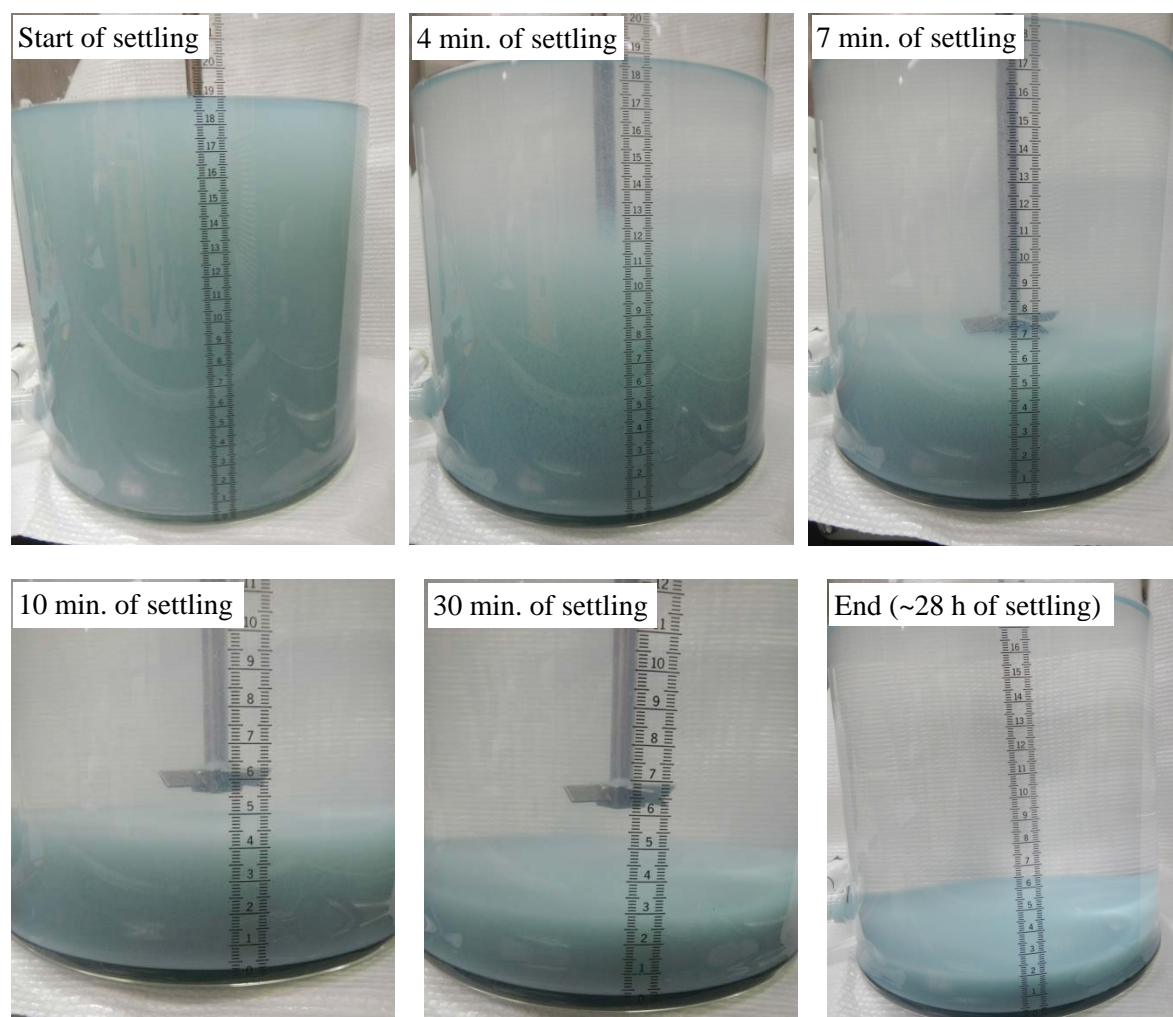
**Table 3-3. Settling Rates**

<b>Elapsed Time (h)</b>	<b>Settling Rate (in/h)</b>
0.03	12.99
0.07	53.15
0.10	59.06
0.12	59.06
0.13	16.54
0.15	11.81
0.17	9.45
0.20	5.91
0.23	4.72
0.32	1.42
0.40	1.42
0.48	0.94
0.65	0.47
0.90	0.31
1.40	0.08
2.40	0.08
5.77	0.01
28.57	0.002



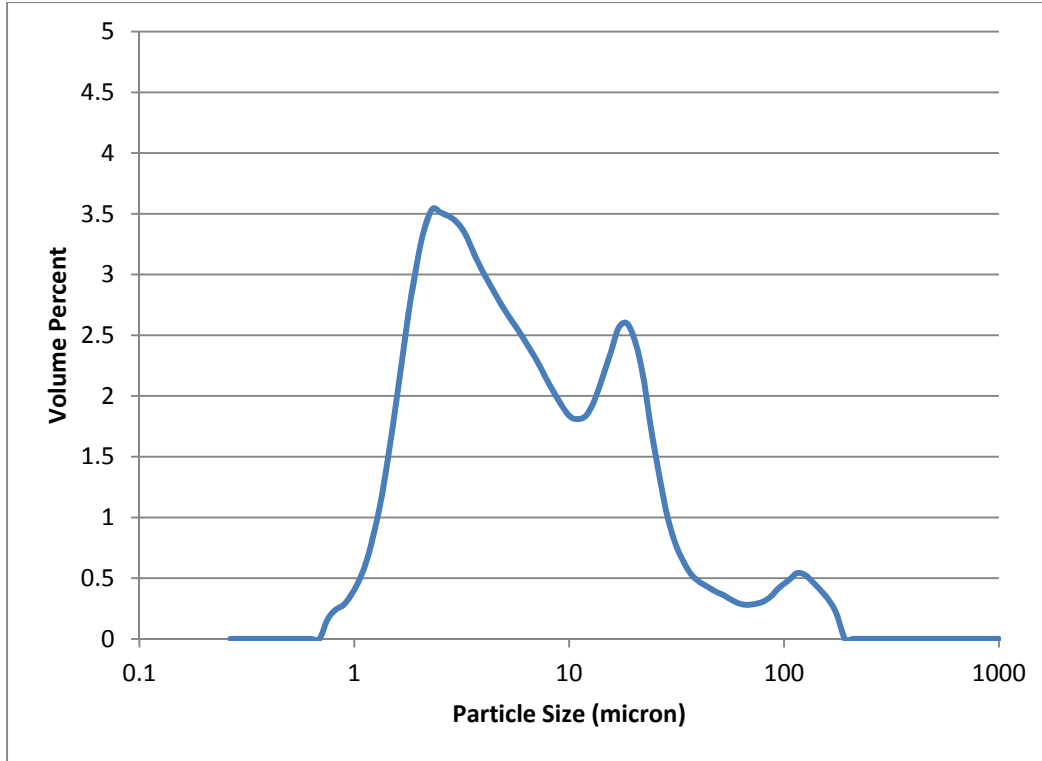
**Figure 3-2. Settling rate versus time.**

Figure 3-3 shows a series of photographs taken during the solids settling experiment. As can be seen from the photographs, the settling is essentially complete within approximately 30 minutes. The level at the top of the settled solids only changed 0.9 cm between the 30 minute time point and the end of the experiment at 28 hours.



**Figure 3-3. Photographs taken at various time points during solids settling.**

At the conclusion of the settling tests, the majority of the supernatant was drained via the port located near the bottom of the vessel, but above the settled solids. The concentrated solids slurry was then transferred to a separate bottle. After mixing, a sample of the slurry was taken from this bottle and was submitted for particle size analysis. The results showed a fairly broad distribution of particles ranging in size from 0.750 to 176 microns or 1.73 microns at the 10<sup>th</sup> percentile and 55.5 microns at the 95<sup>th</sup> percentile. The mean diameter of the volume distribution was 13.5 microns, and the mean diameter of the number distribution was 1.58 microns. The median particle size based on volume distribution was 5.14 microns. The volume distribution is shown in Figure 3-4.



**Figure 3-4. Particle size distribution (volume) of precipitate from stannous chloride reductive precipitation reaction.**

### 3.4 Sample Calculation for Full-Scale Reaction Vessel

This section provides an example calculation to design a full-scale vessel to perform the precipitation reaction. The design flow rate is 555 gallons per hour (9.25 gpm).<sup>23</sup> Assuming 70% attainment, the precipitation process needs to run at 13.2 gpm. At bench-scale, the precipitation process occurred within 5 minutes. A precipitation reaction time of 50 minutes is assumed for full-scale. In order to have one vessel turnover in 50 minutes, the reactor working volume should be 660 gallons. To have three vessel turnovers, the reactor working volume should be 1980 gallons (~2,000 gallons), and is used as a conservative upper bound on the reactor size needed for this process in WTP. Assuming the vessel diameter is the same as the liquid height, the vessel diameter can be calculated with equation [4]

$$T = \sqrt[3]{\frac{4V}{\pi}} = \sqrt[3]{\frac{4(2,000 \text{ gallons})(3785 \frac{\text{cm}^3}{\text{gallon}})}{\pi}} = 213 \text{ cm} = 7 \text{ ft} \quad [4]$$

where T is the vessel diameter and V is the vessel working volume. Solving equation [1] for the full-scale vessel, assuming the impeller is a 4 blade pitch blade impeller with a diameter 1/4 of the vessel diameter

$$N\theta_{95} = 5.2 \frac{T^{1.5}H^{0.5}}{D^2N_p^{1/3}} = 5.2 \frac{(7)^{1.5}(7)^{0.5}}{(1.75)^2(1.27)^{1/3}} = 77 \quad [1]$$

the calculated value of  $N\theta_{95}$  is 77. Selecting an impeller speed of 60 rpm (1 rps), the 95% blend time is 77 seconds. Using equation [2], the 99% blend time is 118 seconds, approximately 2 minutes, which is much less than the turnover time of the vessel and the time observed for the precipitation reaction in the bench-scale testing.

This calculation gives an example of a vessel design that will work to ensure the precipitation reaction to remove technetium is complete. The design of the full-scale system needs to consider feed addition and product removal to ensure the process operates as designed. In addition, other impeller types, sizes and speeds could achieve the desired mixing needed. If larger scale testing shows that the reaction time is much less than 50 minutes, the vessel size could be reduced.

### 3.5 Solid-Liquid Separation

Once the precipitation reaction is complete, the solid particles must be separated from the decontaminated supernate. Possible technologies to perform the solid-liquid separation are clarifiers, hydrocyclones, and filters. Although other technologies also exist (e.g. centrifuges), these three are expected to be the methods with the lowest maintenance requirements, which is key for radioactive facilities.

Clarifiers are settling tanks built with mechanical means to remove the solids deposited by settling. The solid particles settle by gravity and are removed from the tank bottom by a device, such as a scraper or a conveyor belt. Clarifiers work best when separating large, dense (i.e., fast settling) particles from liquids. Flocculants are often added to the feed slurry to increase particle size and settling rate. The hold-up time in the clarifier must be long enough for the solid particles to settle to the bottom. Inlet and outlet velocities to the tank must be minimized to prevent turbulence, which would reduce the settling rate. The advantage of clarifiers is their simplicity. The disadvantage is that with slow settling particles the required tanks can be very large, and they often require flocculants, which are not compatible with downstream processes, although the rapid settling observed in this preliminary test suggest that these particles settle quickly and so would not have these disadvantages.

A hydrocyclone is a static device that applies centrifugal force to a flowing liquid mixture to promote the separation of solid particles from a liquid. A hydrocyclone converts incoming liquid velocity into rotary motion by directing inlet flow tangentially near the top of the vessel. The tangential flow creates centrifugal force in the liquid. Heavy components move outward toward the wall of the vessel where they agglomerate and spiral down the wall to the outlet at the bottom of the vessel. Light components (i.e., the liquid or small, slow settling particles) move toward the center axis of the hydrocyclone where they move up toward the overflow at the top of the vessel. Hydrocyclones work well in separating large, dense (i.e., fast settling) particles from liquids. The advantages of hydrocyclones are the small size and the lack of moving parts. The disadvantage is that they will not separate small, light (i.e., slow settling) particles from liquids.

Filters separate solids from liquid with a semi-permeable barrier. The barrier contains pores which allow liquids and dissolved solids to pass, but which block insoluble solids that are larger than the pores. The filter could be dead-end or crossflow. The advantages of filters are that they can remove small, slow settling particles with high efficiency. The filter pore size can be selected to remove the expected particles in the feed. A filter system would be smaller than a clarifier. The disadvantages of a filter are that it will foul from the particles present in the feed and it will require a method such as chemical cleaning, backpulsing, and/or replacement to recover from the fouling.

These three solid-liquid separation technologies have not been tested or fully evaluated at this phase of the program. Further work is needed to determine the best path forward for separating the Tc-containing slurry from the bulk of the liquid. To ensure the required DF is achieved by removing all of the precipitated Tc, the system would likely include a polishing filter as a second step. This would be especially important for the clarifier or hydrocyclone technologies, as these systems can allow bypass of particles.

## 4.0 Conclusions

Previous work has shown  $\text{SnCl}_2$  to be an effective precipitation agent for the  $^{99}\text{Tc}$  through reductive precipitation in the SBS/WESP simulant when added at a ratio of 1.5 equivalents relative to the electrons needed to reduce the  $^{99}\text{Tc}$  and Cr. This is believed to work by reducing the Tc(VII) ion in the soluble pertechnetate ( $\text{TcO}_4^-$ ) to Tc(IV), precipitating as technetium dioxide ( $\text{TcO}_2$ ). A similar reaction is expected for the Cr, reducing from the Cr(VI) ion in the soluble chromate ( $\text{CrO}_4^{2-}$ ) to Cr(III), precipitating as chromium oxide ( $\text{Cr}_2\text{O}_3$ ). All prior testing had been done at small scale, typically ~20 mL, with mixing provided by a shaker oven. Experiments described in this report were aimed at maturing the technology by performing the reaction at a larger scale (5 L versus 20 mL) and with an impeller-mixed vessel rather than a laboratory-scale shaker. Since the previous work has shown that the Cr reaction kinetics bound the Tc reaction kinetics, this experiment was performed as a non-radioactive experiment and the Tc was omitted. The larger scale mixing experiment confirmed that the reaction is complete within 5 minutes with good mixing provided by the pitch blade impeller under these conditions. These results indicate that the precipitation process can be conducted in a short time, if the process is designed to have adequate miscible liquid blending to contact the stannous chloride with the pertechnetate. The key is to design the process so the miscible liquid blend time is only a few minutes and to ensure that any layer of solid particles on the vessel volume is thin to allow the stannous chloride and pertechnetate to contact each other in a short time. The method and time of chemical addition needs to be considered when designing the full-scale process, since chemical addition can affect fluid mixing.

As a preliminary look at what solid-liquid separation technologies would be needed to implement this process, the settling rate of the precipitate formed was measured. Since Cr is the dominant species that reacts and precipitates in solution, the physical properties of the Cr and Sn precipitate will dominate the rheological properties and the solid-liquid separation behavior. While the Tc precipitate could theoretically behave differently, i.e., stay suspended rather than settle, in practice the Tc solids would likely co-precipitate or sorb onto the bulk precipitate and not remain in suspension. Since the objective was not to determine the DF, Tc was not included in these experiments. This test was designed to provide information to help with the design of the solid-liquid separation portion of the flowsheet. The precipitate was found to settle relatively quickly, and indicates a settle and decant process with a polishing filter may be sufficient for separating the precipitate from this stream. Again, the fast settling time indicates that this process could be deployed in relatively small equipment. The results indicate that routine engineering-scale clarifiers or hydrocyclones followed by polishing filters could be used for this process. However, the particle size and morphology will also be impacted by factors such as mixing and stannous chloride addition rate as well as factors such as flow rate, temperature, and geometry at full scale. The particle size obtained under these bench scale conditions may not necessarily represent the particle size and morphology from a full scale process.

## 5.0 Future Work

Additional tasks needed to further develop this process include slurry rheology measurements, solid-liquid separation, corrosion and erosion studies, and slurry storage and immobilization.

## 6.0 References

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## Appendix A. Detailed Results

**Table A-1. Summary of Concentration Data from Bench Scale Mixing Experiment (HLAW-221)**

Sample Time	Cr Concentration (µg/L)		Sn Concentration (µg/L)	
	Original Sample	Re-filtered Sample	Original Sample	Re-filtered Sample
<b>5 min.</b>	168	218	1440	1020
<b>15 min.</b>	222	236	1480	674
<b>30 min.</b>	237	204	1490	505
<b>60 min.</b>	312	351	1590	686

**Table A-2. Solids Settling Raw Data**

Date/Time	Level at top of suspended solids (cm)
4/10/2017 10:36	19.1
4/10/2017 10:38	18
4/10/2017 10:40	13.5
4/10/2017 10:42	8.5
4/10/2017 10:43	6
4/10/2017 10:44	5.3
4/10/2017 10:45	4.8
4/10/2017 10:46	4.4
4/10/2017 10:48	3.9
4/10/2017 10:50	3.5
4/10/2017 10:55	3.2
4/10/2017 11:00	2.9
4/10/2017 11:05	2.7
4/10/2017 11:15	2.5
4/10/2017 11:30	2.3
4/10/2017 12:00	2.2
4/10/2017 13:00	2
4/10/2017 14:06	2
4/10/2017 16:22	1.9
4/11/2017 15:10	1.8