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Laboratory Optimization Tests of Decontamination of Cs, Sr, and Actinides from Hanford Waste Treatment Plant Low Activity Waste Off-Gas Condensate Simulant

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EXECUTIVE SUMMARY

The Hanford Waste Treatment and Immobilization Plant (WTP) Low Activity Waste (LAW) vitrification facility will generate an aqueous condensate recycle stream (LAW Off-Gas Condensate) from the off-gas system. The baseline plan for disposition of this stream 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. Alternate disposition of this stream would eliminate recycling of problematic components, and would enable less integrated operation of the LAW melter and the Pretreatment Facilities. Eliminating this stream from recycling within WTP would also substantially decrease the LAW vitrification mission duration and quantity of glass waste.

This LAW Off-Gas Condensate stream contains components that are volatile at melter temperatures and are problematic for the glass waste form. Because this stream recycles within WTP, these components accumulate in the Condensate stream, exacerbating their impact on the number of LAW glass containers that must be produced. The current construction of WTP includes two LAW melters, and additional melters will be needed as Supplemental capacity to complete the mission. Approximately 32% of the sodium in Supplemental LAW comes from glass formers used to make the extra glass to dilute the halides to acceptable concentrations in the LAW glass, and diverting the stream reduces the halides in the recycled Condensate and is a key outcome of this work. Additionally, under possible scenarios where the LAW vitrification facility commences operation prior to the WTP Pretreatment facility (a.k.a. Direct Feed LAW), identifying a disposition path becomes vitally important. This task seeks to examine the potential treatment of this stream to remove radionuclides and subsequently disposition the decontaminated stream elsewhere, such as the Effluent Treatment Facility (ETF), for example. The treatment process envisioned is very similar to that used for the Actinide Removal Process (ARP) that has been operating for years at the Savannah River Site (SRS), and focuses on using mature radionuclide removal technologies that are also compatible with long-term tank storage and immobilization methods. For this new application, testing is needed to demonstrate acceptable treatment sorbents and precipitating agents and measure decontamination factors for additional radionuclides in this unique waste stream.

Testing reported here includes results from two different simulants. The *baseline simulant* is based on the computer modeling of the entire WTP mission processing 177 waste tanks. The Direct Feed LAW (DFLAW) simulant is based on modeling of only the first several years of operation of the LAW melters prior to start-up of the entire WTP process. The waste feed composition to the LAW melters during DFLAW will be different from the baseline because the waste originates in fewer tanks, and, the internal streams generated during processing within the WTP Pretreatment Facility such as sludge washing and leaching will not be included. Since the waste feed is different, the off-gas condensate will be different.

The origin of this LAW Off-Gas Condensate stream will be the liquids from the Submerged Bed Scrubber (SBS) and the Wet Electrostatic Precipitator (WESP) from the LAW melter off-gas system. The 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 components are expected to be

mostly sodium and ammonium salts of nitrate, chloride, and fluoride. This stream has not been generated yet and will not be available until the WTP begins operation, but a simulant has been produced based on models, calculations, and comparison with pilot-scale tests.

Several radionuclides are expected to be in appreciable concentration in the LAW Off-Gas Condensate, including ^{129}I, ^{90}Sr, ^{137}Cs, and ^{241}Am. This report discusses results of optimized test conditions for radionuclide decontamination of the simulant. Testing examined use of Monosodium Titanate (MST) to remove ^{90}Sr and actinides, and zeolites for ^{137}Cs.

Prior work had shown that removal of the cesium was more effective at alkaline pH because the ammonium ion interferes with the zeolite uptake of ^{137}Cs. A Cs decontamination factor (DF) of 17.9 was obtained with IE-95 at pH 12 in the prior work. The IE-911 (crystalline silicotitanate, CST) had not been tested at alkaline pH, and it was hypothesized that this material may give superior DFs to the IE-95 under alkaline conditions. Similarly, Monosodium Titanate (MST) was more effective at alkaline pH at removing Sr, Pu, and U, with DF values of 319, 11.6, and 10.5, respectively, after 24 hours of contact, at a phase ratio of 5000:1.

Based on the results of the previous testing, all separations were performed at alkaline pH. The target pH was raised slightly from the previous testing (i.e., 12.3 vs. 12.0) to convert additional ammonium to ammonia to improve Cs removal. Results of the current separation testing indicate that sorption/precipitation is a viable concept and has the potential to decontaminate the off-gas stream. The target DFs for all radionuclides were achieved with one or more of the materials tested. Based on the results, a possible treatment scenario could involve the use of Ionsiv® IE-911 to remove the Cs, Sr, and actinides. Addition of MST to remove Sr and actinides may not be needed.

Further testing is needed to identify the larger scale conditions for the decontamination process. Once these conditions are established, follow-on tasks likely include evaluation and testing of applicable solid-liquid separation technologies, slurry rheology measurements, composition variability testing and evaluations, corrosion and erosion testing, slurry storage and immobilization investigations, and decontaminated LAW Off-Gas Condensate evaporation and solidification.

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LIST OF ACRONYMNS and ABBREVIATIONS

Avg	Average
CST	Crystalline Silicotitanate
DF	Decontamination Factor
dpm	Disintegrations per minute
ETF	Effluent Treatment Facility
g	grams
h	hour
HLW	High Level Waste
HTWOS	Hanford Tank Waste Operations Simulator
ICP-ES	Inductively Coupled Plasma – Emission Spectroscopy
IDF	Integrated Disposal Facility
L	Liter
LAW	Low Activity Waste
LERF	Liquid Effluent Retention Facility
mg	milligram
mL	milliliter
MST	Monosodium titanate
NM	Not Measured
PA	Performance Assessment
SALDS	State-Approved Land Disposal Site
SBS	Submerged Bed Scrubber
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SWRT	Secondary Waste Receiver Tanks
TIC	Total Inorganic Carbon
µg	micrograms
µL	microliter
µm	micrometer (micron)
WESP	Wet Electrostatic Precipitator
WG	Weapons Grade
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. This stream, which will contain substantial amounts of chloride, fluoride, ammonium, and sulfate ions, will get recycled within the WTP process by returning 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 at some point within the process. The halide and sulfate components are only marginally soluble in glass, and can dictate the waste loading and thereby impact waste glass volume. Additionally, long-lived ^{99}Tc and ^{129}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}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) [Mann, 2003]. 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. The modeling calculations indicate that diverting this stream decreases the quantity of LAW glass canisters by over 43,000 and ends the mission nearly three years earlier [Arakali, 2012]. Most of the reduction is during the "Supplemental LAW" period, i.e., after additional melters are constructed to supplement the two that are currently under construction.

An alternate operating scenario is also under evaluation where tank supernate is decontaminated in another facility, called LAW Pretreatment System, and sent directly to the LAW melter (a.k.a. Direct Feed LAW or DFLAW) without start-up of the Pretreatment Facility. This scenario would require alternate disposal of the Off-Gas Condensate stream because the Pretreatment Facility would not be available. Further, the DFLAW feed stream to the melter is likely to have a different chemical and radionuclide composition from the full WTP baseline operations feed stream, and thus will have a different Off-Gas Condensate composition.

The objective of this development task is to evaluate decontamination of the Off-Gas Condensate stream in both scenarios (full WTP baseline operations and DFLAW) using sorbents and precipitating agents so that they can be diverted elsewhere (Figure 2). Although Figure 2 shows the baseline scenario where the stream is routed to the Pretreatment facility, the diversion point is the same for the DFLAW scenario. The process would be comparable to the ARP[1] at SRS that has been operating successfully for years, although that process treats tank waste rather than off-gas condensate. The concept for this process adapts the use of technically mature absorbents where feasible, such as Monosodium Titanate (MST), commercially available zeolites previously used in radioactive DOE applications, and common industrial chemicals. Use of these inorganic materials is expected to simplify down-stream issues, such as storage and immobilization. Implementation would make available 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 the Figure indicates sending the decontaminated

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[1] The Actinide Removal Process (ARP) at SRS decontaminates ^{90}Sr and actinides from aqueous tank waste before it is further treated for ^{137}Cs removal by solvent extraction. 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}Sr and actinides is washed with water, and sent for vitrification as HLW glass in the DWPF.

liquid to the ETF, other paths may also be identified. The ETF is used here as an example of a potential path that is used for an estimation of decontamination requirements. The objective of this task is to evaluate removing the Cs, Sr, and actinides from this stream so that it could be disposed at another facility, such as the ETF. A companion task is being performed to examine removal of ^{99}Tc from this stream [Taylor-Pashow, 2014b].

The overall plan for technology development of the options for disposal of the off-gas condensate stream has been documented [McCabe, 2013]. Other alternative disposal paths are being investigated, including tank farm storage options.

1.1 Simulant Formulation Basis

Because the Off-Gas Condensate streams are not yet available for characterization, the simulant formulations were based on input from two sources. The projected solution chemistry and radionuclide content were based on version 7.4 of the Hanford Tank Waste Operations Simulator (HTWOS) modeling of the flow sheet [Belsher, 2012] performed by WRPS [SVF-2732 for baseline and SVF-3002, Rev. 1 for DFLAW]. Insoluble solids composition was primarily based on analysis of LAW Off-Gas Condensate obtained from pilot-scale simulant melter testing [Matlack, 2006]. Basing the solution chemistry and radionuclide content on the computer modeling rather than melter testing results extended the range of compositions and allowed evaluation of process conditions for treatment of all tank wastes. This approach also accounts for internal WTP process streams (for the WTP baseline operations stream), making it more comprehensive. However, since the computer model does not account for carryover of solids by physical entrainment, the insoluble solids were based on results from pilot-scale melter off-gas system testing. Those results showed that the insoluble solids were high in iron, indicating that they are largely glass-formers. Therefore, glass formers were added as the insoluble solid phase. After collecting and comparing this information, the major individual components were further assessed by comparison between the measured and computed values, and adjustments were made based on scientific judgment. Further, the composition was evaluated using chemical thermodynamic modeling software to determine potential precipitation of insoluble solids, acid neutralization, and dissolution of glass-former solids. Laboratory testing and analysis dictated the final, actual composition. Potential variability in the waste stream composition was not evaluated yet, but is recommended for later phases of this program. The HTWOS model run scenario selected as the basis for the baseline solution chemistry was full operation of all of the WTP facilities, including Supplemental LAW melters, albeit with diversion of the LAW Off-Gas Condensate streams from the LAW melter facilities. This diversion has the effect of lowering the concentration of volatile problematic species (versus the condition where it is recycled and concentrations escalate), but is more realistic of the condition that would be encountered if the stream is diverted from WTP. For DFLAW, there are no recycling or internal WTP streams.

More detail on the synthesis of the simulants has been documented [Adamson, 2013 and Nash, 2014]. Radionuclides selected for inclusion were based on a comparison to the Liquid Effluent Retention Facility (LERF)/ETF limits [McCabe, 2013]. Radionuclides that exceeded the limits were included, except for ^{129}I and ^{151}Sm. The ^{129}I was excluded because current aqueous separation technologies are expected to be overwhelmed by the high total halide concentrations

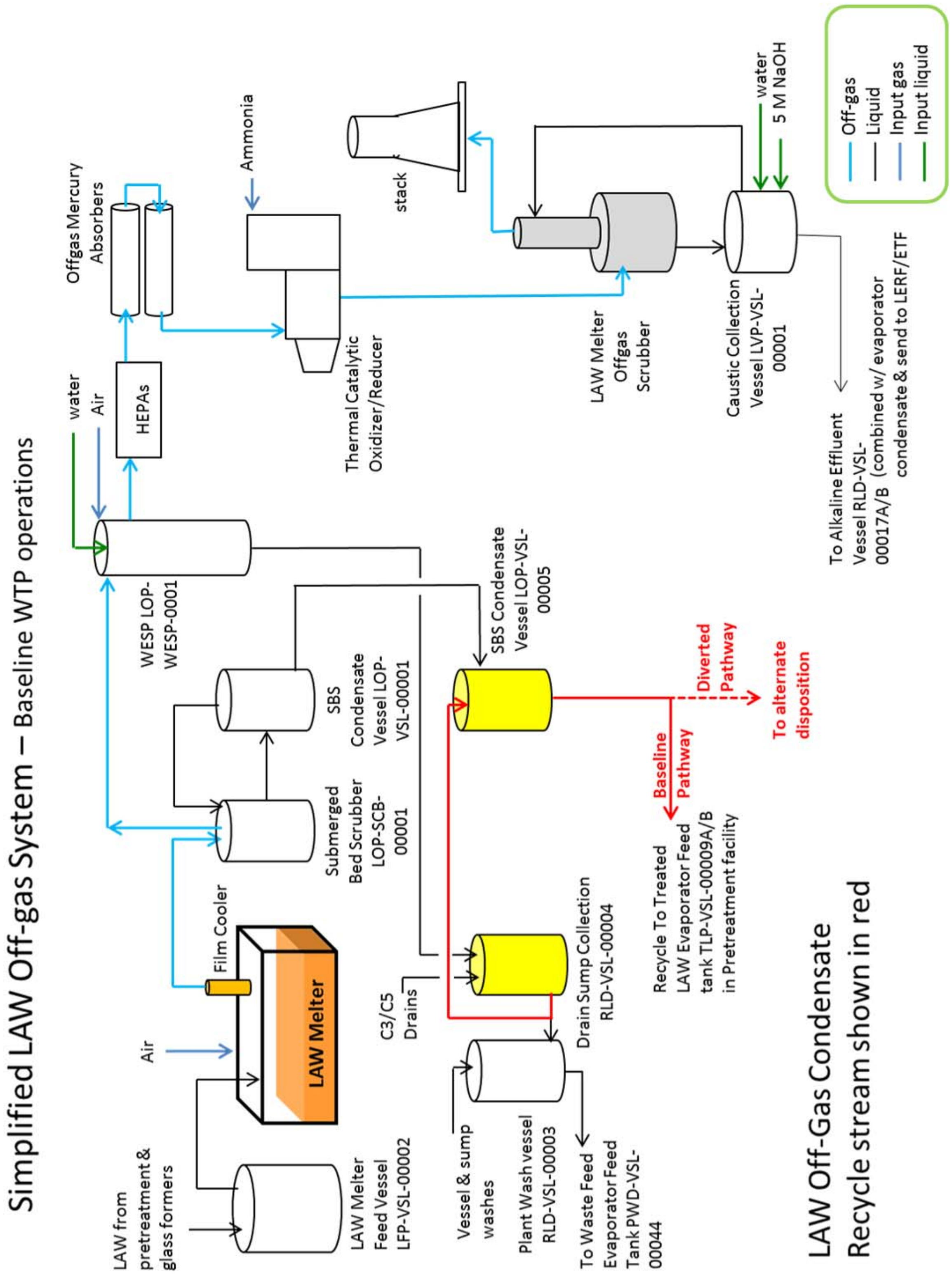

Figure 1 Simplified LAW Off-gas System

(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)

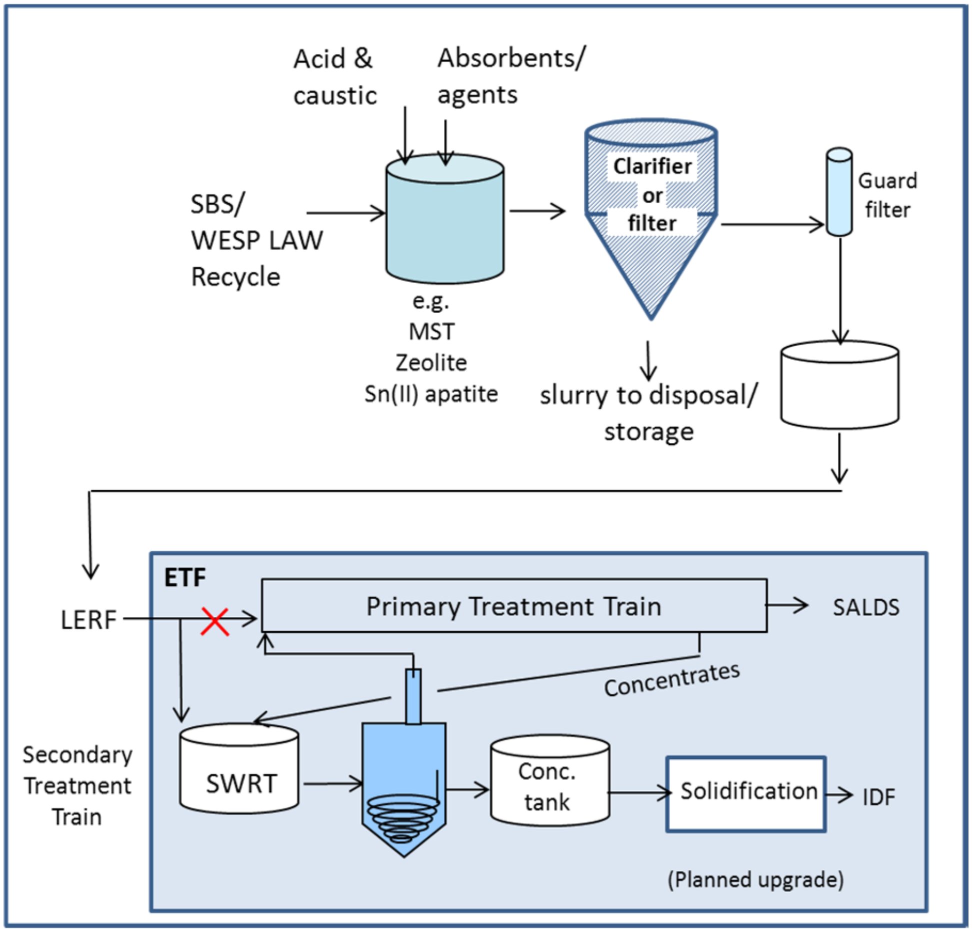

Figure 2 Schematic of a Proposed Decontamination Process and Disposition Path of LAW Off-Gas Condensate

(with I being a minor contributor), and because the ETF is currently equipped to handle some ^{129}I. Furthermore, since one potential disposition path of the contaminated solids generated from the decontamination process is vitrification, the ^{129}I would vaporize again in the melter, so a more comprehensive evaluation of its fate is needed. The ^{151}Sm was excluded because it is evidently a calculation issue in the waste inventory and not expected to actually be present. It is also not appreciably soluble in LAW, based on comparison with SRS waste samples, and would be filtered in High Level Waste (HLW) and not be present in the LAW or LAW Off-Gas Condensate streams.

1.2 Decontamination Process

One processing option that has been previously evaluated is disposal of the LAW Off-Gas Condensate stream directly to the ETF. This option has a number of consequences to ETF including increases in waste volume, halide levels, and radioactivity [Lueck, 2008; May, 2009]. The amount of halide sent to ETF will increase substantially, which is expected to impact corrosion [Lueck, 2008]. Likewise, the radionuclide content would substantially increase, and would challenge existing treatment capabilities [May, 2009]. If the radionuclides are removed from the Condensate stream in an alternate process and the decontaminated liquid is then sent to the ETF (Figure 2), the fluoride, sulfate, and chloride would be purged from the LAW system, yielding substantial benefits to WTP and mitigating the consequences of radioactive contamination at ETF, but still impacting the operation of ETF due to high halide levels.

The LAW Off-Gas Condensate stream is expected to contain ^{99}Tc due to its volatility at melter temperatures. Other radionuclides have either volatile forms (e.g. ^{129}I) or are carried over as particulates from the melter into the off-gas system, or some combination of both mechanisms. The estimated level of activity and soluble salts in the Condensate stream is expected to generally exceed that in the streams (e.g. evaporator overheads and groundwater) currently processed routinely at the ETF [Lueck, 2008].

For this proposed alternative treatment process, separation of the radionuclides is accomplished by precipitation with chemical reagents, or sorption onto pre-formed materials, and settling and/or filtration, similar to the SRS ARP. 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) coupled with absorbents (e.g. hydroxyapatite). For cesium removal, the primary material tested was zeolites. The strontium and actinide removal was examined using the same MST used at SRS ARP.

For this proposed alternative treatment process, disposal of the aqueous decontaminated Condensate stream at ETF is used as an example pathway. To accomplish this, the stream will be routed to the LERF, and transferred into the Secondary Waste Receiver Tanks (SWRT) in the ETF Secondary Treatment Train (similar to *case 2* in [May, 2009]). Ultimately, disposition of the solidified waste in IDF would likely require a PA calculation. In the absence of such a calculation, constituents that are in appreciable quantities will also be removed by the currently available technology to the extent practical.

The target DF for the radionuclides was derived from comparison of the average calculated composition from the modeling versus several acceptance criteria for the Hanford LERF and ETF [McCabe, 2013]. The key radionuclides that exceeded the current limits and their target decontamination factors are shown in Table 1-1. These should all be considered as estimates for the average DF, since they are based on several assumptions of the disposition path and processing steps.

The target DF for ^{137}Cs is based on an estimate of the achievable concentration in the evaporator coupled with the design of the shielding of a future modification planned for immobilizing the concentrate in the ETF. The target DF for ^{99}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 (^{99}Tc removal testing is not included in this report, but was measured in some cases to determine if there was an effect of the treatment). The target DF for ^{90}Sr is 6 because of the sum-of-fractions rule and the total inventory that could be present in ETF. (Note that the isotope used in testing may not be the same isotope that is projected to be in highest concentration in the LAW Off-Gas Condensate stream. For example, ^{85}Sr was used in tests because of its easy gamma analysis versus ^{90}Sr in the stream; similarly,

^{238}U and $^{239/240}$Pu are among the isotopes projected to be present, and all uranium and plutonium isotopes are assumed to have the same DF.)

Table 1-1. Target Decontamination Factors

Isotope	Target DF
^{137}Cs	83
^{238}U	6
$^{239/240}$Pu	2
^{241}Am	166
^{85}Sr	6
^{99}Tc	100

The process is envisioned to be very similar to the equipment used for the SRS ARP, and can utilize designs and lessons learned from that process. The solid-liquid separation equipment may include a clarifier prior to filtration to reduce the burden on the filter and maximize throughput, but that will be determined in a later phase of this program.

The adsorbent/precipitate slurry containing the radionuclides will be characterized in a future phase, and its potential disposition pathways will be evaluated. Immobilization testing will be in a subsequent phase of this program, once the slurry composition and quantities are defined.

2.0 Experimental Procedure

2.1 Baseline Simulant Preparation

(Note: this detail has been previously reported [Taylor-Pashow, 2014b], but is included again here for completeness)

Detail on the basis and synthesis of the simulant has been documented [Adamson, 2013]. The target concentrations of chemicals and radionuclides were derived from the output from the HTWOS calculation, documented in SVF-2732. The aqueous phase was prepared from dissolution of laboratory chemicals, as shown in Table 2-1. A single batch of an optimized formulation was used for this testing, earlier described as “Phase 2” simulant. This formulation reduces the sodium, ammonium, and nitrate concentrations from the previous version and brings it closer to the predicted composition. The change in these three species is not expected to impact the Sr or actinide removal processes, but would impact the Cs removal because of competition from ammonium ion. 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 prepared. Note that the information in Table 2-1 does not necessarily reflect the final composition of the aqueous phase because it is impacted by precipitation and reaction with the glass formers, and with the nitric acid added during pH adjustment. The glass formers were then added, and mixed for five days at ambient temperature of ~ 23 ˚C. The filtrate of the Phase 2 simulant (1.75 L batch) required approximately 12.5 mL of 0.4 M nitric acid and was pH 7.54 after adjustment. This pH adjustment ensures that the simulant is initially at the expected condition so that all species are in the expected ionic form.

Table 2-1. Aqueous Simulant Formulation Targets

Chemical	Formula	Phase 2 Mass (g)/L simulant	Phase 2 Molarity
Aluminum nitrate nonahydrate	$Al(NO_3)_3 \cdot 9H_2O$	0.400	0.0011
Sodium chromate	Na_2CrO_4	0.283	0.0017
Potassium chloride	KCl	0.219	0.0029
Sodium chloride	$NaCl$	1.395	0.0239
Sodium fluoride	NaF	3.209	0.0764
Ammonium nitrate	NH_4NO_3	2.820	0.0352
Sodium nitrate	$NaNO_3$	0	0*
Sodium nitrite	$NaNO_2$	0.016	0.0002
Ammonium sulfate	$(NH_4)_2SO_4$	3.220	0.0244
Dibasic sodium phosphate dihydrate	$Na_2HPO_4 \cdot 2H_2O$	0.040	0.0002

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

The glass formers added to the simulant are shown in Table 2-2. These were derived from the overall mission average quantity [Arakali, 2012]. Sucrose was excluded because it is destroyed in the melter.

Table 2-2. Target Glass Former Quantities

Mineral	Formula	Mass (g)/L simulant
kyanite	Al_2SiO_5	0.745
borax	$Na_2B_4O_7 \cdot 10H_2O$	0.0123
boric acid	H_3BO_3	1.430
wollastonite	$CaSiO_3$	0.772
iron oxide (hematite)	Fe_2O_3	0.430
lithium carbonate	Li_2CO_3	0.392
forsterite olivine	Mg_2SiO_4-Fe_2SiO_4	0.257
sodium carbonate	Na_2CO_3	0.003
silica	SiO_2	2.857
rutile	TiO_2	0.114
zinc oxide	ZnO	0.286
zircon	$ZrSiO_4$	0.372
sucrose	$C_{12}H_{22}O_{11}$	0
	Total	7.67

The solids were then removed from the neutralized solution by filtration with a 0.45-µm Nalgene®[2] filter. A portion of the filtrate was then spiked with the radioisotope tracers. Samples were analyzed for elemental composition by Inductively Coupled Plasma – Emission Spectroscopy (ICP-ES) and for anions and ammonium by Ion Chromatography.

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[2] Nalgene is a registered trademark of the Nalge Company Corporation, Rochester, New York.

2.2 DFLAW Simulant Preparation

(Note: this detail has been previously reported [Nash, 2014], but is included again here for completeness)

The target concentrations of chemicals and radionuclides were derived from the output from the HTWOS calculation, documented in SVF-3002, Rev. 1. The aqueous phase was prepared from dissolution of laboratory chemicals, as shown in Table 2-3. A single batch of 1.0 L of simulant was prepared and used for this testing. 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 prepared. Minor adjustments (5%) were made to some components versus the targets to account for this and to use readily available chemicals, and sodium and nitrate were adjusted by 25%. Note that the information in Table 2-3 does not necessarily reflect the final composition of the aqueous phase because it is impacted by precipitation and reaction with the glass formers, and with the nitric acid added during pH adjustment. The glass formers were then added as shown in Table 2-4, and mixed for four days at ambient temperature of ~ 23 ˚C. After mixing, the solution was adjusted to be within the pH range of 7.0-7.5 with 20.2953 g of 0.18 M nitric acid, mixed for over one hour, and then filtered with a 0.45-µm Nalgene®[3] filter.

Table 2-3. DFLAW Simulant Formulation Targets and Actual

Chemical	Formula	Target Mass (g)/L simulant	Actual Mass (g)/L Added
Aluminum nitrate nonahydrate	$Al(NO_3)_3 \cdot 9H_2O$	0.12	0.1203
Sodium chromate	Na_2CrO_4	0.093	0.0934
Potassium chloride	KCl	0.456	0.4563
Sodium chloride	$NaCl$	2.05	2.0506
Sodium fluoride	NaF	1.23	1.2302
Ammonium nitrate	NH_4NO_3	3.05	3.0520
Sodium nitrate	$NaNO_3$	4.6	4.6031
Sodium nitrite	$NaNO_2$	0.0094	0.0097
Ammonium sulfate	$(NH_4)_2SO_4$	1.8	1.8021
Dibasic sodium phosphate dihydrate	$Na_2HPO_4 \cdot 2H_2O$	0.086	0.0864

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[3] Nalgene is a registered trademark of the Nalge Company Corporation, Rochester, New York.

Table 2-4. Target Glass Former Quantities for DFLAW Simulant

Mineral	Formula	Target Mass (g)/L simulant	Actual Mass (g)/L simulant
kyanite	Al_2SiO_5	0.745	0.7453
borax	$Na_2B_4O_7 \cdot 10H_2O$	0.0123	0.0125
boric acid	H_3BO_3	1.430	1.4300
wollastonite	$CaSiO_3$	0.772	0.7727
iron oxide (hematite)	Fe_2O_3	0.430	0.4306
lithium carbonate	Li_2CO_3	0.392	0.3920
forsterite olivine	Mg_2SiO_4-Fe_2SiO_4	0.257	0.2576
sodium carbonate	Na_2CO_3	0.003	0.0031
silica	SiO_2	2.857	2.8570
rutile	TiO_2	0.114	0.1138
zinc oxide	ZnO	0.286	0.2866
zircon	$ZrSiO_4$	0.372	0.3723
sucrose	$C_{12}H_{22}O_{11}$	0	0
	Total	7.67	7.6741

2.3 Simulant Spiking with Radionuclides

Two separate aliquots of the baseline simulant were spiked with the radiotracer solutions shown in Table 2-5. The 1.0 L of DFLAW simulant was also spiked with radiotracer solutions as shown in Table 2-6. The ^{239}Pu and ^{241}Am target concentrations were increased over the calculated amount in an effort to raise the concentration high enough to be well above the analytical detection limit.

Table 2-5. Radiotracer Solutions added to Baseline Simulant

Isotope	Matrix	Target concentration (dpm/mL)
^{137}Cs	^{137}Cs in 0.1 M HCl	1.16E4
^{238}U	$UO_2(NO_3)_2 \cdot 6H_2O$ solid	6.24E-1
$^{239/240}$Pu	1.5 g/L WG Pu in 0.45 M HNO_3	8.42E1
^{85}Sr	^{85}Sr radionuclide in 0.5 M HCl	5.79E4
^{99}Tc	Ammonium pertechnetate solution	9.21E4
^{241}Am	^{241}Am aqueous stock solution	5.15E2

Table 2-6. Radiotracer Solutions added to DFLAW Simulant

Isotope	Matrix	Target concentration (dpm/mL)
^{137}Cs	^{137}Cs in 0.1 M HCl	1.12E4
^{238}U	$UO_2(NO_3)_2 \cdot 6H_2O$ solid	8.59E-2
$^{239/240}$Pu	1.5 g/L WG Pu in 0.45 M HNO_3	8.42E1
^{85}Sr	^{85}Sr radionuclide in 0.5 M HCl	2.58E3
^{99}Tc	Ammonium pertechnetate solution	2.29E5
^{241}Am	^{241}Am aqueous stock solution	5.15E2

After stirring overnight, the simulants were filtered with a 0.45-µm Nalgene® filter. The filtrate was then analyzed for radionuclide content. Analysis methods utilized Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS) (^{99}Tc, ^{238}U), gamma spectroscopy (^{85}Sr, ^{137}Cs, ^{241}Am), and alpha pulse height analysis after an extraction with thenoyltrifluoroacetone ($^{239/240}$Pu). Non-radioactive constituents were analyzed as described above. Results are summarized in Section 3 and detailed in Appendix B.

2.4 Sorption/Precipitation Tests

These tests were performed after pH adjustment of the simulant. For the initial tests, a 375 mL aliquot of the baseline simulant was adjusted to pH ~12.3 using 5.216 g of 50 wt% sodium hydroxide solution. For the optimized tests, a 175 mL aliquot of the baseline simulant was adjusted to pH ~12.3 with 2.528 g of 50 wt% sodium hydroxide solution. For the DFLAW tests, a 215 mL aliquot of the DFLAW simulant was adjusted to pH ~12.3 with 4.909 g of 50 wt% sodium hydroxide solution. No obvious precipitation was observed following pH adjustment and the simulants were not filtered prior to contact with the sorbents/reagents, but a control sample was filtered and analyzed along with each test sample to examine the effect of the pH adjustment without any sorbents/reagents. In general, tests were performed by adding a small amount of each sorbent/reagent to separate poly bottles, followed by addition of 20 mL of the radioactive simulant solution to each. The bottles were then agitated in a shaker oven at ~25 ˚C for the specified time. Each sample was then filtered through a 0.1-µm filter. The filtrate was then analyzed for the radionuclide of interest. Results are summarized in Section 3.

Table 2-7. Sorbent/Reagent Test Matrix for Initial Tests (Baseline Simulant)

Sorbent/Reagent	**Mass Phase ratio (liquid g:solid g)**	**Target Duration (hours)**
pH Adjust ALL to pH = 12.3 +/- 0.2 (0.174 moles NaOH added/L)		
Blank filtered	NA	
Cs Removal		
IE-95	100:1	7, 24, 48 hr
CST (IE-911 beads)	100:1	7, 24, 48 hr
IE-95	500:1	7, 24, 48 hr
CST (IE-911 beads)	500:1	7, 24, 48 hr
Sr/Actinide Removal		
MST	5000:1	7, 24 hr
mMST	5000:1	7, 24 hr
SrTreat	5000:1	7, 24 hr
$Fe_2(oxalate)_3$	100:1	7, 24 hr

Table 2-8. Sorbent/Reagent Test Matrix for Optimized Tests (Baseline Simulant)

Sorbent/Reagent	**Mass Phase ratio (liquid g:solid g)**	**Target Duration (hours)**
pH Adjust ALL to pH = 12.3 +/- 0.2 (0.180 moles NaOH added/L)		
Blank filtered	NA	
Cs Removal		
CST (IE-911 beads)	200:1	6, 24, 48 hr
CST (IE-911 beads)	250:1	6, 24, 48 hr
Sr/Actinide Removal		
MST	2500:1	6, 24 hr

Table 2-9. Sorbent/Reagent Test Matrix for DFLAW Tests

Sorbent/Reagent	**Mass Phase ratio (liquid g:solid g)**	**Target Duration (hours)**
pH Adjust ALL to pH = 12.3 +/- 0.2 (0.285 moles NaOH added/L)		
Blank filtered	NA	
Cs Removal		
CST (IE-911 beads)	100:1	6, 24 hr
CST (IE-911 beads)	200:1	6, 24 hr
Sr/Actinide Removal		
MST	5000:1	6, 24 hr
MST	2500:1	6, 24 hr

The sources of the sorbents and precipitation reagents were:

IE-95: UOP Ionsiv® IE-95, 20x50 mesh (commercial zeolite)
CST: Crystalline Silicotitanate, UOP Ionsiv® IE-911, batch # 899902081000009 (not caustic washed) (commercial zeolite)
MST ($NaTi_2O_5 \cdot xH_2O$): 15.6 wt% aqueous slurry, Harrell Industries batch # 46000619120
mMST: modified MST, 17.3 wt% aqueous slurry, SRNL prepared batch # LS-14
SrTreat: Fortum Engineering Ltd., Finland, Lot #8 (grain size < 0.045 mm)
Ferric oxalate: Alfa Aesar, Lot # H20D23

2.5 Quality Assurance

This test program is described in the Task Technical and Quality Assurance Plan for Technetium Ion Exchange Resin Manufacturing Maturation [McCabe, 2014]. Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. Savannah River National Laboratory (SRNL) documents the extent and type of review in WSRC-IM-2002-00011, Rev. 2.

3.0 Results and Discussion

3.1 Baseline Simulant Composition

(Note: results were previously reported [Taylor-Pashow, 2014b], but are included here for completeness)
Results of the average and standard deviation of the duplicate chemical analysis of the neutralized, filtered simulant are shown in Table 3-1. These match the target compositions reasonably well, except for nitrate which was low. This small variation is 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. Although boron, lithium, silicon, and zinc were not in the aqueous simulant preparation, they are present in the solution due to addition of the glass former solids and their subsequent dissolution. The sulfur analysis by ICP-ES indicated higher than expected concentrations, which may be due to matrix effects, but the sulfate analysis by IC indicated it is very close to the target. The target concentration for soluble aluminum was ~28 mg/L, based on computer modeling and comparison to the pilot scale melter off-gas condensate sample analyses [Matlack, 2006]. However, attempts to dissolve the aluminum (added as 0.4 g/L aluminum nitrate nonahydrate) by manipulation of the sequence of chemical addition and temperature were unsuccessful.

Table 3-1. Neutralized Baseline Simulant Filtrate Chemical Composition.

Component	Simulant Concentration (mg/L)	Std. Dev.		HTWOS projection (avg. SVF-2732) (mg/L)
Al	<1.0			28
B	242	8.8		GF[1]
Ca	1.1	0.014		GF[1]
Cr	89.9	0.86		91
Fe	<1.0			GF[1]
K	103	0.54		115
Li	80.9	0.80		GF[1]
Mg	<1.0			GF[1]
Na	2.28E3	14		2.29E3
P	<1.0			7 (as PO_4^{-3})
S	1.26E3	15		780 (as SO_4^{-2})
Si	55.0	0.42		GF[1]
Ti	<1.0			GF[1]
Zn	14.6	0.15		GF[1]
Zr	<1.0			GF[1]
F^-	1.41E3	0*		1.45E3
Cl^-	945	1.4		950
NO_2^-	<500			10.7
NO_3^-	3.20E3	57		5.53E3
SO_4^{-2}	2.23E3	14		2.34E3
PO_4^{-3}	<31 (based on P)			21.5
NH_4^+	1.54E3**			1.51E3

*Standard Deviation of zero indicates the two analysis results were identical
**analysis of a single sample
[1] Glass Former component; minimal HTWOS projected concentration

3.2 DFLAW Simulant Composition

(Note: results were previously reported [Nash, 2014], but are included here for completeness) Chemical analysis results of a sample of the neutralized and filtered simulant is shown in Table 3-2. These match the target compositions reasonably well. Although boron, lithium, silicon, zinc, and carbonate were not in the aqueous simulant preparation, they are present in the solution due to addition of the glass former solids and their subsequent dissolution. It appears that all of the lithium carbonate and boric acid dissolved, but only a small amount of a silicon mineral and a small amount of the zinc oxide dissolved; which is expected due to their relative solubilities. Additionally, the analyzed soluble fluoride in the simulant was about 50 mg/L lower than the target, and the phosphate is much lower than the target. Presumably, both fluoride and phosphate precipitate from the solution. The target concentration for soluble aluminum was 8.7

mg/L, based on computer modeling. However, consistent with prior results, the aluminum is not sufficiently soluble.

Table 3-2. Neutralized DFLAW Simulant Filtrate Chemical Composition

Component	DFLAW Simulant Concentration (mg/L)	Method uncertainty (% RSD)	HTWOS projection Adjusted (avg. SVF-3002, R. 1) (mg/L)
Al	<0.309		8.7
B	243	10	GF[1] (251 mg added as GF)
Ca	3.67	10	GF[1] (266 mg added as GF)
Cr	28.4	10	30
Fe	<0.07		GF[1] (385 mg added as GF)
K	242	10	239
Li	72	10	GF[1] (73.6 mg added as Li_2CO_3)
Mg	1.01		GF[1] (36 mg added as GF)
Na	2.66E3	10	2.78E3
P	4.38	11.8	15 (added as PO_4^{-3})
S	465	10.3	435 (added as SO_4^{-2})
Si	37.9	10	GF[1] (1752 mg added as GF)
Ti	<0.047		GF[1] (68 mg added as GF)
Zn	4.8	10	GF[1] (230 mg added as GF)
Zr	<0.031		GF[1] (185 mg added as GF)
F^-	511	10	559
Cl^-	1.45E3	10	1.46E3
NO_2^-	<10		6.3
NO_3^-	5.90E3	10	5.78E3
SO_4^{-2}	1.29E3	10	1.31E3
PO_4^{-3}	<20		46
CO_3^{-2}	319 (63.4 as TIC)	10	GF[1] (318 mg added as Li_2CO_3)
NH_4^+	1.14E3		1.18E3

[1] Glass Former component; minimal HTWOS projected concentration

3.3 Decontamination Test Results

For each test, a freshly filtered control sample was submitted for analysis alongside the decontaminated test sample to ensure consistency, and those results were used to calculate the DF. Averaged results of the radionuclide analyses on the control samples of the spiked filtered simulant for aliquots 1 and 2 of the baseline simulant are shown in Tables 3-2 and 3-3, respectively, and results for the DFLAW simulant are shown in Table 3-4. Results shown for Tc and U analysis are from ICP-MS as mg/L, and the dpm/mL are calculated from a specific activity of 0.0170 Ci/g and 3.36E-7 Ci/g, respectively.

The ^{241}Am appears to be insoluble in both simulants. Spiking of the simulants with a recently analyzed stock solution of known activity was unsuccessful at achieving a measurable amount of ^{241}Am in either filtered simulant. The $^{239/240}$Pu also appears to have limited solubility, as the target activity could not be achieved. Immediately after spiking, there was more soluble Pu than shown below, and it was observed that there was less soluble Pu present as time progressed, suggesting it was not at equilibrium. Slow precipitation kinetics are frequently observed in solutions that contain Pu and carbonate. The reported DF measurements account for this potential non-equilibrium condition by using the initial Pu concentration measured in that same sample batch that was filtered and analyzed at the same time. In the baseline simulant, the Pu concentration was found to decrease an additional ~60% between testing performed in August 2014 [Taylor-Pashow, 2014b] and September 2014 (this testing). This precipitation of Pu could also be attributed to the pH adjustment performed on this sample of simulant. The control values reported in Tables 3-3 through 3-5 were after pH adjustment to 12.3 unless otherwise indicated. The ^{137}Cs content was higher than the target in the spiking of the first aliquot of the baseline simulant, which appears to be due to a higher than expected concentration in the stock solution, which was also observed in other tests. This was taken into account when spiking the 2nd aliquot of the baseline simulant and when spiking the DFLAW simulant. The ^{85}Sr activity was also higher than the target in the DFLAW simulant, which could be attributed to a dilution error as the stock solution had to be diluted prior to spiking.

Table 3-3. Average Aliquot 1 Baseline Simulant Control Sample Radionuclide Composition

Isotope	**Concentration (dpm/mL)**	**Reported Method Uncertainty**	**% of Target Concentration**
^{137}Cs	2.61E4	5.0%	225
^{238}U	5.65E-1 (0.758 mg/L)	20%	91
$^{239/240}$Pu	8.36E0	32.7%	9.9
^{85}Sr	5.94E4	5.0%	103
^{99}Tc	7.59E4 (2.01 mg/L)	20%	82
^{241}Am	<5.66E1	Mda	<11

mda = minimum detectable activity

Table 3-4. Average Aliquot 2 Baseline Simulant Control Sample Radionuclide Composition

Isotope	**Concentration (dpm/mL)**	**Reported Method Uncertainty**	**% of Target Concentration**
^{137}Cs	1.20E4	5.0%	103
^{238}U	4.43E-1 (0.594 mg/L)	20%	72
$^{239/240}$Pu	5.83E0	33.5%	6.9
^{85}Sr	4.85E4	5.0%	84
^{99}Tc	6.19E4 (1.64 mg/L)	20%	67

Table 3-5. Average DFLAW Simulant Control Sample Radionuclide Composition

Isotope	Concentration (dpm/mL)	Reported Method Uncertainty	% of Target Concentration
^{137}Cs	1.00E4	5.0%	89
^{238}U	9.96E-2 (0.132 mg/L)	20%	116
$^{239/240}$Pu	7.17E0*	42.8%	8.5
^{85}Sr	5.31E3	5.0%	206
^{99}Tc	1.74E5 (4.60 mg/L)	20%	76
^{241}Am	<1.78E1*	mda	<26

*Measured after initial spiking, before pH adjustment to 12.3. Due to low levels not measured during testing.

Previous scoping studies [Taylor-Pashow, 2014a] had shown that increasing the pH from 7.3 to 12 resulted in improvements of the materials tested for Cs, Sr, and actinide removal; therefore, these experiments were all performed with pH adjusted simulant. Results from the first set of sorbent/reagent tests are shown in Tables 3-6 and 3-7. Based on the results from these tests, a second set of conditions were tested, and those results are shown in Table 3-8. Details of the test results can be found in Appendix B. (note that removal of Tc was not an objective of this testing, but results are shown where available)

Table 3-6. Summary of Results of Sorption Tests at pH 12.3 (Baseline, Initial Tests)

Sorbent	IE-95 (100:1)			IE-95 (500:1)			CST (IE-911) (100:1)			CST (IE-911) (500:1)		
Contact Time (h)	7	24	48	7	24	48	7	24	48	7	24	48
Avg. Sr DF	2.06 (0.00)	6.93 (1.03)	14.1 (0.77)	1.36 (0.04)	2.02 (0.18)	3.25 (0.24)	11.7 (2.29)	241 (11.7)	809 (277)	2.61 (0.04)	26.2 (0.47)	243 (15.1)
Avg. Cs DF	5.17 (0.04)	17.3 (1.39)	20.9 (0.93)	2.08 (0.21)	4.09 (0.47)	4.88 (0.06)	13.2 (4.24)	188 (23.2)	321 (31.5)	2.61 (0.08)	20.1 (0.77)	50.6 (1.50)
Avg. Pu DF	n/a[a]	nm	nm	n/a[a]	nm	nm	n/a[a]	3.35 (1.12)	nm	n/a[a]	nm	nm
Avg. U DF	2.03 (0.02)	nm	nm	1.37 (0.07)	nm	nm	2.85 (0.54)	31.4 (7.85)	nm	1.34 (0.01)	nm	nm
Avg. Tc DF	1.01 (0.00)	nm	nm	1.01 (0.01)	nm	nm	1.01 (0.01)	1.00 (0.00)	nm	1.01 (0.00)	nm	nm

a) All Pu-239/240 activities for the 7 h samples, including the controls were below the method detection limit, and therefore DFs could not be calculated.
b) nm = not measured.
c) Values in parentheses are the standard deviation of duplicate trials.
d) Outlined boxes indicate target sorbate(s) for each material. Yellow highlight indicates exceeded target DF from Table 1-1.

Table 3-7. Summary of Results from Sorption Tests at pH 12.3 (Baseline, Initial Tests) continued

Sorbent	**MST (5000:1)**		**mMST (5000:1)**		**SrTreat (5000:1)**		**$Fe_2(oxalate)_3$**	
Contact Time (h)	7	24	7	24	7	24	7	24
Avg. Sr DF	873 (457)	> 1229 (13.7)	> 1578 (16.0)	> 997 (2.90)	199 (81.8)	866 (9.60)	nm	36.9 (3.90)
Avg. Cs DF	1.01 (0.01)	1.02 (0.01)	1.00 (0.01)	1.02 (0.01)	1.00 (0.02)	1.04 (0.02)	nm	1.05 (0.01)
Avg. Pu DF	n/a[a]	> 2.86 (0.10)	n/a[a]	> 6.53[b] (4.39)	n/a[a]	4.37 (1.60)	nm	1.02 (0.02)
Avg. U DF	2.94 (0.09)	2.51 (1.84)	1.24 (0.02)	2.79 (2.20)	1.05 (0.00)	1.07 (0.00)	nm	1.39 (0.00)
Avg. Tc DF	0.99 (0.02)	0.99 (0.00)	0.98 (0.01)	0.99 (0.01)	0.99 (0.00)	1.00 (0.01)	nm	1.00 (0.01)

a) All Pu-239/240 activities for the 7 h samples, including the controls were below the method detection limit, and therefore DFs could not be calculated.
b) One duplicate gave an activity below the detection limit and one did not. The two DFs were >9.63 and 3.43.
c) nm = not measured.
d) Values in parentheses are the standard deviation of duplicate trials.
e) Outlined boxes indicate target sorbate(s) for each material. Yellow highlight indicates exceeded target DF from Table 1-1.

Table 3-8. Summary of Results from Optimized Sorption Tests at pH 12.3 (Optimized Tests)

Sorbent	**CST (IE-911) (200:1)**			**CST (IE-911) (250:1)**			**MST (2500:1)**	
Contact Time (h)	6	24	48	6	24	48	6	24
Avg. Sr DF	5.07 (1.14)	105 (20.9)	435 (73.6)	3.19 (0.11)	63.1 (0.85)	297 (78.0)	> 2220[b] (883)	> 1850[c] (1050)
Avg. Cs DF	4.60 (1.59)	59.4 (9.02)	142 (8.70)	2.67 (0.18)	40.2 (7.05)	118 (0.71)	0.90 (0.01)	0.79 (0.01)
Avg. Pu DF	2.30 (0.14)	n/a[a]	n/a[a]	> 1.88[d] (0.28)	n/a[a]	n/a[a]	> 1.55[e] (0.56)	n/a[a]
Avg. U DF	1.78 (0.20)	7.34 (1.31)	31.0 (1.06)	1.49 (0.04)	4.98 (0.78)	22.8 (0.01)	7.45 (0.27)	14.5 (0.36)
Avg. Tc DF	1.01 (0.00)	0.98 (0.01)	0.99 (0.00)	1.01 (0.00)	0.99 (0.01)	0.99 (0.01)	0.99 (0.01)	0.99 (0.01)

a) All Pu-239/240 activities for the 24 and 48 h samples, including the controls were below the method detection limit, and therefore DFs could not be calculated.
b) One duplicate gave an activity below the detection limit and one did not. The two DFs were >2850 and 1600.
c) One duplicate gave an activity below the detection limit and one did not. The two DFs were >2590 and 1110.
d) One duplicate gave an activity below the detection limit and one did not. The two DFs were 1.68 and > 2.08.
e) One duplicate gave an activity below the detection limit and one did not. The two DFs were 1.95 and > 1.16.

f) Values in parentheses are the standard deviation of duplicate trials.
g) Outlined boxes indicate target sorbate(s) for each material. Yellow highlight indicates exceeded target DF from Table 1-1.

Results from the initial testing showed that CST was much more effective for Cs removal at pH 12.3 than the IE-95. This is in contrast to earlier results at near neutral pH, where IE-95 was more effective [Taylor-Pashow, 2014a]. At a phase ratio of 100:1, the CST exceeded the target DF within 24 hours, and continued to remove Cs through 48 hours. Therefore, for the optimized testing, the amount of CST was reduced. The lower amounts of CST were less effective at removing the Cs; however, even the 250:1 phase ratio was sufficient to reach the target DF after 48 hours. The CST was also quite effective at removing the Sr and actinides. At the 200:1 phase ratio, the target DFs were exceeded for all sorbates (except Cs) by 24 hours.

Of the materials tested to target Sr and actinide removal, MST and mMST performed comparably. Both materials removed essentially all of the Sr and Pu to below the method detection limits; however, neither material reached the target DF for U during the initial tests. In the optimized tests, a higher concentration of MST was tested (2500:1 phase ratio), and the U DF was easily reached by 6 hours. The SrTreat material was also quite effective at Sr and Pu removal, but much less effective for U removal. Finally, the ferric oxalate did remove some Sr, meeting the target DF, but had little effect on the actinides.

Based on the results of the initial testing with the baseline simulant, the conditions to test with the DFLAW simulant were determined. The results from the testing with the DFLAW simulant are shown in Table 3-9. Details are provided in Appendix B.

Table 3-9. Summary of Results from Sorption Tests with DFLAW Simulant at pH 12.3

Sorbent	**CST (IE-911) (100:1)**		**CST (IE-911) (200:1)**		**MST (5000:1)**		**MST (2500:1)**	
Contact Time (h)	6	24	6	24	6	24	6	24
Avg. Sr DF	11.2 (0.13)	245 (76.6)	4.81 (0.92)	337 (92.6)	206 (103)	170 (23.4)	> 347 (82.4)	> 223 (43.3)
Avg. Cs DF	12.0 (1.06)	134 (3.95)	5.39 (1.29)	61.2 (2.64)	0.99 (0.02)	1.00 (0.00)	1.02 (0.01)	1.00 (0.02)
Avg. U DF	3.91 (2.10)	> 13.1 (0.00)	1.82 (0.19)	6.53 (2.06)	2.97 (0.40)	4.65 (0.37)	4.34 (2.39)	12.2 (1.22)
Avg. Tc DF	1.01 (0.00)	1.01 (0.00)	1.01 (0.01)	0.99 (0.00)	1.00 (0.00)	1.00 (0.01)	1.01 (0.01)	0.99 (0.01)

a) Values in parentheses are the standard deviation of duplicate trials.
b) Outlined boxes indicate target sorbate(s) for each material. Yellow highlight indicates exceeded target DF from Table 1-1.

The results from the DFLAW testing were similar to those obtained for the baseline simulant. The CST at 100:1 phase ratio, reached the target DFs for Cs, Sr, and U after 24 hours. The lower amount of CST (200:1 phase ratio) was sufficient to reach the target Sr and U DF, but only reached a Cs DF of 61.2 after 24 hours. It is likely that the target DF of 83 could be reached with longer contact times. The benefits of less sorbent versus shorter contact times would have to be weighed to determine the best conditions. With either scenario, it appears MST may not be necessary, and the CST alone is adequate to remove the Cs, Sr, and actinides, in part due to their limited solubility at this pH.

4.0 Conclusions

Sorption testing with various inorganic sorbents proved successful at achieving the target DF for all sorbates (Cs, Sr, Pu, and U). Of the materials tested, the CST (IE-911) appears the most effective for the suite of radionuclides. At a phase ratio of 100:1, the target DF for all radionuclides can be reached within 24 hours. If the amount of CST is reduced to 250:1, this can be achieved within 48 hours. The MST and SrTreat® materials are also very effective at Sr and Pu removal, but have no affinity for Cs, as expected. At sufficient concentration, MST is also effective at removing U, reaching the target DF within 6 hours.

This work demonstrates that very small amounts of sorbents can be used to decontaminate the stream, removing most of the activity from the large majority of the volume so that it can be managed in a more cost-effective way. The most radioactive portion that must be stored and disposed is concentrated, so that storage and disposal costs are also minimized.

5.0 Future Work

Additional work is needed to examine the Tc removal from the DFLAW simulant, and demonstrate the integrated Tc removal step followed by removal of the Cs, Sr, and actinides. Additional testing should also be performed to further optimize the conditions for Cs, Sr, and actinide removal, looking at the impacts of pH and contact time.

Now that this process has been successfully demonstrated at the laboratory scale, further testing is needed to identify the larger scale conditions for the decontamination process. Once these conditions are established, follow-on tasks likely include evaluation and testing of applicable solid-liquid separation technologies, slurry rheology measurements, composition variability testing and evaluations, corrosion and erosion testing, slurry storage and immobilization investigations, and decontaminated LAW Off-Gas Condensate evaporation and solidification.

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Appendix A.

Sorbent/Reagent Decontamination Test Detail

Below are the details of the results from the Sorbent/Reagent decontamination tests.

Table 6-1. Summary of Concentration Data from Tests 65-73 (Initial Tests).

Test ID	**HLAW-65**	**HLAW-66**	**HLAW-67**	**HLAW-68**	**HLAW-69**	**HLAW-70**	**HLAW-71**	**HLAW-72**	**HLAW-73**
Sorbent	None (control)	IE-95	IE-95	CST	CST	IE-95	IE-95	CST	CST
Sorbent Conc. (g/L)	0.0	9.98	10.01	10.04	10.04	2.04	2.00	2.00	2.00
Contact Time (h)	6.87	6.87	6.88	6.88	6.90	6.92	6.93	6.93	6.95
Sr-85 (dpm/mL)	6.03E+04	2.92E+04	2.93E+04	4.51E+03	5.96E+03	4.54E+04	4.35E+04	2.29E+04	2.33E+04
Cs-137 (dpm/mL)	2.59E+04	5.11E+03	5.06E+03	1.62E+03	2.57E+03	1.36E+04	1.18E+04	1.03E+04	9.88E+03
Pu-239/240 (dpm/mL)	<1.15E+01	<2.53E+00	<5.20E+00	<4.26E+00	<7.15E+00	<3.48E+00	<3.76E+00	<2.95E+00	<2.76E+00
Tc-99 (µg/L)	2.00E+03	1.99E+03	2.00E+03	1.99E+03	2.01E+03	1.97E+03	2.01E+03	2.00E+03	1.99E+03
U-238 (µg/L)	7.58E+02	3.78E+02	3.73E+02	2.36E+02	3.09E+02	5.77E+02	5.38E+02	5.66E+02	5.73E+02
Contact Time (h)	24.00	24.00	24.02	24.02	24.03	24.03	24.07	24.07	24.08
Sr-85 (dpm/mL)	5.62E+04	7.57E+03	9.35E+03	2.33E+02	2.49E+02	3.06E+04	2.70E+04	2.25E+03	2.19E+03
Cs-137 (dpm/mL)	2.59E+04	1.41E+03	1.58E+03	1.26E+02	1.50E+02	6.87E+03	5.83E+03	1.32E+03	1.25E+03
Pu-239/240 (dpm/mL)	3.46E-02	NM	NM	2.09E-02	1.29E-02	NM	NM	NM	NM
Tc-99 (µg/L)	2.03E+03	NM	NM	2.01E+03	2.00E+03	NM	NM	NM	NM
U-238 (µg/L)	7.56E+02	NM	NM	2.04E+01	2.92E+01	NM	NM	NM	NM
Contact Time (h)	48.63	48.65	48.67	48.68	48.70	48.72	48.73	48.75	48.78
Sr-85 (dpm/mL)	6.07E+04	4.15E+03	4.49E+03	6.04E+01	9.90E+01	1.97E+04	1.78E+04	2.61E+02	2.39E+02
Cs-137 (dpm/mL)	2.65E+04	1.23E+03	1.31E+03	7.72E+01	8.87E+01	5.48E+03	5.39E+03	5.13E+02	5.35E+02

Table 6-2. Summary of Concentration Data from Tests 74-82 (Initial Tests).

Test ID	**HLAW-74**	**HLAW-75**	**HLAW-76**	**HLAW-77**	**HLAW-78**	**HLAW-79**	**HLAW-80**	**HLAW-81**	**HLAW-82**
Sorbent	None (control)	MST	MST	mMST	mMST	SrTreat	SrTreat	Fe_2(oxalate)$_3$	Fe_2(oxalate)$_3$
Sorbent Conc. (g/L)	0.0	0.201	0.201	0.200	0.200	0.209	0.219	10.02	10.00
Contact Time (h)	7.12	7.13	7.13	7.15	7.15	7.15	7.18	7.18	7.20
Sr-85 (dpm/mL)	6.01E+04	1.09E+02	5.04E+01	<3.84E+01	<3.79E+01	4.25E+02	2.34E+02	NM	NM
Cs-137 (dpm/mL)	2.67E+04	2.61E+04	2.59E+04	2.61E+04	2.63E+04	2.58E+04	2.66E+04	NM	NM
Pu-239/240 (dpm/mL)	<4.98E+00	<1.22E+00	<3.11E+00	<2.68E+00	<2.74E+00	<3.09E+00	<1.40E+00	NM	NM
Tc-99 (µg/L)	2.02E+03	2.00E+03	2.06E+03	2.06E+03	2.03E+03	2.03E+03	2.03E+03	NM	NM
U-238 (µg/L)	7.67E+02	2.65E+02	2.54E+02	6.07E+02	6.24E+02	7.31E+02	7.26E+02	NM	NM
Contact Time (h)	24.65	24.67	24.67	24.68	24.70	24.70	24.73	24.75	24.77
Sr-85 (dpm/mL)	5.98E+04	<4.68E+01	4.76E+01	<5.80E+01	<5.83E+01	6.65E+01	6.75E+01	1.70E+03	1.46E+03
Cs-137 (dpm/mL)	2.57E+04	2.55E+04	2.53E+04	2.55E+04	2.52E+04	2.46E+04	2.52E+04	2.45E+04	2.47E+04
Pu-239/240 (dpm/mL)	1.13E+01	<3.00E+00	<2.85E+00	8.68E-01	2.44E+00	2.58E+00	1.52E+00	8.32E+00	8.08E+00
Tc-99 (µg/L)	2.00E+03	2.04E+03	2.04E+03	2.05E+03	2.03E+03	2.01E+03	2.03E+03	2.00E+03	2.04E+03
U-238 (µg/L)	7.50E+02	1.97E+02	6.20E+02	1.73E+02	6.08E+02	7.02E+02	7.02E+02	5.42E+02	5.42E+02

NM = not measured

Table 6-3. Summary of Concentration Data from Tests 95-104 (DFLAW).

Test ID	**HLAW-95**	**HLAW-96**	**HLAW-97**	**HLAW-98**	**HLAW-99**	**HLAW-100**	**HLAW-101**	**HLAW-102**	**HLAW-103**	**HLAW-104**
Sorbent	None (control)	None (control)	MST	MST	MST	MST	CST	CST	CST	CST
Sorbent Conc. (g/L)	0.0	0.0	0.20	0.20	0.41	0.40	10.01	10.00	5.02	5.04
Contact Time (h)	5.98	6.00	6.00	6.02	6.02	6.03	6.03	6.05	6.07	6.07
Sr-85 (dpm/mL)	6.46E+03	6.93E+03	5.04E+01	2.40E+01	<1.65E+01	<2.32E+01	5.90E+02	6.00E+02	1.23E+03	1.61E+03
Cs-137 (dpm/mL)	9.75E+03	1.03E+04	1.02E+04	9.98E+03	9.91E+03	9.74E+03	7.86E+02	8.91E+02	1.59E+03	2.24E+03
Tc-99 (μg/L)	4.61E+03	4.65E+03	4.64E+03	4.63E+03	4.60E+03	4.56E+03	4.61E+03	4.60E+03	4.57E+03	4.62E+03
U-238 (μg/L)	1.32E+02	1.35E+02	4.97E+01	4.11E+01	2.22E+01	5.04E+01	2.48E+01	5.52E+01	6.84E+01	7.91E+01
Contact Time (h)	24.10	24.12	24.12	24.13	24.13	24.15	24.13	24.15	24.17	24.17
Sr-85 (dpm/mL)	3.83E+03	4.02E+03	2.10E+01	<2.56E+01	<1.55E+01	<2.04E+01	1.31E+01	2.06E+01	1.45E+01	9.75E+00
Cs-137 (dpm/mL)	9.95E+03	1.01E+04	9.99E+03	1.00E+04	9.90E+03	1.02E+04	7.62E+01	7.31E+01	1.59E+02	1.69E+02
Tc-99 (μg/L)	4.59E+03	4.56E+03	4.60E+03	4.53E+03	4.65E+03	4.62E+03	4.53E+03	4.52E+03	4.61E+03	4.61E+03
U-238 (μg/L)	1.31E+02	1.31E+02	2.99E+01	2.66E+01	<1.00E+01	1.15E+01	<1.00E+01	<1.00E+01	1.64E+01	2.58E+01

Table 6-4. Summary of Concentration Data from Tests 105-112 (Optimized Tests).

Test ID	HLAW-105	HLAW-106	HLAW-107	HLAW-108	HLAW-109	HLAW-110	HLAW-111	HLAW-112
Sorbent	None (control)	None (control)	CST	CST	CST	CST	MST	MST
Sorbent Conc. (g/L)	0.0	0.0	5.00	5.00	4.00	4.04	0.40	0.40
Contact Time (h)	6.02	6.03	6.03	6.05	6.05	6.07	6.07	6.10
Sr-85 (dpm/mL)	4.85E+04	4.80E+04	8.22E+03	1.13E+04	1.55E+04	1.48E+04	<1.70E+01	3.02E+01
Cs-137 (dpm/mL)	1.03E+04	1.36E+04	2.09E+03	3.44E+03	4.70E+03	4.27E+03	1.32E+04	1.34E+04
Pu-239/240 (dpm/mL)	7.72E+00	3.94E+00	2.66E+00	2.44E+00	3.50E+00	<2.83E+00	2.99E+00	<5.09E+00
Tc-99 (μg/L)	1.67E+03	1.64E+03	1.65E+03	1.64E+03	1.64E+03	1.65E+03	1.66E+03	1.68E+03
U-238 (μg/L)	5.94E+02	6.05E+02	3.12E+02	3.67E+02	4.09E+02	3.94E+02	7.85E+01	8.27E+01
Contact Time (h)	24.12	24.13	24.13	24.15	24.17	24.18	24.18	24.20
Sr-85 (dpm/mL)	4.63E+04	4.80E+04	3.92E+02	5.21E+02	7.41E+02	7.55E+02	<1.82E+01	4.26E+01
Cs-137 (dpm/mL)	1.21E+04	8.70E+03	1.58E+02	1.96E+02	2.30E+02	2.95E+02	1.33E+04	1.30E+04
Pu-239/240 (dpm/mL)	<4.44E+00	<4.14E+00	<7.39E+00	<2.82E+00	<1.31E+00	<2.85E+00	2.28E+00	<1.87E+00
Tc-99 (μg/L)	1.63E+03	1.64E+03	1.68E+03	1.66E+03	1.65E+03	1.66E+03	1.65E+03	1.64E+03
U-238 (μg/L)	5.92E+02	5.99E+02	7.20E+01	9.27E+01	1.08E+02	1.35E+02	4.05E+01	4.19E+01
Contact Time (h)	48.05	48.07	48.07	48.08	48.08	48.10	n/a	n/a
Sr-85 (dpm/mL)	4.99E+04	5.00E+04	1.30E+02	1.03E+02	1.42E+02	2.07E+02	NM	NM
Cs-137 (dpm/mL)	1.37E+04	1.37E+04	1.01E+02	9.26E+01	1.17E+02	1.16E+02	NM	NM
Pu-239/240 (dpm/mL)	<3.34E+00	<3.53E+00	<3.62E-01	<9.47E-01	7.88E-01	<3.90E-01	NM	NM
Tc-99 (μg/L)	1.62E+03	1.64E+03	1.66E+03	1.66E+03	1.66E+03	1.64E+03	NM	NM
U-238 (μg/L)	5.89E+02	5.85E+02	1.85E+01	1.94E+01	2.58E+01	2.58E+01	NM	NM

NM = not measured

Distribution:

S. L. Marra, 773-A
F. M. Pennebaker, 773-42A
T. B. Brown, 773-A
E. N. Hoffman, 999-W
D. H. McGuire, 999-W
S. D. Fink, 773-A
C. C. Herman, 773-A
K. M. L. Taylor-Pashow, 773-A
C. A. Nash, 773-42A
C. L. Crawford, 773-42A
D. J. McCabe, 773-42A
W. R. Wilmarth, 773-A
D. T. Herman, 735-11A
A. D. Cozzi, 999-W
D. K. Peeler, 999-W
K. M. Fox, 999-W
H. H. Burns, 773-41A
K. H. Subramanian, WRPS
Records Administration (EDWS)
J. A. Diediker. DOE-ORP
L. Holton, DOE-ORP
W. F. Hamel, DOE-ORP
T. W. Fletcher, DOE-ORP
S. H. Pfaff, DOE-ORP
S. T. Arm, WRPS
P. A. Cavanah, WRPS
T. W. Crawford, WRPS
W. G. Ramsey, WRPS
D. J. Swanberg, WRPS
D. L. Noyes, DOE-ORP
N. P. Machara, DOE-EM
J. A. Poppiti, DOE-EM
D. J. Koutsandreas, DOE-EM
R. A. Gilbert, DOE-ORP
C. J. Winkler, WRPS
R. H. Spires, WRPS
M. R. Thorson, WRPS