PNWD-3189 WTP-RPT-034

# Verification of Process Conditions for Sr/TRU Removal from AN-102/C-104 Waste Blend

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September 2002

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## Verification of Process Conditions for Sr/TRU Removal from AN-102/C-104 Waste Blend

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Test Specification: TSP-W375-01-00003 Test Plan: CHG-TP-41500-019 Test Exceptions: None R&T Focus Area: Pretreatment Test Scoping Statement(s): B-35

By Battelle—Pacific Northwest Division Richland, Washington 99352

#### **COMPLETENESS OF TESTING**

This report describes the results of work and testing specified by Test Specification TSP-W375-01-00003 and Test Plan CHG-TP-41500-019. The work and any associated testing followed the quality assurance requirements outlined in the Test Specification/Plan. The descriptions provided in this test report are an accurate account of both the conduct of the work and the data collected. Test plan results are reported. Also reported are any unusual or anomalous occurrences that are different from expected results. The test results and this report have been reviewed and verified.

Approved:

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#### Summary

The River Protection Project-Waste Treatment Plant baseline for pretreating Envelope C low-activity waste (LAW) at Hanford includes a precipitation step for removing radioactive strontium (Sr-90) and transuranic (TRU) isotopes before the waste is vitrified. The current design basis for the Sr/TRU removal process is the addition of strontium nitrate (0.075M) for isotopic dilution and permanganate (0.05M) for TRU removal at 1M additional sodium hydroxide. Section 5 of the *Research and Technology Plan* (BNI 2002) identifies further research needs, which are illustrated in Figure 5-14, Precipitation Test Matrix.

One need shown in this matrix is optimization of the Sr/TRU precipitation reaction conditions [SOW Ref.: Sec. C.6 Std.2 (a)(3)(ii)(B) and WBS No.: 1.2.10.03 and .05]. The optimization of the Sr/TRU precipitation process and the impact of recycle streams are addressed in Scoping Statement B-35, which is included in Appendix C of the *Research and Technology Plan*. In accordance with Scoping Statement B-35, Test Specification TSP-W375-01-00003, and Test Plan CHG-TP-41500-019, studies were conducted to determine if low levels of reagent provide adequate decontamination conditions for integrated process testing with a mixture of Tank AN-102 waste and high-level waste (HLW) pretreatment streams (filtrate, wash, and leach solution from HLW pretreatment of Tank C-104 wastes) (Hallen et al. 2002). These conditions include determining the minimum amount of strontium and permanganate needed for decontaminating the liquid waste to meet LAW requirements for vitrification.

The success criteria include demonstrating that the treated waste meets Specification 2 of the Bechtel National, Inc. contract (BNI 2001) for removing Sr-90 and TRU elements from the LAW solution; i.e., 20 Ci/m<sup>3</sup> for Sr-90 and 100 nCi/g for TRU. Blending the AN-102 waste with the wash/leach solutions resulted in over 60% dilution in the Sr-90 and Am-241 activity relative to the sodium concentration. As a result of the waste blending, decontamination factors (DFs) of approximately 5 for Sr-90 (80% removal) and 1.6 for TRU (38% removal) are required to meet a target of 50% below the LAW requirements (20 Ci/m<sup>3</sup> and 100 nCi/g, respectively). Since over 90% of the TRU in the AN-102/C-104 blended waste is Am-241, a target DF of 1.6 was established for Am-241.

The objective of the work reported here was to verify that the optimized process conditions would provide adequate Sr/TRU decontamination of the AN-102/C-104 waste blend. For this integrated process verification testing, a blended waste was used that combined samples from Tank AN-102 and sludge washing/leaching solutions from Tank C-104 (Brooks et al. 2000). Mixing the sludge washing/leaching solutions with the AN-102 sample resulted in a waste blend of approximately 3M sodium (Urie et al. 2002). This solution was evaporated to concentrate the sodium to approximately 5M sodium as required for the Sr/TRU removal process (Lumetta et al. 2002). Before integrated process verification tests were conducted on a 1-L batch of blended waste, small-scale experiments were performed to verify process conditions for Sr-90 and TRU removal, specifically, to evaluate if low reagent addition (no additional hydroxide, 0.02M Sr, and 0.02M permanganate) is adequate for Sr/TRU decontamination of the blended waste.

The treatment conditions provided adequate Sr/TRU decontamination of the AN-102/C-104 waste blend. The free hydroxide concentration in the blended waste was adequate for decontamination with added strontium and permanganate. As expected, no, or very little, TRU decontamination occurred without the

addition of permanganate. The integration of sludge wash/leach streams with AN-102 waste had very little impact on meeting the Sr/TRU limits for ILAW. The most significant impact of blending was the reduced levels of Sr-90 and TRU in the feed to the process.

Recommended treatment conditions for integrated process verification testing with the AN-102/C-104 waste blend are as follows: 0.02M strontium, 0.02M permanganate, no additional hydroxide, and treatment/digest at  $25 \pm 5^{\circ}$ C. These conditions are based on results from tests with AN-102 diluted waste (Hallen et al. 2002), and are significantly reduced from the current design basis for Sr/TRU removal conditions (0.075M strontium, 0.05M permanganate, 1M additional hydroxide, and treatment/digest at  $50^{\circ}$ C).

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## 1.0 Introduction

This report summarizes work performed in support of the River Protection Project-Waste Treatment Plant (RPP-WTP) at Hanford. Before the liquid (supernatant) fraction of Envelope C<sup>(a)</sup> wastes (Tank AN-107 and Tank AN-102 waste) can be disposed of as low-activity waste glass, pretreatment is required to remove radioactive strontium (Sr-90) and transuranic (TRU) elements in addition to Cs-137 and Tc-99. Because of the high concentration of organic complexants in this waste, conventional separation processes (e.g., ion exchange) are not effective for Sr-90 or TRU removal. Under earlier work for the RPP-WTP contractor, Battelle—Pacific Northwest Division (PNWD) and Savannah River Technology Center (SRTC) conducted technology development and demonstration of the Sr-90 and TRU (Sr/TRU) removal process with waste samples from Envelope C tanks (Hallen et al. 2000a,b; Nash et al. 2000a,b). The baseline Sr/TRU process was identified as addition of nonradioactive strontium for precipitation/isotopic dilution and addition of permanganate for precipitation of TRU elements.

Work at PNWD (Hallen et al. 2002) determined the optimal conditions and minimal reagent addition that would provide Sr/TRU decontamination of Tank AN-102 waste adequate to meet Specification 2 (BNI 2002) requirements (20 Ci/m<sup>3</sup> Sr and 100 nCi/g TRU). However, the waste treatment plant's proposed process flowsheet for Envelope C waste also includes the potential blending of high-level waste (HLW) pretreatment streams (filtrate from sludge wash/leach solutions) with tank waste and processing the waste blend through the Sr/TRU removal process. The objective of the work reported here was to verify the optimized process conditions would meet a target (50% of Specification 2 limits) Sr/TRU decontamination of the AN-102/C-104 waste blend. The process verification testing involved a specific waste blend of AN-102 waste samples (Urie et al. 2002a,b) and C-104 sludge wash/leach solutions from HLW pretreatment tests (Brooks et al. 2000). The waste blend was concentrated by evaporation (Lumetta et al. 2002) to meet the target sodium concentration of 5.5M, and a subsample was removed to conduct small-scale Sr/TRU removal tests using process conditions from optimization studies with waste from Tank AN-102 (Hallen et al. 2002). The optimization tests identified process conditions that minimized reagent addition and reduced the process temperature to  $25 \pm 5^{\circ}$ C.

The verification tests were conducted at ambient hot cell temperature ( $26^{\circ}$ C) under only two conditions: Sr at 0.02M and Sr at 0.02M plus permanganate at 0.02M. Experiments were performed in radioactive hot cells using approximately 20-mL samples of waste. The precipitate was digested for 4 hours, at ambient hot cell temperature ( $26^{\circ}$ C). Supernatant decontamination data were obtained from the test data. The Sr-90 and TRU decontamination factors (DFs)<sup>(b)</sup> were compared to determine the efficiency of the Sr/TRU removal process. Preferred Sr/TRU removal conditions were identified for the integrated process verification test on a 1-L sample of the waste blend, which will then be used for Cs-137 and Tc-99 ion exchange tests.

The results from this series of small-scale Sr/TRU decontamination tests are presented in this report. Test conditions and experimental procedures are described in Section 2.0. Experimental results from the tests are discussed in Section 3.0. The major conclusions and recommendations are given in Section 4.0. The

<sup>(</sup>a) Envelope designations are explained in Specification 7 of DOE-RL (1996).

<sup>(</sup>b) The decontamination factor is defined as the amount of the contaminant in the waste before treatment divided by the amount present after treatment.

appendices contain the test instructions, data sheets, logbook entries, analytical data, calculations, and staff roles/responsibilities for this work.

## 2.0 Test Conditions and Experimental Procedures

This section describes the conditions used for the verification tests and the procedures for the experiments and analyses.

#### 2.1. Description of AN-102/C-104 Waste Blend

PNWD received 27 bottles of tank waste from Hanford's 222-S Laboratory. The waste material was taken by grab sampling of AN-102 from riser 022 during the period August 7 through 11, 2000, and shipped to the 222-S Laboratory in the 200 West Area. The sample material was transferred to 500-mL bottles before being shipped to the Radiological Processing Laboratory (RPL) in the 300 Area, where they were inspected upon receipt (Hallen et al. 2002, Appendix A). All of the samples contained a settled layer of light brown solids with a dark brownish/black standing liquid. Eight of the bottles were designated for process testing. The eight AN-102 as-received samples were then homogenized to form a slurry and characterized (Urie et al. 2002a). Supernatant and solids from homogenized jars of AN-102 were blended to make a waste material consisting of 2 wt% undissolved solids. Filtrate, leachate, and wash solutions from Tank C-104 processing (Brooks et al. 2000) were blended with the 2 wt% solids AN-102 waste to produce the AN-102/C-104 waste blend. Blending the AN-102 and C-104 materials was conducted according to the test specification TSP-W375-00-00007 (Urie et al. 2002b). No visible gel formation or net solids increase occurred during the blending process.

A sample of the blended waste was characterized, and the sodium concentration was determined to be 3.2M (Urie et al. 2002b). The target concentration for feed to the Sr/TRU removal process was given as 5.5M sodium in the test specification TSP-W375-01-00003 (Reynolds 2001). The waste was evaporated at 50°C until the sodium concentration was estimated to be 5.5M. Samples of the evaporated waste were taken and analyzed before the tests began (Lumetta et al. 2002). The sodium concentration was determined to be within the test specifications,  $5.5 \pm 0.5M$  after evaporation.

#### 2.2. Development of Test Conditions

The RPP-WTP contract (WTP 2001) requires that the immobilized low-activity waste (ILAW) product contain less than 100 nCi/g TRU and that the average Sr-90 be less than 20 Ci/m<sup>3</sup>. However, shielding for the LAW vitrification facility now requires that every ILAW container be less than or equal to 20 Ci/m<sup>3</sup> Sr-90. Supernatant from Envelope C waste contains levels of Sr-90 and TRU too high to meet ILAW requirements. At the design basis waste oxide loading of 15 wt% for Envelope C tanks, waste from AN-102 needs DFs of approximately 10 for Sr-90 (90% removal) and 2 for TRU (50% removal) to meet a target of 50% below the ILAW disposal requirements. Blending the AN-102 waste with the wash/leach solutions resulted in over 60% dilution in the Sr-90 and Am-241 activity relative to the sodium concentration. As a result of the waste blending, DFs of approximately 5 for Sr-90 (80% removal) and 1.6 for TRU (38% removal) are needed to meet a target of 50% below the ILAW disposal requirements. Since over 90% of the TRU in the AN-102/C-104 blended waste is Am-241, a target DF of 1.6 was established for Am-241.

Experimental conditions were defined using the results from the optimization studies with AN-102 diluted waste (Hallen et al. 2002). Based on these studies, strontium and permanganate treatment levels of 0.02M were used. Blending AN-102 waste with the caustic leach and wash solutions from C-104 resulted in increased free hydroxide. The initial free hydroxide of the AN-102/C-104 waste blend was determined by titration to be 0.33M, substantially higher than the diluted AN-102 used for the optimization studies, which was 0.14M. No additional hydroxide was added for the experiments.

The test matrix, shown in Table 2.1, consisted of only two different reaction conditions: 1) 0.02M Sr-only and 2) 0.02M Sr and 0.02M permanganate. The target concentrations were based on the final composition after addition of all reagents. The quantity of each reagent to add to the waste to achieve these values, as well as the actual quantities that were used, can be found in the test instructions included in Appendix A of this report.

All experiments were conducted at ambient hot cell temperature, 26°C, which was within the test specification requirement of  $25 \pm 5$ °C. Earlier studies showed that ambient temperature resulted in adequate Sr-90 and TRU decontamination (Hallen et al. 2000a, 2002). A precipitate digest time of 4 hours was used, which is expected to allow enough time for isotopic exchange of Sr-90 with added nonradioactive Sr.

#### 2.3. Experimental

The AN-102/C-104 waste blend was evaporated in the High Level Radiochemistry Facility hot cells (in the RPL), and an approximately 50-mL sample was removed and transferred to the Shielded Analytical Laboratory hot cells (in the RPL) for testing (Lumetta et al. 2002). Duplicate 10-mL samples were used to determine the density of the waste after evaporation; the average density of the waste was 1.266 g/mL. The small-scale experiments were conducted in 60-mL sample jars with approximately 20 mL or 25.3 g of the waste blend. The reagents were rapidly added to the jars with an adjustable pipette at ambient hot cell temperature (in the order listed from left to right in the test matrix), and were mixed using magnetic stir bars. The samples were stirred for 4 hours, then duplicate samples were filtered with a 0.45-µm disposable syringe filter for analyses. The samples for chemical and radiochemical analyses were acidified and diluted to the appropriate levels for the analytical method.

Test Number	Sample Number	Target [Sr <sup>2+</sup> ]	Target [MnO4]	Comment
1	SP-01	None	None	Initial waste-filtered
	SP-02	None	None	Initial waste-filtered, duplicate
2	SP-03	0.02M	None	Sr-only
	SP-04	0.02M	None	Sr-only, duplicate
3	SP-05	0.02M	0.02 M	Sr and Mn
	SP-06	0.02M	0.02 M	Sr and Mn, duplicate

Table 2.1. Test Matrix for AN-102/C-104 Waste Blend

Stock solutions of the reagents were prepared for addition to the waste. For the experiments, 0.4M strontium nitrate and 0.4M sodium permanganate were used as the stock solutions. Although the design basis specifies 1M strontium nitrate and 1M sodium permanganate solution, the more dilute solutions were used to provide more accurate dispensing and better mixing/distribution of the reagent. The actual quantities of waste and reagents used are given in the test instructions in Appendix A.

## 2.4. Chemical Analyses

For the chemical analyses, the test specification designated the analytes of interest and minimum reportable quantity (Reynolds 2001). Separation and alpha energy analyses (AEA) were required for Am-241 because of the high Cs-137 concentration. Alpha spectroscopy and total alpha measurements were conducted on selected samples after a discrepancy was noted for Am-241 in one sample, SP-06 (see explanation in Appendix B). The Sr-90 concentration was determined by chemical separation followed by beta counting. Sodium concentration was determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES), as were the other metals listed in the test instructions. Samples were also analyzed by titration to determine the free hydroxide concentration. All of the analytical results are included in Appendix B.

### 3.0 Results and Discussion

The small-scale experiments with blended AN-102/C-104 waste were conducted to verify that adding reduced levels of nonradioactive strontium and permanganate at ambient temperature would provide adequate decontamination when HLW pretreatment streams (filtrate, caustic leach, and wash solutions) were blended with tank waste. The results of the experiments are discussed below.

#### 3.1. Decontamination of Sr-90 and TRU

The series of experiments involved multiple samples, and all samples were analyzed as a single analytical batch to determine the change in waste composition upon treatment. Duplicate samples of the starting waste blend were analyzed after filtration to determine the initial composition of the supernatant. The radionuclide composition of the treated samples was compared with the initial composition to determine the extent of decontamination. The DF for a specific radionuclide is defined as the concentration of the component in the initial waste divided by the concentration after treatment, corrected by the amount of dilution that occurred during sample treatment:

#### $DF = [A]_i / ([A] * MD)$

where  $[A]_i$  is the concentration of component A per mass in the initial sample; [A] is the concentration of component A per mass in the treated sample; and MD is the mass dilution ratio, final mass of treated solution divided by the initial mass of solution. The final mass is determined by summing up the mass of initial waste and all dilutions, adjustments, and/or reagent additions.

The DFs for Sr-90 and Am-241 are shown in Figure 3.1. The Sr-90 DFs were above the target of 5 for both experiments. Adding permanganate increased the removal of Sr-90. The target Am-241 DF of 1.6 was exceeded when permanganate was added. Very little Am-241 was removed when only Sr was added.

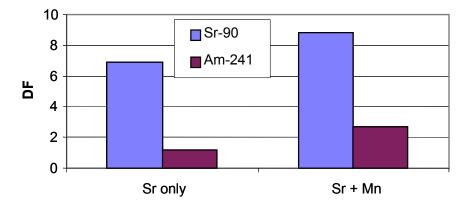


Figure 3.1. Strontium-90 (Target = 5) and Americium-241 (Target = 1.6) Decontamination Factors. Sr-only:  $0.02M \text{ Sr}^{2+}$ ; Sr + Mn:  $0.02M \text{ Sr}^{2+}$  and  $0.02M \text{ MnO}_4^-$ .

The Sr-90 DF and total Sr solubility for AN-102/C-104 waste blend and AN-102 diluted waste are compared in Table 3.1. The Sr-90 DFs are consistently higher for the waste blend than for the diluted waste; 6.9 versus 5.5 for Sr-only and 8.8 versus 7.1 for Sr and  $MnO_4^-$ . The total Sr solubility is higher for the waste blend, but shows a similar reduction with permanganate treatment. The decreased Sr solubility is likely a result of permanganate oxidation of the complexants, EDTA and HEDTA. The increased Sr-90 DF is a direct result of the decrease in Sr solubility. The primary reason for the increased DF with the waste blend is that the initial concentration of Sr-90 was approximately 60% lower, and in turn the isotopic dilution was greater; i.e., the ratio of nonradioactive Sr to Sr-90 was higher. The increased DF was not a result of decreased total Sr solubility; total Sr was actually higher for the waste blend.

	AN-102/C-104 Waste Blend		AN-10 Diluted V	
Test Condition	Sr-90 DF	[Sr] (µg/g)	Sr-90 DF	[Sr] (µg/g)
Sr-only	6.9	197	5.5	167
Sr + Mn	8.8	172	7.1	142

<b>Table 3.1</b> .	Comparison of Sr-90 DFs and Total Sr Solubility for Treated Samples of
	AN-102/C-104 Waste Blend and AN-102 Diluted Waste

The Am-241 and Cm-243+244 DFs for AN-102/C-104 waste blend and AN-102 diluted waste are compared in Table 3.2. Both wastes showed little or no Am and Cm decontamination with Sr addition only. The permanganate addition was required for Am and Cm decontamination; however, significantly less decontamination occurred with the waste blend.

**Table 3.2**. Comparison of Am-241 and Cm-243+244 DFs for AN-102/C-104 Waste Blend and<br/>AN-102 Diluted Waste

Test	AN-102/C-104 Waste Blend		AN-102 Diluted Waste	
Condition	Am-241 DF	Cm <sup>(a)</sup> DF	Am-241 DF	Cm <sup>(a)</sup> DF
Sr-only	1.2	1	0.9	0.8
Sr + Mn	2.7	1.7	4.8	4.8
(a) $Cm = Cm - 243 + 244$ .				

The reduced Am and Cm DFs for the permanganate-treated waste blend is not well understood. The levels of Am and Cm were lower in the initial waste blend as a result of being combined with the wash/leach solutions. The reduced DFs are likely a result of the change in AN-102 chemical composition caused by blending with the C-104 streams. The chemical compositions of the initial wastes are compared in Table 3.3. The caustic leach solution from C-104 was high in caustic and aluminum, resulting in a significant increase in Al and OH<sup>-</sup> in the waste blend. The initial sodium concentration of the waste blend was also 10% higher than the AN-102 diluted waste, but the increased Al and Na are not expected to have a significant impact on TRU DFs. The increased OH<sup>-</sup> could result in higher solubility levels of TRU complexes and have an impact on TRU decontamination. Two key elements found in past studies (Hallen 2000a,b) to correlate with TRU removal, Fe and Mn, were much lower in the waste

blend. This could be the reason for the significantly different TRU decontamination for the waste blend when compared with Envelope C waste alone (AN-102) in Table 3.3, but additional experiments are needed to confirm the effects of chemical composition on TRU decontamination.

	AN-102/C-104	AN-102		
Analyte	(µg/g)	(µg/g)		
Al	7905	5670		
Ca	185	183		
Cd	24.4	24.2		
Co	[1.8]	[1.6]		
Cr	96.6	101		
Cu	10.3 8.4	9.47 29.4		
Fe				
K	856	844		
La	[5.2]	[6.3]		
Mn	[2.7]	[9.3]		
Mo	21.25	20.6		
Na	107000	97000		
Nd	[11.5]	[13]		
Ni	169	162		
Р	397.5	735		
Pb	62.2	68		
Sr	[1.1]	[1]		
Zn	[2.8]	[2.1]		
Zr	[3.3]	[4.2]		
	(M)	(M)		
OH-	0.33	0.14		
	(µCi/g)	(µCi/g)		
Sr-90	21.0	30.0		
Am-241	5.45E-2	6.42E-2		
Cm-243+244	2.15E-3	2.15E-3		
Values in brackets are in low concentration,				
values less than 10 times the detection limit, and				
analytical error likely to exceed 15%.				

 Table 3.3. Comparison of the Starting Composition of AN-102/C-104 Waste Blend and AN-102 Diluted Waste

#### 3.2. Change of Chemical Composition

Chemical analyses of each sample were conducted using ICP-AES. The ICP data from each sample can be used to determine the impact of reagent addition on the chemical composition of the supernatant. The impact of the process conditions on the chemical composition of the treated supernatant is calculated as

percent removed relative to the starting waste. Table 3.4 shows the composition of the initial waste blend in  $\mu$ g/g and the percent change that occurred for the two treated samples. A number of the analytes show little or no significant change on treatment: Al, Cd, Co, Cr, Cu, K, Mo, Na, Ni, P, and Pb. It is important that Al, Cr, and P remain in the supernatant so they are vitrified in the low-activity glass melters. The low Cr removal for permanganate treatment is in contrast to the AN-102 diluted waste, which showed significant Cr removal (30 to 50%).

	Initial	Sr-only	Sr + Mn	
Analyte	(µg/g)	% Removal	% Removal	
Al	7900	0	-2	
Ca	185	21	16	
Cd	24.4	1	-4	
Со	[1.8]	[1]	[-27]	
Cr	96.6	4	8	
Cu	10.3	0	-5	
Fe	8.4	-51	67	
K	856	0	-11	
La	[5.2]	[38]	[55]	
Mn	[2.7]	[13]	[30]	
Мо	21.3	1	-7	
Na	107000	1	-6	
Nd	[11.5]	[18]	[31]	
Ni	169	1	[-7]	
Р	398	2	-5	
Pb	62.2	1	2	
Sr	[1.1]	[-19000]	[-17000]	
Zn	[2.8]	[7]	[46]	
Zr	[3.3]	[11]	[47]	
Values in brackets are in low concentration, values less than 10 times the detection limit, and analytical error likely to exceed 15%.				
	u 1570.			

<b>Table 3.4</b> . Initial Concentration and Percent Removal of ICP Metals for the Treate
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Strontium addition had the most impact on chemical composition; the large negative values represent the large increase in Sr concentration in the treated supernatant. The Sr addition also removed Ca from solution. The Ca removal was not significantly impacted by the addition of permanganate. The apparent Fe addition with Sr-only is likely due to analytical error in the initial or Sr-only samples. The Sr-only addition resulted in some La and Nd removal. Significant Fe, La, Mn, Nd, Zn, and Zr were removed when both Sr and permanganate were added. The La removal was similar to that obtained for Am-241, 63% removal. However, the Mn and Nd percent removals were significantly lower for the AN-102/C-104 waste blend. Chemical composition differences in the waste blend have resulted in slightly less TRU removal by the permanganate.

#### 3.3. Estimated Sr-90 and TRU Levels in ILAW Glass

The data from these experiments can be used to estimate the Sr-90 and TRU loadings that would be expected in ILAW glass made from the treated supernatant. Values listed in Table 3.5 are given for the current baseline design waste glass concentration of 15 wt% Na<sub>2</sub>O in the ILAW. The results show that both treated samples were below the contract limits for ILAW glass. Strontium addition at 0.02M and ambient temperature (26°C) treatment/digest is adequate to meet the contract requirement. The TRU loading of the initial waste blend was below the contract limit because of the low TRU content in the C-104 filtrate, leach, and wash streams. However, significant TRU removal/decontamination only occurred when permanganate was added.

Test Condition	Sr-90 (Ci/m <sup>3</sup> )	TRU (nCi/g)
<b>ILAW Limits</b>	20	100
Initial	58	59
Sr-only	9	49
Sr + Mn	6	22

Table 3.5. Sr-90 and TRU ILAW Glass Loading for 15 wt% Waste Na<sub>2</sub>O

## 4.0 Conclusions and Recommendations

Waste samples from Tank AN-102 were blended with filtrate, leach, and wash solutions from the pretreatment of HLW waste from Tank C-104. The waste blend was concentrated by evaporation from approximately 3.2M sodium to 5.5M sodium for Sr/TRU removal testing. Two experiments were conducted with reduced reagent concentration to verify process conditions for integrated testing. The results of these experiments demonstrated adequate Sr/TRU removal to meet ILAW requirements. The blending of Tank AN-102 waste with Tank C-104 HLW pretreatment streams resulted in higher Sr-90 decontamination but slightly less TRU decontamination when compared with diluted tank waste.

Reagent addition and precipitate digest were conducted at ambient hot cell temperature  $(26^{\circ}C)$ , which was within the test specification requirement of  $25 \pm 5^{\circ}C$ . Reagent addition was  $Sr(NO_3)_2$  at 0.02M with and without permanganate at 0.02M. Both experiments were at the initial free hydroxide level of the waste blend, 0.33M. The decontamination factors for Sr-90 from both experimental conditions evaluated were 5 or higher, which is an adequate decontamination to meet ILAW disposal requirements. The addition of permanganate slightly increased the Sr-90 decontamination. These results verify that, for Sr-90 removal, the precipitation temperature can be  $25 \pm 5^{\circ}C$  and the Sr(NO<sub>3</sub>)<sub>2</sub> addition can be reduced to 0.02M for the AN-102/C-104 waste blend.

The Sr-90 DFs were higher for the AN-102/C-104 waste blend than were obtained for similar treatment conditions with AN-102 diluted waste. The primary mechanism for Sr-90 removal is isotopic dilution with the added nonradioactive  $Sr(NO_3)_2$ . Comparing the total soluble Sr data (by ICP-AES) for the two wastes showed higher total Sr solubility for the waste blend. The higher Sr-90 DF for the waste blend was a result of the lower concentration of Sr-90, approximately 60% less than the AN-102 diluted waste; thus, at a fixed reagent addition level, 0.02M, more isotopic dilution occurred with the waste blend. The permanganate addition reduced the total Sr levels, which resulted in a higher Sr-90 DF than Sr addition alone. The reduction in total soluble Sr by permanganate treatment is likely a result of partial oxidation of the chelating agents, EDTA and HEDTA.

The TRU decontamination in the AN-102/C-104 waste blend only occurred when permanganate was added. The TRU removal exceeded the requirements for ILAW glass by a factor of approximately 5. However, the TRU DFs were lower for the AN-102/C-104 waste blend than the TRU DFs obtained for similar treatment conditions with AN-102 diluted waste. Initial TRU levels were decreased by approximately 80% by the waste blending, but after treatment the TRU levels were higher than for the corresponding AN-102 diluted waste. The reason for the decreased DF for the waste blend is not clearly understood. The chemical composition of the AN-102 waste changed when combined with the filtrate, leach, and wash solutions from C-104. The reduced TRU DF could be a result of the increased hydroxide concentration in the waste blend, 0.33M versus 0.14M for the AN-102 diluted waste. Further studies with AN-102 will provide a better understanding of the impact of recycle streams on TRU removal with permanganate.

The recommended Sr/TRU removal process conditions for integrated process verification tests with AN-102/C-104 waste blend are as follows:

- 1. Evaporate to a final target sodium concentration of 5.5M.
- 2. Then, at a temperature of  $25 \pm 5^{\circ}$ C with the waste stirring, add Sr(NO<sub>3</sub>)<sub>2</sub> or nonradioactive Sr and sodium permanganate to give a final concentrations of 0.02M.
- 3. After both reagents are added, stir/digest the precipitate at  $25 \pm 5^{\circ}$ C for 4 hours.
- 4. Conduct crossflow filtration testing at  $25 \pm 5^{\circ}$ C.

These conditions are much lower than the current baseline Sr/TRU removal conditions of 0.075M Sr, 0.05M permanganate, 1M additional hydroxide, and treatment/digest at 50°C. The recommended conditions would result in substantial savings in procurement costs for the reagents; less precipitation and filter cycle time; fewer solids to filter, hence less filter capacity required, and less wash stream for recycle; and fewer HLW solids that need to be stored and incorporated into HLW glass. Eliminating the additional sodium hydroxide could allow higher waste loading in ILAW glass that will reduce the total volume of low-activity waste glass.

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