Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Assessment of the Mechanisms for Sr-90 and TRU Removal from Complexant-Containing Tank Wastes at Hanford

R. T. Hallen[a]; J. G. H. Geeting[a]; M. A. Lilga[a]; T. R. Hart[a]; F. V. Hoopes[a]
[a] Battelle-Pacific Northwest Division, Richland, WA, USA

To cite this Article Hallen, R. T. , Geeting, J. G. H. , Lilga, M. A. , Hart, T. R. and Hoopes, F. V.(2005) 'Assessment of the Mechanisms for Sr-90 and TRU Removal from Complexant-Containing Tank Wastes at Hanford', Separation Science and Technology, 40: 1, 171 — 183

To link to this Article: DOI: 10.1081/SS-200041906

URL: http://dx.doi.org/10.1081/SS-200041906

Separation Science and Technology, 40: 171–183, 2005
ISSN 0149-6395 print/1520-5754 online
DOI: 10.1081/SS-200041906

Assessment of the Mechanisms for Sr-90 and TRU Removal from Complexant-Containing Tank Wastes at Hanford

R. T. Hallen, J. G. H. Geeting, M. A. Lilga,
T. R. Hart, and F. V. Hoopes
Battelle-Pacific Northwest Division, Richland, WA, USA

Abstract: Small-scale tests (~20 mL) were conducted with samples from Hanford underground storage tanks AN-102 and AN-107 to assess the mechanisms for removing Sr-90 and transuranics (TRU) from the liquid (supernatant) portion of the waste. The Sr-90 and TRU must be removed (decontaminated), in addition to Cs-137 and the entrained solids, before the supernatant can be disposed of as low-activity waste. Experiments were conducted with various reagents and modified Sr/TRU removal process conditions to more fully understand the reaction mechanisms. The optimized treatment conditions—no added hydroxide, addition of Sr (0.02 M target concentration) followed by sodium permanganate (0.02 M target concentration) with mixing at ambient temperature—were used as a reference for comparison. The waste was initially two orders of magnitude undersaturated with Sr; the addition of nonradioactive $Sr(NO_3)_2$ saturated the supernatant, resulting in isotopic dilution and precipitation of Sr-90 as $SrCO_3$. The reaction chemistry of Mn species relevant to the mechanism of TRU removal by permanganate treatment was evaluated, along with the importance of various mechanisms for decontamination, such as precipitation, absorption, ligand exchange, and oxidation of organic complexants. For TRU removal, permanganate addition generally gave the highest DF. The addition of Mn of lower oxidation states (II, IV, and VI) also resulted in good TRU removal, as did complexant oxidation with periodate and addition of Zr(IV) for ligand exchange. These results suggest that permanganate treatment leads to TRU removal by multiple routes.

Address correspondence to R. T. Hallen, Battelle-Pacific Northwest Division, Richland, WA 99352, USA. E-mail: richard.hallen@pnl.gov

INTRODUCTION

The world's largest radiochemical processing facility is being constructed on the U.S. Department of Energy's (DOE) Hanford Site, near Richland, Washington. When completed, the Waste Treatment Plant (WTP), occupying 65 acres, will immobilize Hanford's radioactive tank waste by vitrification. Before vitrification, WTP's Pretreatment Facility will separate waste into high-level waste, HLW (or the solids portion, which will contain the highest concentration of radioactivity), and low-activity waste (LAW) [or the liquid (i.e., supernatant, portion, which will contain lower levels of radioactivity)]. In two Hanford tanks (AN-102 and AN-107), the high levels of Sr-90 and transuranics (TRU) in the supernatant require that additional pretreatment be performed to remove the Sr-90 and TRU elements, in addition to Cs-137 and the entrained solids, before the waste can be disposed of as LAW. This paper describes a study conducted with samples from these tanks, which are designated as Envelope C wastes in the WTP contract, to assess the mechanisms for Sr-90 and TRU removal (decontamination).

The Sr-90 removal process consists of isotopic dilution by nonradioactive $Sr(NO_3)_2$ addition and precipitation of $SrCO_3$. The TRU removal process involves addition of permanganate, stepwise manganese reduction, Mn(VII) to Mn(VI) to Mn(IV); precipitation of MnO_2; and concomitant TRU precipitation. This TRU decontamination method is based on work conducted at Pacific Northwest National Laboratory (PNNL) by Orth et al. (1). Entrained solids and Sr/TRU precipitate are to be removed via cross-flow filtration; Cs-137 is to be removed by ion exchange. In previous work for the WTP contractor, conducted by Battelle-Pacific Northwest Division and the Savannah River Technology Center, Sr/TRU removal was demonstrated with actual waste samples from Envelope C tanks (2–5) by added nonradioactive Sr and permanganate. Additional work has been conducted to further evaluate and optimize the pretreatment process (6–8).

EXPERIMENTAL

Small-scale laboratory tests (~20 mL) were conducted with samples of waste from Tanks AN-102 and AN-107. This testing method allowed several different reagents and reaction conditions to be examined with the limited quantities of samples available for testing. The volume was an adequate sample for the required analytical analyses.

Samples of waste from Tank AN-102, collected by grab sampling over the period August 7–11, 2000, were shipped to the Hanford Radiochemical Processing Laboratory (RPL) and characterized (9). Analyses showed that the AN-102 samples contained approximately 0.25 M free hydroxide, 1 M carbonate (total inorganic carbon, TIC), and 2 M total organic carbon

(TOC). The samples were diluted with an appropriate amount of 0.01 M NaOH to give a diluted feed of approximately 5.5 M sodium prior to reagent addition, which represents the WTP. Free OH^- in the diluted samples was determined, by titration, to be 0.14 M.

A 500 mL bottle of AN-107 diluted feed, which had been retained for future testing during benchscale testing (3), was used for these studies. Earlier, the diluted feed had been prepared to a target of 7.7 M sodium and 1.1 M added hydroxide (10). The carbonate (TIC) and TOC concentrations were determined to be 1.4 M and 2.5 M, respectively. Since the waste had been stored in the hot cell and aged for approximately 4 ys, samples were taken and reanalyzed before the experiments were begun. The sodium concentration was determined to be 8.6 M by inductively coupled plasma–atomic emission spectrometry (ICP-AES), and titration showed the free hydroxide to be 0.55 M. Since excess free hydroxide was still present in the AN-107 diluted feed, no caustic adjustment was required. The [Na] data were used to determine the quantity of 0.01 M NaOH to add to the AN-107 sample to yield approximately 5.5 M sodium in the waste prior to treatment.

Experimental conditions were defined using the results from earlier reaction mechanism studies with AN-102/C-104 waste blend simulant (11). Based on those studies, minimum levels (0.02 M) of reagents were added to determine the differences in the effectiveness of treatment conditions. The addition of 0.02-M $Sr(NO_3)_2$ and permanganate had also showed adequate Sr-90 and TRU decontamination in earlier studies (6). The test matrix for each tank sample was slightly different. The total number of tests was held to a minimum to limit the volume of waste used and to keep associated analytical costs within the budget. This information was used to construct the test matrices shown in Tables 1 and 2 for AN-102 and AN-107, respectively. The target concentrations listed in the test matrices are based on the final composition after addition of all reagents. Both test matrices included a repeat of the second experimental conditions as the final experiment in the matrix to examine the variability in conducting duplicate experiments.

The waste samples were diluted with 0.01 M NaOH just prior to testing in the Shielded Analytical Laboratory hot cells (in the RPL). The small-scale experiments were conducted in 60 mL sample jars with approximately 20 mL of the diluted tank waste. The reagents were added rapidly to the wastes with an adjustable pipette, in the order listed in Tables 1 and 2 (from left to right), at ambient hot cell temperature, and mixed with magnetic stir bars when specified. The ambient hot cell temperatures ranged from 26°C to 28°C on the days of these tests. Each series of experiments was conducted over a 2-day period, approximately 1 month apart. Samples were collected at the specified times of 4 h and/or 24 h and filtered immediately with a 0.2 μm disposable syringe filter. Duplicate samples of initial waste, RX-01 and SS-01, were filtered, along with the other samples, but no chemical reagents were added. The samples for chemical and radiochemical analyses were acidified and

Table 1. Test matrix for experiments using Tank AN-102 waste samples

Experiment number	Sr^{+2}	Mn(VII)	Mn(VI)	Mn(IV)	Mn(II)[a]	Other[b]	Stir	Added OH^-
RX-01	–	–	–	–	–	–	No	AR[c]
RX-02	0.02 M	0.02 M	–	–	–	–	Yes	AR
RX-03	–	0.02 M	–	–	–	–	Yes	AR
RX-04	0.02 M	0.02 M	–	–	–	–	Yes	0.3 M
RX-05	0.02 M	0.02 M	–	–	–	–	No[d]	AR
RX-06	0.02 M	–	0.02 M	–	–	–	Yes	AR
RX-07	0.02 M	–	–	Solid[e]	–	–	Yes	AR
RX 08	0.02 M	–	–	–	0.02 M	–	Yes	AR
RX-09	0.02 M	–	–	–	–	IO_4^-[f]	Yes	AR
RX-10	0.02 M	–	–	–	–	Zr(IV)[g]	Yes	AR
RX 11	0.02 M	0.02 M	–	–	–	–	Yes	AR

[a] Mn^{+2} precipitates as $Mn(OH)_2$, which air oxidizes to Mn(IV).

[b] Periodate (IO_4^-) is a nonprecipitating oxidant, and zirconium, Zr(IV), is a nonoxidizing precipitant.

[c] AR = as received, 0.14 M OH^-; no added hydroxide.

[d] Mix sample after 1h of reaction.

[e] Solid reagent, freshly precipitated $MnO(OH)(ONa) \cdot xH_2O$, same number of Mn equivalents added.

[f] 0.06 M KIO_4^-, added as solid reagent.

[g] 0.02 M $Zr(NO_3)_4$.

Table 2. Test matrix for experiments using tank AN-107 waste samples

Experiment number	Sr^{+2}	Mn(VII)	Mn(VI)	Mn(II)[a]	Other[b]	Stir
SS-01	–	–	–	–	–	No
SS-02	0.02 M	0.02	–	–	–	Yes
SS-03	0.02 M	–	0.02 M	–	–	Yes
SS-04	0.02 M	0.02 M	–	–	–	No[c]
SS-05	0.02 M	–	–	0.02 M	–	Yes
SS-06	0.02 M	–	–	–	IO_4^-[d]	Yes
SS-07	0.02 M	–	–	–	Zr(IV)[e]	Yes
SS-08	0.02 M	0.02 M	–	–	–	Yes

[a] Mn^{+2} precipitates as $Mn(OH)_2$, which air oxidizes to Mn(IV).

[b] Periodate (IO_4^-) is a nonprecipitating oxidant, and zirconium, Zr(IV), is a nonoxidizing precipitant.

[c] Mix sample after 1h of reaction.

[d] 0.06 M KIO_4^-, added as solid reagent.

[e] 0.02 M $Zr(NO_3)_4$.

diluted to the appropriate levels for the analytical method. Samples for titration were submitted without any chemical addition. Sample RX-06, Mn(VI) addition, was spilled during weighing after the reagent addition. The sample could not be recovered, and no additional waste remained to re-run this condition. The Mn(VI) test condition, SS-03, was examined with AN-107 waste to provide data for reaction mechanism assessment.

Stock solutions of the reagents were prepared for addition to the waste. The experiments used 0.44 M solutions of $Sr(NO_3)_2$, $NaMnO_4$, $MnCl_2$, and $Zr(NO_3)_4$. The Mn(VI) solution was freshly prepared before each series of tests by reducing $NaMnO_4$ (0.44 M) with an equal volume of 0.22 M $NaHCO_2$ in 2 M NaOH. The resulting solution was 0.22 M Mn(VI), and 2 mL were added to approximately 20 mL of waste to give the target of 0.02 M Mn addition. The 1 M free hydroxide stabilized the Mn(VI) solutions for up to 24 h after preparation. Without additional hydroxide, the Mn(VI) would disproportionate to Mn(VII) and Mn(V), ultimately leading to loss of Mn(VI) from the solution (12).

The Mn(IV) solids were prepared by the $NaMnO_4$ reduction with $NaHCO_2$ in 0.1 M NaOH. After complete precipitation of the Mn (no remaining solution color), the solids were collected on a filter, washed with deionized water, and then dried in a vacuum oven. A sample of the dried solids was digested and analyzed by ICP-AES. The formula weight of the Mn(IV) solids was determined to be 205 g per mole of Mn, which infers the Mn as deprotonated, hydrated, manganese dioxide, $Mn(O)(OH)(ONa) \cdot xH_2O$. The target Mn addition of 0.02 M is equal to 0.09 g of solids added for each approximately 20 mL of sample. Potassium periodate, target 0.06 M or 0.3 g, was added as a solid because of its low solubility in water at room temperature. Caustic addition to RX-04 involved adding 1 mL of 6.6-M NaOH to approximately 20 mL of waste.

DECONTAMINATION OF Sr-90

Each series of experiments using AN-102 and AN-107 waste involved multiple samples, analyzed as multiple analytical batches, and provided analytical results to determine the change in waste composition upon treatment. Duplicate samples of each starting waste 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 decontamination factor (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, which is defined as the final mass of treated solution divided by the initial mass of solution. (The mass dilution factor corrects DF so the dilution by reagent addition is not shown to decontaminate the waste.) The final mass is determined by summing the mass of initial waste and all dilutions, adjustments, and/or reagent additions.

The DFs for Sr-90 from treated AN-102 samples are shown in Fig. 1. Sr-90 removal was observed for all experiments where nonradioactive $Sr(NO_3)_2$ was added, consistent with the mechanism for Sr-90 removal involving isotopic dilution and $SrCO_3$ precipitation. The addition of permanganate alone resulted in no Sr-90 removal; nonradioactive Sr must be added to remove Sr-90 from Envelope C wastes. The Sr-90 DFs were greatly increased by the solids digest time of 24 h vs. 4 h. The added free hydroxide (0.3 M) had little impact on the Sr-90 DFs compared with treatment of the waste without any additional hydroxide (0.1 M). The no-mix experiment for the first hour of reaction did not have a significant impact on the Sr-90 DF, supporting earlier conclusions from simulant tests (110) that mixing during reagent addition and initial permanganate reduction are not important as long as the treated samples are well mixed before filtration. The oxidation state of the Mn added had little impact on the Sr-90 DF. Sr-90 removal was less effective when combined with periodate addition, compared with other experiments where $Sr(NO_3)_2$ was added. Nonradioactive Sr and Zr(IV) addition gave a Sr-90 DF that

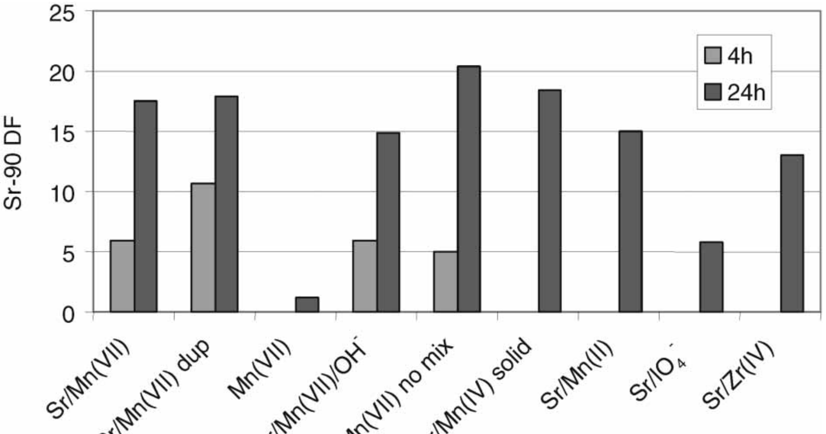

Figure 1. Strontium-90 decontamination factors for treated AN-102 samples as a function of added reagent and reaction time. Target reagent levels: 0.02 M for all except OH^- (target of an additional 0.3 M) and IO_4^- (target concentration of 0.06 M).

was nearly as high as nonradioactive Sr and permanganate. These results suggest that complexant oxidation is not important for Sr-90 removal, since permanganate treatment, oxidation, did little to improve the Sr-90 DF. The primary mechanisms for Sr-90 decontamination are isotopic dilution by adding nonradioactive $Sr(NO_3)_2$ at concentrations well over the solubility limit and the precipitation of Sr-90 from solution as $SrCO_3$.

Addition of 0.02 M nonradioactive Sr would result in 1400 μg/g [Sr] if $SrCO_3$ precipitation did not occur. The [Sr] data for the AN-102 tests are shown in Fig. 2. Note that the relatively high concentration of Sr for periodate addition corresponds to precipitation of 84% of the added Sr, and the highest DFs (permanganate addition) correspond to precipitation of 94% of the added Sr. The [Sr] data also show the same consistent trend as found in earlier tests: at 4 h, the total [Sr] is still relatively high; after 24 h, the values are generally half the 4 h values, resulting in a doubling of the Sr-90 DF. However, the [Sr] is not the only difference in DF for the 4 h samples. When the isotopic dilution ratio, Sr-90/[Sr], is examined (Table 3), three of the four samples taken at 4 h of reaction time do not appear to have reached the final equilibrium ratio (~0.018 μCi/μg). The experimental average of the 24 h samples and the calculated ratio based on the quantity of added $Sr(NO_3)_2$ agree quite well, showing that isotopic exchange is complete. The slow isotopic exchange for the 4 h samples is likely a result of the procedure used for the small-sample tests with reagent addition via an autopipetter, where the $Sr(NO_3)_2$ solution is injected rapidly into the waste solution. In the benchscale tests where complete isotopic exchange was noted at 18 min, the Sr solution was added slowly over 6 min with continuous stirring by an

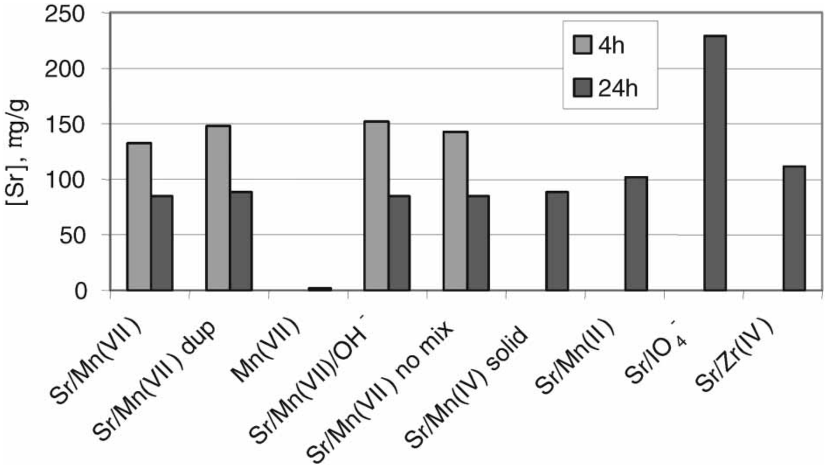

Figure 2. Total strontium solubility in the treated AN-102 samples as a function of reagent addition and reaction time.

Table 3. Isotopic exchange ratio, [Sr-90]/[Sr] in μCi/μg, for AN-102 tests

	[Sr-90], μCi/g		[Sr], μg/g		[Sr-90]/[Sr], μg/g	
Conditions	4 h	24 h	4 h	24 h	4 h	24 h
Initial	28	28	[1.2]	[1.1]	24.1	25.8
Sr/Mn(VII)	4.4	1.5	132	85	0.0330	0.0177
Sr/Mn(VII) dup	2.5	1.5	149	88	0.0165	0.0166
Mn(VII)	–	22	–	[1.0]	–	22.2
Sr/Mn(VII)/OH^-	4.3	1.7	151	85	0.0281	0.0198
Sr/Mn(VII) no mix	5.3	1.3	141	85	0.0373	0.0150
Sr/Mn(IV) solid	–	1.5	–	89	–	0.0164
Sr/Mn(II)	–	1.7	–	101	–	0.0172
Sr/IO_4^-	–	4.7	–	228	–	0.0204
Sr/Zr(IV)	–	2.0	–	111	–	0.0180
				Experimental Avg.		0.0176
				Stddev		0.0018
				Calculated ratio		0.0185

Values in brackets [] are greater than the method detection limit (MDL) but less than the estimated quantitation limit (EQL), with errors likely to exceed 15%.

overhead-driven impeller. The undersaturation of Sr in the initial waste can be seen by comparing the [Sr] in the initial waste (~1 μg/g) to the [Sr] in any of the samples with added nonradioactive Sr (85–228 μg/g).

The results for experiments with AN-107 waste can be compared to those for AN-102 waste. The comparison of Sr-90 DFs at 24 h of reaction is shown in Fig. 3. Sr-90 removal was high (DF $>$ 10) for the AN-107 waste and in most cases quite similar to AN-102. The corresponding TRU removal process did not significantly impact Sr-90 removal from AN-107, as was noted for AN-102. Only minor differences appear for Sr-90 removal from the two Envelope C wastes for the no-mix and periodate experiments. The AN-102 sample results appear to be more sensitive to changes in process conditions than for corresponding treatment conditions with AN-107 samples. The carbonate (TIC) concentrations are high ($>$0.5 M) in both these wastes and likely have little impact on the differences observed, as only 0.02 M is consumed on $SrCO_3$ precipitation.

DECONTAMINATION OF TRU

The effectiveness of the various treatment conditions for TRU removal from AN-102 can be seen by examining the DFs for the sum of the alpha shown in Fig. 4. The DFs were significantly higher when oxidation occurred (i.e., experiments where permanganate, Mn(VII), or periodate was added). The

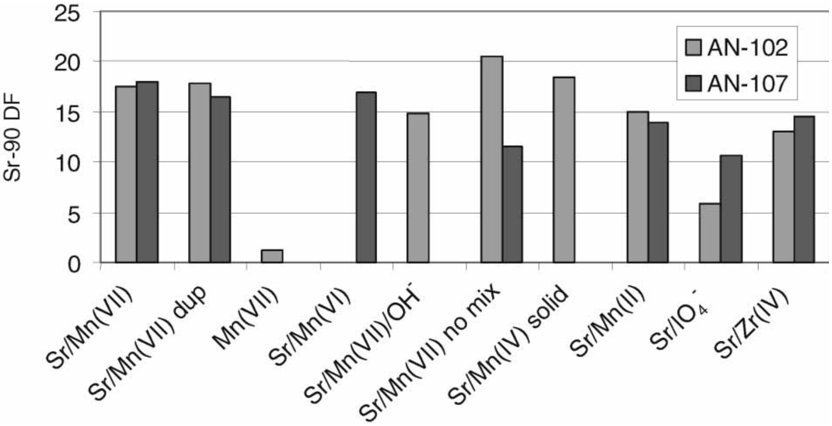

Figure 3. Comparison of Sr-90 DF for AN-102 and AN-107 experiments.

periodate treatment (oxidation) showed even greater TRU removal than permanganate, possibly because the target concentration of periodate was 0.06 M, which corresponds to about twice the oxidation equivalence as permanganate at 0.02 M. Process changes coupled with permanganate addition (no Sr addition; hydroxide added; or no mixing for the first hour of reaction) did not significantly impact TRU decontamination. Precipitation, sorption, and ligand exchange by reduced states of Mn(IV and II) and

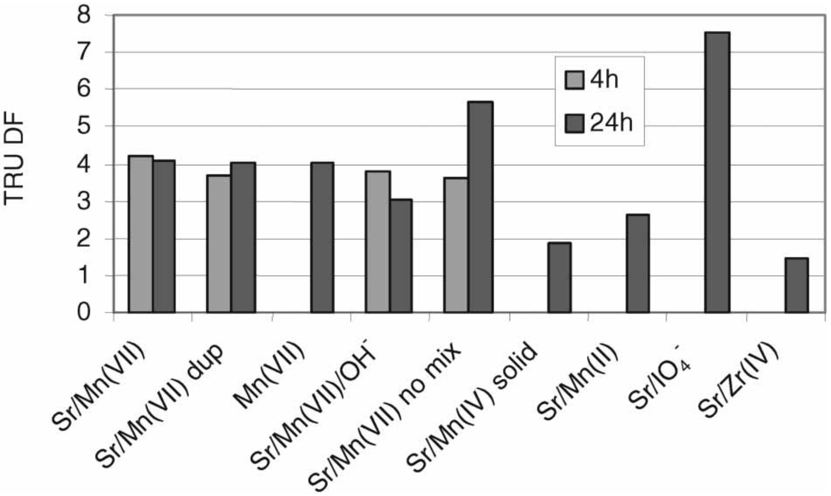

Figure 4. TRU (sum of alpha) decontamination factors for treated AN-102 samples as a function of reagent addition and reaction time.

Zr(IV) resulted in only low levels of TRU removal, DF ∼2 (50% removal). The precipitation of Mn solids in the waste solution (no mix) resulted in higher TRU DFs than addition of preformed Mn(IV) solids. The high organic content in the waste solution likely resulted in the formation of more reduced Mn solids, which were more effective at TRU removal than preformed Mn(IV) solids. There was very little difference in time dependence in the TRU DFs, with little difference between DFs at 4h and 24h sample times.

Two different radiochemical methods were used to determine the alpha content of the waste samples. Individual isotope pairs were determined by the alpha counting technique and added together to give the sum of alpha. Total alpha was determined in a separate technique by counting the entire sample. The DFs for the two techniques and individual isotopes can be plotted individually for comparison (Fig. 5). DFs for the two techniques and Am and Cm isotopes are very similar. The Am and Cm isotopes are most likely in the most stable +3 oxidation state. Since Am-241 accounts for approximately 90% of the TRU in AN-102 waste, the Pu-238 + Am-241 data can be considered all Am-241. The Cm isotopes, primarily 243 and 244, account for approximately 5% of the total TRU. The Cm isotope data have greater variability due to increased analytical error associated with the very low concentrations of these isotopes. The DFs for the Pu isotopes (Pu-239 + 240) are consistently lower than found for the Am and

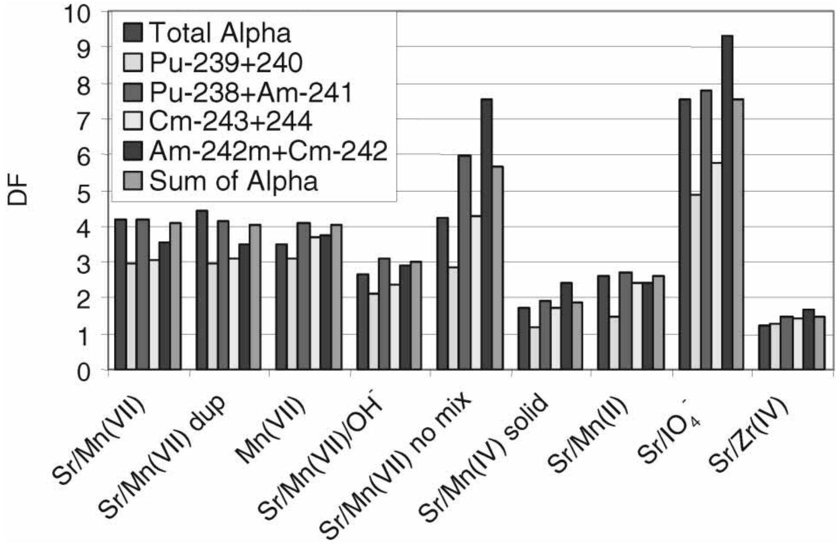

Figure 5. Comparison of decontamination factors for total alpha, the various isotope pairs, and the sum of alpha (treated AN-102 at 24 h).

Cm isotopes. Both Pu and U + Np isotopes were reported to have lower DFs than Am and Cm in earlier bench-scale tests (8). Permanganate treatment is most effective for removing the +3 valence TRU elements.

Treatment of AN-107 waste under similar conditions gave much different results than AN-102 (see the comparison shown in Fig. 6). TRU removal from AN-107 was significantly higher for most treatment schemes compared to the corresponding treated AN-102 sample. The results for Mn(II), IO_4^-, and Zr(IV) show that oxidation is not as important for TRU decontamination in AN-107 waste. This was somewhat surprising because AN-107 is initially higher in TOC and concentrations of soluble Fe and Mn. Co-precipitation, sorption, and ligand exchange result in high TRU removal from AN-107 waste. AN-102 waste may have higher concentrations of active complexants that require oxidation for effective TRU removal.

SUMMARY AND CONCLUSIONS

Sr-90 is removed by addition of nonradioactive $Sr(NO_3)_2$. The addition of a chemical oxidant did not increase Sr-90 decontamination. None of the various treatment schemes or reagents added showed an improved Sr-90 DF. Strontium concentrations, and, therefore, decontamination levels, are time dependent. In all experiments in which Sr was added, the Sr concentrations decreased significantly between 4 h and 24 h. In some cases, the concentration was halved over this time (i.e., a doubling of the Sr-90 DF). This behavior is the same as that observed for simulated waste

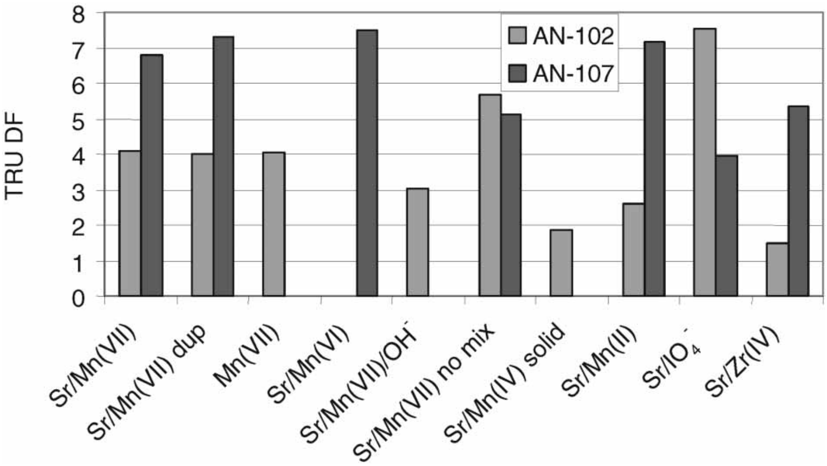

Figure 6. Comparison of TRU (sum of alpha) DF for AN-102 and AN-107 waste samples (24 h).

samples (11). The Sr concentrations in the actual waste and the simulated waste were also similar, again corresponding to the simulant studies.

The important factors for determining Sr-90 decontamination are the isotopic dilution ratio and the [Sr]. Approximately 95% of the added Sr is precipitated from solution. The isotopic dilution is quite rapid relative to $SrCO_3$ precipitation. The precipitation of $SrCO_3$ is slow, and adequate time must be allowed for equilibrium to be approached. The results from this study, therefore, suggest that the treated waste should not be filtered at 4h after reagent addition; instead, additional time should be allowed for Sr to precipitate.

For evaluating treatments that employed various species of Mn(VII, VI, IV, and II); an alternative oxidant; and competing metal ion, Zr(IV), the DF of TRU (sum of alpha) was used as an indication of extent/effectiveness. The performance of the reference treatment conditions was very similar to earlier tests with actual waste (8). In general, the reference process conditions gave the best TRU removal of the treatments tested. For AN-102, the treatments involving oxidation performed the best, although addition of Mn(IV), Mn(II), and Zr(IV) resulted in some TRU removal. For AN-107, oxidation appears to be less important, and similar TRU removal was found for permanganate and reduced Mn(II) and Zr(IV). AN-107 appears easier to treat for TRU removal. Ligand displacement, precipitation, and sorption of TRU ions appear to be important for AN-107.

Time has very little impact on the TRU removal when comparing the 4h and 24h data. However, increased levels of Mn were noted for the treated samples after only 4h of reaction, whereas all Mn was removed by 24h of reaction. These results again suggest that the treated waste should not be filtered after only 4h of reaction; in this case, the increased Mn levels will likely lead to postfiltration precipitation. Instead, if filtration begins after 24h of reaction, all Mn will already have precipitated from solution.

The results for AN-102 simulant and active tests were very similar, supporting the earlier conclusions of Lilga et al. (11). The lanthanide elements used in simulant studies as surrogates for the TRU elements showed very similar removals as the actual waste samples and correlated well with the TRU decontamination. This correlation is likely because 95% of the TRU in the Envelope C wastes are Am and Cm isotopes that are in a similar oxidation state as the lanthanide elements.

The initial waste from Tank AN-107 was higher in both Sr-90 and TRU compared to waste from Tank AN-102. The experimental data from both wastes can be used to predict the loading of Sr-90 and TRU (sum of alpha) expected in immobilized low-activity waste (ILAW) at a fixed waste sodium loading of 15 wt% (contract limit > 10 wt%). At 24h of reaction, the levels of Sr-90 and TRU in the supernatant from treatment of both wastes were quite similar. For Sr-90 loading, the levels were three times below the ILAW requirements of $20\,\mathrm{Ci/m^3}$ and, for TRU loading, the levels were four times below the requirements of 100 nCi/g when treated by addition of nonradioactive Sr and sodium permanganate at reagent levels of 0.02 M. These reagent levels are significantly below the baseline treatment conditions of 0.075 M Sr and 0.05 M permanganate.

REFERENCES

1. Orth, R.J., Zacher, A.H., Schmidt, A.J., Elmore, M.R., Elliott, K.R., Neuenschwander, G.G., and Gano, S.R. (1995) *Removal of Strontium and Transuranics from Hanford Tank Waste Via Addition of Metal Cations and Chemical Oxidant–FY 1995 Test Result*; Pacific Northwest National Laboratory: Richland, WA, PNL-10766.
2. Hallen, R.T., Brooks, K.P., and Jagoda, L.K. (2000) *Demonstration of Entrained Solids and Sr/TRU Removal Processes with Archived AN-107 Waste*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3033.
3. Hallen, R.T., Bredt, P.R., Brooks, K.P., and Jagoda, L.K. (2000) *Combined Entrained Solids and Sr/TRU Removal from AN-107 Diluted Feed*; Battelle, Pacific Northwest Division: Richland, WA, PNWD.
4. Nash, C.A., Rosencrance, S.W., and Wilmarth, W.R. (2000) *Entrained Solids, Strontium-Transuranic Precipitation, and Crossflow Filtration of AN-102 Small C*; Savannah River Technology Center: Aiken, SC, BNF-033-98-0218.
5. Nash, C.A., Hiroshi, H.H., and Wilmarth, W.R. (2000) *Strontium-Transuranic Precipitation and Crossflow Filtration of 241-AN-102 Large C*; Savannah River Technology Center: Aiken, SC, BNF-033-98-0317.
6. Hallen, R.T., Burgeson, I.E., Hoopes, F.V., and Weier, D.R. (2002) *Optimization of Sr/TRU Removal Conditions with Samples of AN-102 Tank Waste*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3141.
7. Hallen, R.T., Burgeson, I.E., and Hoopes, F.V. (2002) *Verification of Process Conditions for Sr/TRU Removal from AN-102/C104 Waste Blend*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3189.
8. Hallen, R.T., Geeting, J.G.H., Jackson, D.R., and Weier, D.R. (2002) *Combined Entrained Solids and Sr/TRU Removal from AN-102/C-104 Waste Blend*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3264.
9. Urie, M.W., Fiskum, S.K., Campbell, J.A., Farmer, O.T., Greenwood, L.R., Hoppe, E.W., Mong, G.M., Soderquist, C.Z., and Swoboda, R.G. (2002) *Chemical Analysis and Physical Property Testing of 241-AN-102 Tank Waste-Supernatant and Centrifuged Solids*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3229.
10. Urie, M.W., Greenwood, L.R., Wagner, J.J., Farmer, O.T., Fiskum, S.K., Ratner, R.T., and Soderquist, C.Z. (1999) *Inorganic and Radiochemical Analysis of AW-101 and AN-107 "Diluted Feed" Materials*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-2463.
11. Lilga, M.A., Hallen, R.T., Boettcher, K.M., Hart, T.R., and Muzatko, D.S. (2003) *Assessment of the Sr/TRU Removal Precipitation Reaction Mechanisms Using Waste Simulant Solutions*; Battelle, Pacific Northwest Division: Richland, WA, PNWD-3263.
12. Cotton, F.A. and Wilkinson, G. (1972) *Advanced Inorganic Chemistry*; Interscience: New York.