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Effects of Sodium Hydroxide and a Chelating Agent on the Removal of Aluminum from Radioactive Sludge

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Abstract: An essential step during the remediation of nuclear waste by the U.S. Department of Energy involves the separation of nonradioactive components such as aluminum from high-level waste sludges to minimize the ultimate volume to be stored in a nuclear waste repository. Plans for waste treatment at Hanford and the Savannah River Site include the use of 1 to 3 m sodium hydroxide (NaOH) at an elevated temperature to leach the aluminum from the sludge. Triethanolamine (TEA) was added to caustic leaching solutions in an effort to improve the solubility of aluminum from authentic tank-waste sludge. High-level radioactive waste sludge with significant amounts of gibbsite and hard-to-dissolve boehmite phases was used in these tests. In concept, a chelating agent such as TEA can both improve the dissolution rate and increase the aluminum concentration in the liquid phase. However, TEA could also increase the solubility of other sludge components that are potentially problematic to downstream processing.

Six tests were performed with leachate concentrations ranging from 0.1 to 3.0 m NaOH, 0 to 3.0 m TEA, and 0 to 2.9 m $NaNO_3$. One test was performed using 3.0 m NaOH at 80°C in order to simulate the baseline process, while the other tests were performed at 60°C. As expected, more aluminum entered the solution at 80°C than at 60°C when other test conditions were held constant. With caustic alone, equilibrium was achieved at both temperatures within 10 days. The addition of TEA significantly increased the concentration of aluminum in the leachate, and the aluminum concentration continued to increase even after 11 days of processing. The fraction of aluminum dissolved at 60°C increased from 35% using 3.0 m NaOH alone to

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87% using a combination of 3.0 *m* NaOH and 3.0 *m* TEA. The high-nitrate, low-hydroxide solutions did not significantly dissolve the aluminum because aluminate ion could not be produced. A small addition of TEA to these low-caustic solutions had no effect on aluminum removal.

The use of TEA also increased the solubility of copper, nickel, and iron, which are only minor constituents. The TEA also had a significant effect on the solubility of the radionuclides ^{137}Cs and ^{60}Co. The significant presence of ^{137}Cs in the leachates was expected with and without TEA. The high-nitrate leaches, which were the least effective of the leaching solutions, removed 69% of the ^{137}Cs from the washed sludge, while a combination of 3.0 *m* NaOH and 3.0 *m* TEA removed 96%. Very little ^{60}Co was removed from the sludge except with the use of the 3.0 *m* NaOH–3.0 *m* TEA solution, which removed 53%. These results indicate that only TEA and ^{60}Co need to be examined for potential chemical and radiological impacts, respectively, on downstream processes.

INTRODUCTION

The U.S. Department of Energy (DOE) is required by law to treat and safely dispose of its radioactive wastes from the reprocessing of spent nuclear reactor fuel and irradiated targets. These nuclear wastes are in a variety of physical forms, which include supernatant, saltcake, and sludge. It is expected that various separation technologies will be used to divide these nuclear wastes into high-level waste (HLW) and low-level waste (LLW) fractions. Successful separation technologies will concentrate the radionuclides into a relatively small volume of HLW, which requires very expensive immobilization and disposal processes. In sharp contrast, the vast majority of the treated nuclear waste will be considered to be LLW, which can then be immobilized and disposed at a small fraction of the HLW costs (1).

In 1993, the DOE considered three separation options for the Hanford sludges. The treatment options (2) included simple sludge washing, enhanced sludge washing, and advanced separations. Simple sludge washing uses only water or inhibited water, which contains 0.01 *M* sodium nitrite and 0.01 *M* sodium hydroxide (NaOH). Enhanced sludge washing (ESW) refers to the simple sludge washing, which is followed by leaching with 2 to 3 *M* NaOH at an elevated temperature. The leached solids are then washed with the inhibited water to remove the dissolved components and any residual NaOH. Advanced separations consist of complete dissolution, if possible, followed by extensive radionuclide separation. An analysis of the options concluded that the simple sludge washing would result in an unreasonably large volume of immobilized HLW, and that advanced separation would require extensive technology development and complex facilities. Therefore, ESW was selected as the baseline process to treat the Hanford sludges. A very similar caustic-based separation process, which is called Extended Sludge

Processing (ESP), was evaluated for the high-aluminum sludges at the Savannah River Site (SRS).

The ESW and ESP treatment processes are expected to solubilize the aluminum in the sludge by converting different forms of aluminum oxides to soluble sodium aluminate (3).

If the proper chelating agent is added to the caustic leaching solution, the agent will complex the aluminate ion [$Al(OH)_4^-$] and drive the dissolution process [Eqs. (1) and (2)].

$$\text{Boehmite} \quad AlOOH(s) + OH^-(aq) + H_2O \longrightarrow Al(OH)_4^-(aq), \qquad (1)$$

$$\text{Gibbsite} \quad Al(OH)_3(s) + OH^-(aq) \longrightarrow Al(OH)_4^-(aq). \qquad (2)$$

While the sludge processing can be performed in the 1,000,000-gal storage tanks, the entire contents of the tank must be heated. Out-of-tank processes for the sludges can also be problematic due to the slow rate of aluminum dissolution. Depending upon the temperature and nitrate concentration, weeks to months may be needed to achieve the desired degree of dissolution. Simulant tests at Oak Ridge National Laboratory (ORNL) have shown that the addition of chelating agents to the ESW or the ESP could lower the temperature required to achieve the desired extent of dissolution and/or increase the dissolution rate. Preliminary tests with bayerite, boehmite, and gibbsite indicated that triethanolamine (TEA) was the most promising candidate. It has a high solubility in water ($\sim$4400 g/L at 30°C) and complexes metals in a molar ratio of 6 TEA : 1 metal (4). Therefore, TEA was selected to determine the effectiveness of chelating agents in the ESP with an actual sludge sample from the SRS. The test procedures and results are presented below. A detailed description of the test apparatus is presented elsewhere (5).

EXPERIMENTAL PROCEDURES

The overall strategy for the preparation of uniform sludge samples for the leaching tests was to transfer the as-received sludge samples into a single bottle, add inhibited water to the sludge, continue to mix and blend the composite sludge slurry, subdivide the slurry into small, uniform aliquots of 1 to 2 g, and decant the inhibited water from the wet sludge solids with the aid of an International Equipment Company Centra-GP8 tabletop centrifuge. These initial steps were performed in a hot cell due to the high radiation field even though the total mass of sludge samples was only 32.5 g. Fortunately, a significant fraction of the water-soluble ^{137}Cs was removed from the sludge solids during this procedure. Once washed and subdivided, the aluminum leaching experiments were conducted on the smaller samples in a glovebox, which provided sufficient shielding and better access to small laboratory equipment.

Hot Cell Operations

Sludge samples from tank 12H at the SRS were shipped to ORNL in January 2002 for testing. This sludge contains a high concentration of aluminum (~30.4% by wt) which was verified by our analysis (see Table 1). The sludge samples were transferred from the stainless steel shipping containers to a single 250-mL, wide-mouthed centrifuge bottle. The sludge was dark brown in color, powdery, and appeared very dry. No visible quantity of sludge remained in the stainless steel container. Then, 25 mL of inhibited water was used to rinse each of the stainless steel containers. This 100 mL of inhibited water was then added to the combined sludge sample. To complete the initial sludge wash, more inhibited water was added to the sludge sample so the mass ratio of wash solution to as-received sludge was 5 : 1. The mixture was agitated on a Thermolyne Maxi-mix II™ vortex mixer and allowed to sit overnight at room temperature. It was then centrifuged for 20 min at 2000 rpm (equivalent to a relative centrifugal force of 850 g). The liquid was decanted to separate it from the settled solids, and the liquid was saved.

A sufficient quantity of inhibited water was added to the sludge to produce a mixture having a concentration of 1.5 g of sludge per 20-mL mixture. This slurry was transferred to a custom-built mixing vessel, which was fabricated from 304 stainless steel schedule 40 pipe components. A Plexiglas cover was fabricated to fit over the vessel. An electrically driven mixer with mechanical speed adjustment was mounted with the mixing vessel on a heavy

Table 1. Analyses of tank 12H sludge for selected components[a]

Component	Amount/g
Process metals	
Al	0.304 g
Ba	0.000771 g
Fe	0.0315 g
Mg	0.00157 g
Mn	0.0199 g
Ni	0.00723 g
Radionuclides	
Co-60	0.000914 mCi
Cs-137	0.0724 mCi
Eu-154	0.0524 mCi
Eu-155	0.00511 mCi

[a] Alpha emitters include ^{all}U, ^{238}Pu, ^{239}Pu, ^{241}Pu, and ^{241}Am but were not quantified in this work.

laboratory stand. Due to previously undetected large particles, the initial attempt to use the mixing vessel was unsuccessful. The sludge slurry (containing all 32.5 g of sludge) was poured through a stainless steel wire mesh with 1.4-mm free space between wires to remove the large particles, which weighed 0.89 g. All of the inhibited water used to clean the storage containers and mesh was added to the slurry, so there were no unaccounted losses of sludge.

Mixing of the sludge slurry then proceeded as expected. Sludge was dispensed into 30-mL centrifuge bottles to produce two sizes of samples. The samples were allowed to settle overnight before the centrifugation step. The samples were centrifuged for 20 min at 3000 rpm (equivalent to a relative centrifugal force of 3600 g), and the clear supernatant was decanted. The samples were weighed before and after decanting to obtain data on the ratio of supernatant to wet sludge. The decanted liquid was added to the previously saved wash solution to produce a composite wash solution with a mass of 518.67 g.

Glove Box

Leaching solutions were prepared in a clean lab and transferred to the glovebox in plastic bottles. Solutions were prepared by weight using one of two calibrated balances, a Mettler model PR8002 or a Mettler model AE163. The amount of water in the sludge samples was estimated from the results from the hot cell portion of the work. This estimate was verified with a sludge sample, which was dried at 60°C. It reached a constant weight after less than 1 day. The temperature was increased successively to 80, 90, and 105°C with no further weight loss. The water content was needed to determine the proper quantity of reagents so the desired solids:liquid ratio and the desired reagent concentrations would be achieved following mixing. Reagents used for preparation of leaching solutions included deionized water, 4.0 *M* NaOH (Baker Analyzed reagent, lot V29501, purity 99 + %), solid $NaNO_3$ (EM Science, lot 41227, purity 99 + %), and TEA [$(HOCH_2CH_2)_3N$, Aldrich Chemical Co., purity 98 + %).

Preweighed stir bars were added to the bottle containing the sludge. The reagents were transferred from a preweighed transfer bottle to be mixed with the sludge, which had previously been weighed during hot cell operations. The lid on the sludge-containing bottle was then sealed, and both it and the transfer bottle were weighed to verify transfer of the proper amount of reagent. The sludge was then well mixed with the aid of the vortex mixer, and the bottle was placed in the sample receiving slot of a preheated aluminum block whose temperature was maintained with a Jenway Model 1103 hot plate/stirrer equipped with an integral temperature controller. The sludge samples were checked at least twice per day to verify that sludge was mixed and that the system was operating properly. When the aluminum blocks were in place on the hot plate/stirrer, it was noted that the rotational speed of the magnetic stirrer was dampened. To ensure that the sludge made good contact with the leach solution, the sludge was mixed manually each time that a sample was checked.

During the leaching tests, the samples were periodically cooled so a small analytical sample could be obtained without loss of the TEA through evaporation. These 1.4-mL aliquots were taken at various time intervals, which ranged from 0.3 day to 11+ days, and did not deplete the liquid phase because of the initial excess. (Because it was anticipated that sampling would remove a portion of the liquid, an excess was selected so that sampling would remove only ~20% of the liquid.) Initially, a mechanical pipettor was used to remove a small portion of the clarified liquid with very little carryover of visible particles. When the settling rate for the solids slowed, the sampling method was changed to support filtering of the leachate. The liquid samples were withdrawn with 5-mL syringes, which were fitted with Tygon® tubing to reach down to the liquid in the centrifuge bottle. While the liquid was in the syringe, the tubing was removed and replaced with a 0.22 μm porosity filter (Millipore Millex®-GS). With one exception, each sample was filtered with ease. In each case, the filtered sample was collected in preweighed polyethylene bottles.

Analytical Methods

Preliminary Characterization of Sludge Samples

It was critical to obtain chemically uniform aliquots of sludge from the hot-cell operations so the chemical analysis of a single washed sludge sample could be used for the other aliquots. The uniformity of the aliquots was verified by weighing the wet sludge after the supernatant was decanted and with on-site measurement of the ^{60}Co, ^{137}Cs, ^{154}Eu, and ^{155}Eu content in each sample. The gamma counting system consisted of an ORTEC model GMX-45220-P-S intrinsic germanium detector, an ORTEC model 672 counting system amplifier, a Canberra Accuspec-A MCA card, and Canberra Genie-2000 spectroscopy software running on an IBM personal computer. With verified uniformity, the rest of the sludge samples were available for the leaching tests, and the amount of required chemical analysis was reduced significantly.

Analysis of the Sludges and Leachates

Because the targeted species for the leaching studies was aluminum, it was the most important component for which to analyze. The methods used also provided data on other metal species with little additional effort. It was also decided that information on the extent of the solubilization of the radionuclides would be important to verify that the aluminum was selectively leached and separated from the high-level waste sludge.

Analytical methods that were used by the Chemical Sciences Division personnel for this study were similar to those previously described in an ORNL report (6). Samples of sludge solids were solubilized by a microwave digestion with nitric acid, based on SW-846 Method 3051, Microwave Assisted Acid Digestion of Sediments, Sludges, Soils, and Oils. This method is considered by regulatory agencies to be a total digestion for metals and radionuclides. However, a simple nitric acid treatment will not dissolve most siliceous materials. Therefore, the residue after the microwave-assisted acid digestion was assumed to be silicon dioxide. The residues may have also contained sodium aluminosilicates, which could decrease the concentration of soluble cesium.

The leachates and wash solutions were filtered (if not already filtered) when obtained from the test. Each sample was analyzed by gamma spectrometry for most of the radionuclides and by Inductively Coupled Plasma–Atomic Emission Spectroscopy (ICP-AES) for metals. Gross alpha and total activity analyses were also performed on the last sample taken from each leaching experiment to ascertain the overall separation of the aluminum from the radionuclides. The analytical error for the metal measurements depends upon the analytical method, the concentration level and dilution factors, and the sample matrix. ICP-AES is a multiple-element measurement technique designed for the best average performance for all elements and is not optimized for any single component. Analysis for anions was not performed.

The standard radiochemical methods for radioactive waste characterization are EPA Method 600/900.0, Gross Alpha and Beta Radioactivity in Drinking Water, and EPA Method 600/901.1, Gamma Emitting Radionuclides in Drinking Water. EPA Method 901.1 was used to determine ^{60}Co, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{155}Eu, and ^{241}Am. Gross beta measurements were obtained by liquid scintillation counting.

RESULTS AND DISCUSSION

Effects of Sludge Washing

Inhibited water was used to transfer the sludge from the shipping containers and to produce a sludge slurry, which was subsequently divided into several uniform aliquots. The equivalent of two washing operations was performed. During the transfer operation, approximately 150 mL of inhibited water was used. The sludge was well contacted with the solution and allowed to sit for 11 days before separation. During the sample preparation, approximately

400 mL of inhibited water was used, and the solution was in contact with the sludge for more than 16 h. In each case, most of the liquid was separated from the sludge by centrifugation and decantation. The clarified liquids were accumulated in one large sample bottle, mixed before sampling and analysis, and referred to as the composite wash solution. The total mass of the collected wash solution, which was identified as COMP-1-S, was 518.67 g.

An analysis of the composite wash solution was used to identify those components that were removed from the sludge. This information may be combined with an analysis of the sludge to estimate the composition of the original as-received sludge and to calculate the fraction of each component removed. Table 2 provides the analysis of the sludge and wash solution and the values calculated from Eqs. (3)–(5). The ratio of dry sludge to wet sludge was estimated at 0.3552 g/g based on the assumption that a negligible amount of sludge was dissolved during washing. The concentration of each component in the dry sludge was calculated from

$$C_{i,dry} = C_{i,wet}/\chi/10^6 \tag{3}$$

where

$C_{i,dry}$ is the concentration of component i in the dry solids, g/g;

$C_{i,wet}$ is the concentration of component i in the wet solids, μg/g; and

χ is the ratio of dry sludge mass to wet sludge mass, 0.3552 g/g.

All metals in the wash solution, other than sodium, originated from the sludge. Therefore, the total amount of each component was calculated by

$$m_i = m_s C_{i,dry} + V_l C_{i,l}/10^6 \tag{4}$$

where

m_i is the total mass of component i in the original sludge, g;

m_s is the total mass of dry sludge solids, g;

V_l is the volume of decanted wash liquid, mL; and

$C_{i,l}$ is the concentration of component i in the wash solution, μg/mL.

The fraction of each component dissolved in the wash solution was calculated with

$$f_i = (V_l C_{i,l}/10^6)/m_i \tag{5}$$

where

f_i is the fraction of each component dissolved by the wash solution.

Table 2. Analysis of sludge and wash solution and calculated fraction of components dissolved

	Sludge[a]			Wash solution[b]		
Analyte metals	Results µg/g	Standard deviation µg/g	Dry basis[c] g/g	Result µg/mL	Standard deviation µg/mL	Fraction dissolved[d] g/g
Ag	2.51E+01	5.00E+00	7.07E−05	<1.70E+00	*e*	*f*
Al	1.08E+05	1.10E+04	3.04E−01	5.00E+00	1.00E+00	2.69E−04
Ba	2.74E+02	2.70E+01	7.71E−04	<2.00E−01	*e*	*f*
Be	<1.14E+00	*e*	*f*	<1.00E−01	*e*	*f*
Ca	1.66E+03	1.70E+02	4.67E−03	3.54E+01	3.50E+00	1.10E−01
Cd	<1.85E+02	*e*	*f*	<1.62E+01	*e*	*f*
Co	<1.14E+02	*e*	*f*	<1.00E+01	*e*	*f*
Cr	3.31E+01	6.60E+00	9.32E−05	<2.10E+00	*e*	*f*
Cu	1.96E+02	2.00E+01	5.52E−04	<7.00E−01	*e*	*f*
Fe	1.12E+04	1.10E+03	3.15E−02	<1.05E+01	*e*	*f*
K	<2.85E+02	*e*	*f*	<2.50E+01	*e*	*f*
Mg	5.59E+02	1.12E+02	1.57E−03	<1.06E+01	*e*	*f*
Mn	7.07E+03	7.10E+02	1.99E−02	<2.00E−01	*e*	*f*
Ni	2.57E+03	2.60E+02	7.23E−03	<5.00E+00	*e*	*f*
Sb	<7.07E+01	*e*	*f*	<6.20E+00	*e*	*f*
Th	1.88E+03	1.90E+02	5.29E−03	<2.51E+01	*e*	*f*
U	<3.86E+02	*e*	*f*	<3.39E+01	*e*	*f*

(*continued*)

Table 2. Continued

	Sludge[a]			Wash solution[b]		
Analyte metals	Results μg/g	Standard deviation μg/g	Dry basis[c] g/g	Result μg/mL	Standard deviation μg/mL	Fraction dissolved[d] g/g
V	<7.07E+01	*e*	*f*	<6.20E+00	*e*	*f*
Zn	<6.58E+02	*e*	*f*	<5.77E+01	*e*	*f*
Radionuclides	Bq/g	Bq/g	Bq/g	Bq/mL	Bq/mL	Bq/Bq
Co-60	1.20E+04	2.00E+03	3.38E+04	*e*	*e*	*f*
Cs-137	5.50E+05	1.00E+04	1.55E+06	6.90E+04	1.00E+03	4.22E−01
Eu-152	3.70E+04	9.00E+03	1.04E+05	*e*	*e*	*f*
Eu-154	6.90E+05	2.00E+04	1.94E+06	*e*	*e*	*f*
Eu-155	6.70E+04	1.80E+04	1.89E+05	*e*	*e*	*f*
Gross alpha	1.00E+07	1.00E+06	2.82E+07	2.60E+03	1.00E+02	1.51E−03
Total activity	3.90E+08	1.00E+07	1.10E+09	8.80E+04	1.00E+03	1.31E−03

[a] Sludge sample #10, analysis is on basis of washed wet sludge.

[b] Composite wash solution sample COMP-1-S.

[c] Concentration corrected to dry mass of sludge (assuming washing dissolved a negligible amount of the as-received sludge).

[d] Based on total mass of dry (as-received) sludge of 31.69 g and a total wash solution mass of 518.67 g that has an assumed density of 1.0 g/mL.

[e] Not applicable because value is below detectable limit.

[f] Not computed because required measured values are below detection limit.

Except for two metals, the concentrations of all process metals in the wash solution were below the detectable limit. The calculated values indicate that approximately 0.027% of the aluminum and approximately 11% of the calcium dissolved in the wash solution. Both values are insignificant, because only a small amount of aluminum was removed and because calcium was a minor constituent in the original sludge. Cesium-137 was the only radionuclide significantly washed from the sludge; approximately 42.2% of the ^{137}Cs was removed. Due to the high activity of ^{137}Cs, its total mass is a negligible fraction of the mass of the sludge. The original assumption, that the mass of material dissolved by the wash solution is negligible, is thus confirmed.

Leaching with Caustic and Caustic Chelating Agents

Six leaching tests were performed with the sludge aliquots. The ratio of sludge to liquid leaching solution was selected so the leachate concentration would not be a limiting factor for the dissolution of aluminum. Two sizes of sludge aliquots were available for the tests. The first two tests were performed with the larger aliquot size, which contained the equivalent of 2.02 g of dry sludge. With the volume available in the test equipment, the maximum feasible ratio of sludge to leaching solution was approximately 1 : 15, so that ratio was used for the experiments. The same ratio was used for leaching tests using the smaller sludge aliquot size, which contained the equivalent of 1.55 g of dry sludge. Leaching solutions were selected based on previous tests and to maintain ionic strength as a controlled variable. A concentration of 3 *m* TEA was selected based on previous studies with simulants. Sodium nitrate was used to maintain a constant ionic strength when NaOH addition was reduced. An operating temperature of 60°C was chosen because that temperature is the lowest operating temperature proposed for the ESP. Table 3 summarizes the actual conditions of the six tests. Each sample was leached for a minimum of 10 days with samples taken at intervals throughout that period.

The liquid leaching solution samples were analyzed for metal cations with ICP-AES, and radionuclides were determined with gamma spectrum, gross alpha, and total activity counting methods. The results for the 6 tests are shown in Tables 4 through 9. A review of the data reveals that antimony, beryllium, cadmium, chromium, cobalt, magnesium, nickel, silver, thorium, uranium, and vanadium were below the detectable limit in all leachates. Therefore, only the detection limits for these 11 elements are listed in Table 4. Aluminum and cesium were detected in all leachates. Other metals and radionuclides appear in the leachates of some of the tests in which TEA was present.

Table 3. Actual conditions of the tests

Test	Temp (°C)	Sludge : Leaching solution (g/g)	Concentration in leaching solution (*m*)		
			NaOH	TEA[a]	$NaNO_3$
1	80	1 : 14.9	3.0	0	0
2	60	1 : 14.9	3.0	0	0
3	60	1 : 14.9	3.0	0.1	0
4	60	1 : 14.9	3.0	3.0	0
5	60	1 : 14.9	0.1	0	2.9
6	60	1 : 14.9	0.1	0.1	2.9

[a] Triethanolamine.

Removal of Aluminum from the Sludge

Aluminum is a major component of tank 12H sludge, accounting for 30.4% by mass. The concentration of aluminum in each leachate sample from the 6 experiments is shown in Fig. 1 as a function of time. Experiments using 3.0 *m* NaOH as the leaching solution at 60°C and 80°C were performed to observe the effect of temperature. At the higher temperature, the aluminum concentration rose more rapidly and achieved a higher final concentration. The decrease in concentration at the last point on the curve for the 80°C test is likely an artifact of an analytical error or due to the formation of a sodium aluminosilicate, which essentially removes aluminum from solution by precipitation. In both cases, it appears that the solubility limit of aluminum was approached. At 60°C, the maximum concentration was reached in approximately 8 to 10 days, while the maximum at 80°C was achieved in approximately 6 days.

Two leaching tests with 3.0 *m* NaOH were performed at a temperature of 60°C using 0.1 *m* TEA and 3.0 *m* TEA. As shown in Fig. 1, the leaching power of the 3.0 *m* NaOH and 0.1 *m* TEA solution lies between the results obtained at 60°C and 80°C using 3.0 *m* NaOH alone. At the end of the 10-day test, the rate of rise in aluminum concentration had decreased, but the equilibrium value had not yet been reached. The concentration of aluminum in the leachate using the 3.0 *m* NaOH and 3.0 *m* TEA solution continued to rise rapidly even after 10 days. The aluminum concentration clearly exceeded the values obtained in all the other tests. The fraction of aluminum dissolved at 60°C increased from approximately 35% using 3.0 *m* NaOH alone to approximately 87% using a combination of 3.0 *m* NaOH and 3.0 *m* TEA.

The last two tests with 0.1 *m* NaOH and 2.9 *m* $NaNO_3$ were performed at a temperature of 60°C; one was conducted using no TEA, and 0.1 *m* TEA was used in the other. As shown in Fig. 1, the aluminum concentration was lower than in the other tests performed with a higher NaOH concentration.

Table 4. Analysis of the leachate solution: Test with 3.0 *m* NaOH at 80°C

Time	1.073 d	2.042 d	4.083 d	6.743 d	10.000 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Ag	<1.70E+00	<1.70E+00	<1.70E+00	<1.70E+00	<1.70E+00
Al	3.33E+03	8.63E+03	1.12E+04	1.18E+04	1.00E+04
Ba	1.00E+00	1.00E+00	8.00E−01	8.00E−01	8.00E−01
Be	<1.00E−01	<1.00E−01	<1.00E−01	<1.00E−01	<1.00E−01
Ca	4.97E+01	1.55E+01	<3.00E+00	1.25E+01	6.40E+00
Cd	<1.62E+01	<1.62E+01	<1.62E+01	<1.62E+01	<1.62E+01
Co	<1.00E+01	<1.00E+01	<1.00E+01	<1.00E+01	<1.00E+01
Cr	<2.10E+00	<2.10E+00	<2.10E+00	<2.10E+00	<2.10E+00
Cu	2.30E+00	2.90E+00	1.50E+00	<7.00E−01	8.00E−01
Fe	<1.05E+01	<1.05E+01	<1.05E+01	<1.05E+01	<1.05E+01
K	3.26E+01	3.04E+01	<2.50E+01	<2.50E+01	<2.50E+01
Mg	<1.06E+01	<1.06E+01	<1.06E+01	<1.06E+01	<1.06E+01
Mn	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Ni	<5.00E+00	<5.00E+00	<5.00E+00	<5.00E+00	<5.00E+00
Sb	<6.20E+00	<6.20E+00	<6.20E+00	<6.20E+00	<6.20E+00
Th	<2.51E+01	<2.51E+01	<2.51E+01	<2.51E+01	<2.51E+01
U	<3.39E+01	<3.39E+01	<3.39E+01	<3.39E+01	<3.39E+01
V	<6.20E+00	<6.20E+00	<6.20E+00	<6.20E+00	<6.20E+00
Zn	<5.77E+01	6.95E+01	<5.77E+01	<5.77E+01	<5.77E+01

(*continued*)

Table 4. Continued

Time	1.073 d	2.042 d	4.083 d	6.743 d	10.000 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	*a*	*a*	*a*	*a*	*a*
Cs-137	7.40E+04	8.50E+04	8.30E+04	8.60E+04	7.90E+04
Eu-152	*a*	*a*	*a*	*a*	*a*
Eu-154	*a*	*a*	*a*	*a*	*a*
Eu-155	*a*	*a*	*a*	*a*	*a*
Gross alpha	*a*	*a*	*a*	*a*	4.00E+03
Total activity	*a*	*a*	*a*	*a*	1.40E+05

[a] Either the measurement was not attempted or the species was not detected.

Table 5. Analysis of the leachate solution: Test with 3.0 m NaOH at 60°C

Time	1.073 d	2.042 d	4.083 d	6.743 d	10.000 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Al	1.29E+03	2.75E+03	4.27E+03	5.13E+03	7.19E+03
Ba	9.00E−01	9.00E−01	9.00E−01	9.00E−01	9.00E−01
Ca	1.81E+01	1.20E+01	1.14E+01	2.37E+01	2.99E+01
Cu	1.40E+00	1.40E+00	1.00E+00	1.80E+00	1.30E+00
Fe	<1.05E+01	<1.05E+01	<1.05E+01	<1.05E+01	<1.05E+01
K	<2.50E+01	<2.50E+01	3.08E+01	2.84E+01	<2.50E+01
Mn	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Zn	7.72E+01	<5.77E+01	<5.77E+01	<5.77E+01	<5.77E+01
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	*a*	*a*	*a*	*a*	*a*
Cs-137	9.10E+04	9.50E+04	9.20E+04	9.10E+04	8.70E+04
Eu-152	*a*	*a*	*a*	*a*	*a*
Eu-154	*a*	*a*	*a*	*a*	*a*
Eu-155	*a*	*a*	*a*	*a*	*a*
Gross alpha	*a*	*a*	*a*	*a*	1.10E+04
Total activity	*a*	*a*	*a*	*a*	1.60E+05

[a] Either the measurement was not attempted or the species was not detected.

Table 6. Analysis of the leachate solution: Test with 3.0 *m* NaOH and 0.1 *m* TEA at 60°C

Time	0.292 d	1.000 d	2.000 d	4.021 d	11.021 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Al	3.97E+02	1.87E+03	3.59E+03	6.51E+03	9.69E+03
Ba	<8.50E+00	<8.50E+00	<8.50E+01	<8.50E+00	<8.50E+00
Ca	8.95E+01	8.05E+01	8.00E+01	7.70E+01	8.15E+01
Cu	9.00E+00	1.45E+01	1.45E+01	1.50E+01	1.65E+01
Fe	<5.25E+01	2.99E+02	3.93E+02	3.95E+02	4.50E+02
K	<1.25E+02	<1.25E+02	<1.25E+02	<1.25E+02	<1.25E+02
Mn	1.25E+01	1.15E+01	1.15E+01	1.15E+01	1.30E+01
Zn	<2.89E+02	<2.89E+02	<2.89E+02	<2.89E+02	<2.89E+02
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	*a*	*a*	*a*	*a*	*a*
Cs-137	4.60E+04	8.70E+04	9.20E+04	8.90E+04	9.30E+04
Eu-152	*a*	*a*	*a*	*a*	*a*
Eu-154	4.40E+01	*a*	*a*	*a*	*a*
Eu-155	*a*	*a*	*a*	*a*	*a*
Gross alpha	*a*	*a*	*a*	*a*	1.40E+03
Total activity	*a*	*a*	*a*	*a*	1.70E+05

[a] Either the measurement was not attempted or the species was not detected.

Table 7. Analysis of the leachate solution: Test with $3.0\,m$ NaOH and $3.0\,m$ TEA at 60°C

Time	0.292 d	1.000 d	2.000 d	4.021 d	11.021 d
Analyte metals	Result μg/mL	Result μg/mL	Result μg/mL	Result μg/mL	Result μg/mL
Al	1.72E+02	1.48E+03	3.33E+03	7.15E+03	1.78E+04
Ba	<8.50E+00	<8.50E+00	<8.00E+00	<8.00E+00	<8.00E+00
Ca	1.05E+02	8.55E+01	9.10E+01	9.00E+01	1.02E+02
Cu	9.00E+00	1.45E+01	1.95E+01	2.10E+01	2.65E+01
Fe	8.25E+01	3.08E+02	5.60E+02	6.95E+02	1.09E+03
K	<1.25E+02	<1.25E+02	<1.25E+02	<1.25E+02	<1.25E+02
Mn	5.10E+01	1.81E+02	3.20E+02	3.93E+02	2.62E+02
Zn	<2.89E+02	<2.89E+02	<2.89E+02	<2.89E+02	<2.89E+02
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	4.90E+01	2.70E+02	6.20E+02	8.90E+02	1.20E+03
Cs-137	2.70E+04	9.10E+04	1.00E+05	1.00E+05	1.00E+05
Eu-152	[a]	[a]	[a]	[a]	[a]
Eu-154	6.50E+02	6.50E+02	4.80E+02	4.70E+02	5.00E+02
Eu-155	1.60E+02	[a]	2.80E+02	[a]	[a]
Gross alpha	[a]	[a]	[a]	[a]	5.30E+04
Total activity	[a]	[a]	[a]	[a]	1.60E+06

[a] Either the measurement was not attempted or the species was not detected.

Table 8. Analysis of the leachate solution: Test with 0.1 *m* NaOH and 2.9 *m* $NaNO_3$ at 60°C

Time	0.292 d	1.000 d	2.000 d	5.000 d	11.917 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Al	2.22E+02	3.37E+02	3.65E+02	3.69E+02	3.70E+02
Ba	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Ca	<3.00E+00	<3.00E+00	<3.00E+00	<3.00E+00	<3.00E+00
Cu	<7.00E−01	<7.00E−01	<7.00E−01	<7.00E−01	<7.00E−01
Fe	1.30E+01	<1.05E+01	<1.05E+01	<1.05E+01	<1.05E+01
K	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01
Mn	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Zn	<5.77E+01	<5.77E+01	<5.77E+01	<5.77E+01	<5.77E+01
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	*a*	*a*	*a*	*a*	*a*
Cs-137	6.90E+04	7.00E+04	6.90E+04	7.20E+04	7.10E+04
Eu-152	*a*	*a*	*a*	*a*	*a*
Eu-154	*a*	*a*	*a*	*a*	*a*
Eu-155	*a*	*a*	*a*	*a*	*a*
Gross alpha	*a*	*a*	*a*	*a*	1.10E+02
Total activity	*a*	*a*	*a*	*a*	1.00E+05

[a] Either the measurement was not attempted or the species was not detected.

Table 9. Analysis of the leachate solution: Test with 0.1 m NaOH, 2.9 m $NaNO_3$, and 0.1 m TEA at 60°C

Time	1.073 d	2.042 d	4.083 d	6.743 d	10.000 d
Analyte metals	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL	Result µg/mL
Al	3.76E+02	7.62E+02	7.84E+02	7.40E+02	<4.95E+00
Ba	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Ca	<3.00E+00	<3.00E+00	3.60E+00	<3.00E+00	<3.00E+00
Cu	<7.00E−01	<7.00E−01	<7.00E−01	<7.00E−01	<7.00E−01
Fe	5.08E+01	8.64E+01	1.00E+02	1.21E+02	<1.05E+01
K	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01
Mn	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01	<2.00E−01
Zn	<5.77E+01	<5.77E+01	1.08E+02	<5.77E+01	<5.77E+01
Radionuclides	Bq/mL	Bq/mL	Bq/mL	Bq/mL	Bq/mL
Co-60	*a*	2.10E+01	2.20E+01	3.30E+01	5.10E+01
Cs-137	6.80E+04	7.00E+04	6.90E+04	6.80E+04	7.20E+04
Eu-152	*a*	*a*	*a*	*a*	*a*
Eu-154	*a*	*a*	*a*	*a*	*a*
Eu-155	*a*	*a*	*a*	*a*	*a*
Gross alpha	*a*	*a*	*a*	*a*	1.20E+01
Total activity	*a*	*a*	*a*	*a*	1.30E+05

[a] Either the measurement was not attempted or the species was not detected.

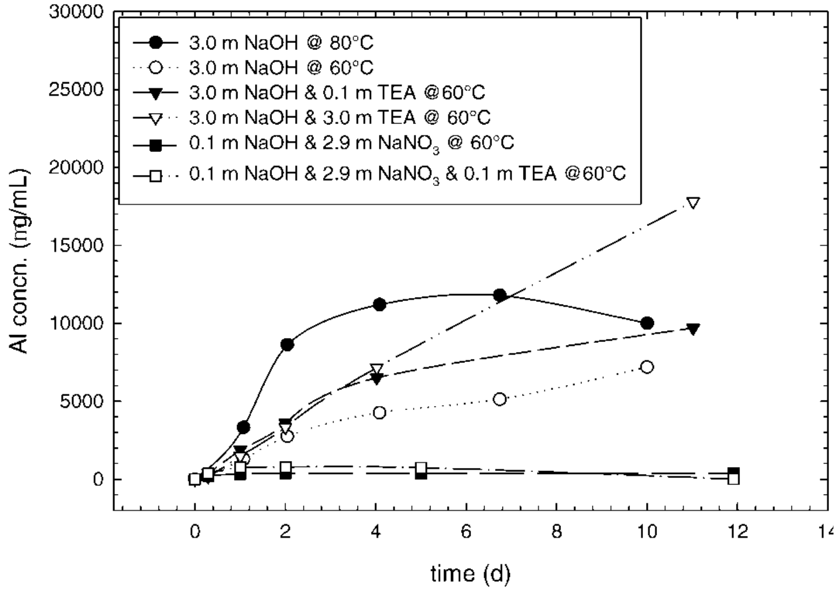

Figure 1. Aluminum concentration in the leachates as a function of time.

Dissolution of Other Metals

Small amounts of process metals were dissolved by the leaching solutions. Their concentrations were generally three or four orders of magnitude smaller than the concentration of aluminum. The concentrations of the metals iron, nickel, and copper showed a correlation with TEA concentration. Generally, the filtered leaching solutions appeared as clear, light-yellow liquids. The only exception was the leachate which contained the high TEA (3.0 *m*) and NaOH (3.0 *m*) concentrations. This solution had a distinct green color, which deepened as the leaching time increased. Because chromium did not dissolve to any appreciable extent, it was thought that the green color arose from dissolution of nickel. Figure 2 shows how the nickel concentration increased during the leaching of sludge using a solution of 3.0 *m* NaOH–3.0 *m* TEA. The nickel concentration saturated at approximately 4 days. Although approximately 13% of the nickel was removed from the sludge, nickel represents only approximately 0.72% of the mass of the dry sludge. Independent tests at ORNL by D.A. Palmer have provided direct evidence that TEA greatly enhances dissolution of nickel and copper oxides (7).

The iron in the sludge was dissolved only when TEA was present. Figure 3 shows that the concentration of iron in the leachate increased with both the NaOH and TEA concentrations at a fixed temperature of 60°C.

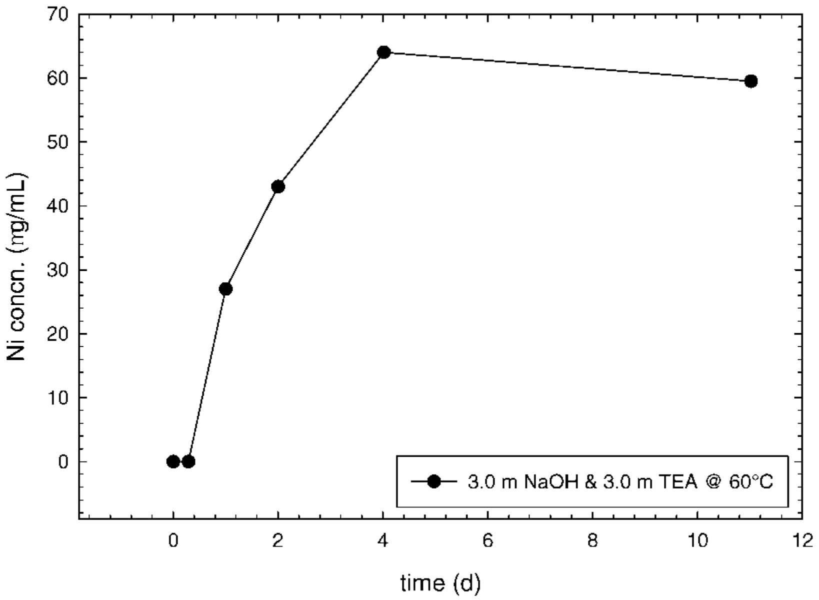

Figure 2. Leaching of nickel at the higher concentration of TEA.

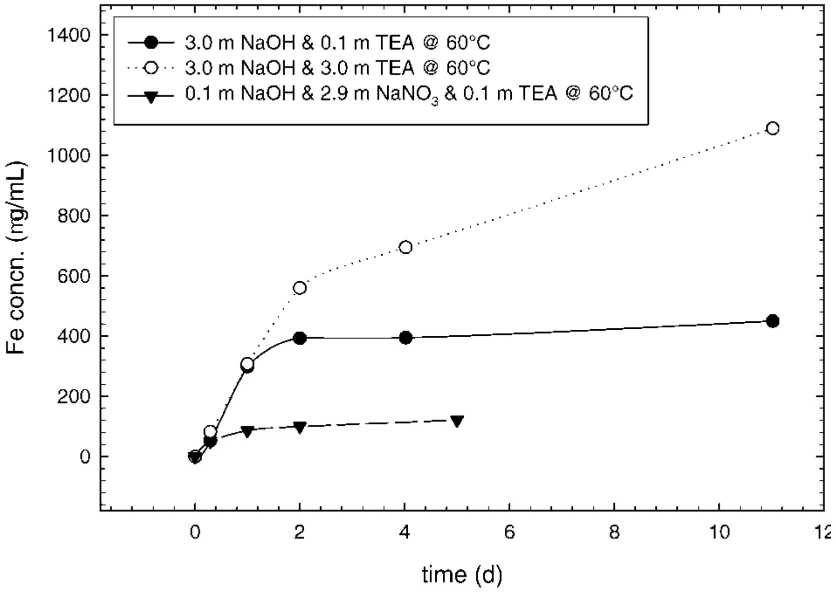

Figure 3. Iron concentration in the leachates, showing increases with the addition of TEA.

Iron concentration reached a limiting value within approximately 2 days when the TEA concentration was 0.1 *m*, but it continued to rise throughout the 11-day experiment when the TEA concentration was 3.0 *m*. Iron represented approximately 3.1% of the mass of dry sludge. When 3.0 *m* NaOH–0.1 *m* TEA was used, approximately 21% of the iron dissolved; however, no detectable amount of iron dissolved when 3.0 *m* NaOH was used alone. The solution containing 0.1 *m* NaOH–2.9 *m* $NaNO_3$ dissolved approximately 5% of the iron when the TEA was present at 0.1 *m*. The solution containing 3.0 *m* NaOH–3.0 *m* TEA dissolved 52% of the iron in the sludge in 11 days, with the expectation that even more iron would dissolve with additional time.

Copper was leached from the sludge at the higher concentrations of NaOH (i.e., 3.0 *m*) and when TEA was present. Figure 4 shows the results from the tests in which copper was measured in the leachates. Concentrations of copper were just above the limits of detection in those tests in which the leaching solution was only caustic. At the baseline condition of 3.0 *m* NaOH and 60°C, approximately 14% of the copper was removed. In the experiment in which 0.1 *m* TEA was used with a high nitrate leach solution (i.e., 0.1 *m* NaOH and 2.9 *m* $NaNO_3$), copper was not detected in the leachate. At NaOH concentrations of 3.0 *m*, the copper concentration saturated in approximately 2 days when the TEA concentration was 0.1 *m*.

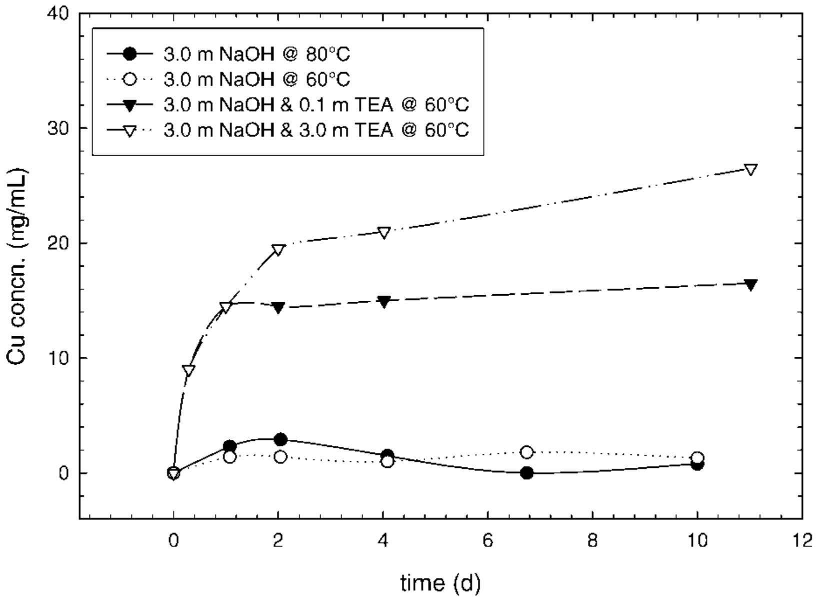

Figure 4. Leaching of copper in 3.0 *m* NaOH, showing increases with addition of TEA.

The copper concentration continued to increase throughout the 11-day test when the TEA concentration was 3.0 *m*. In the latter case approximately 72% of the copper dissolved. However, this quantity is insignificant since the dry sludge was only 0.055% copper by mass.

Dissolution of Radionuclides

The radionuclides detected in the leachate solutions were ^{137}Cs, ^{60}Co, and ^{154}Eu. Because of its lower concentration in the sludge, ^{155}Eu was sometimes, but not always, detected. Concentration profiles for these radionuclides are shown in Figs. 5–7. As expected, ^{137}Cs was found in all leachates. The wet sludge samples contained interstitial liquid containing ^{137}Cs. The results in Fig. 5 show that the equilibrium concentration of ^{137}Cs varies with the concentration of ingredients in the leach solution. The lowest concentrations of ^{137}Cs were measured in the 0.1 *m* NaOH–2.9 *m* $NaNO_3$ leachates, with no significant difference occurring when 0.1 *m* TEA was added. At these conditions approximately 69% of the remaining ^{137}Cs was removed from the washed sludge. Previous studies (8) indicate that up to 72% of the ^{137}Cs may be removed from sludge by washing several times with inhibited water. Since only two washing steps were used during the preparation of the sludge in the present tests, it is reasonable to expect that

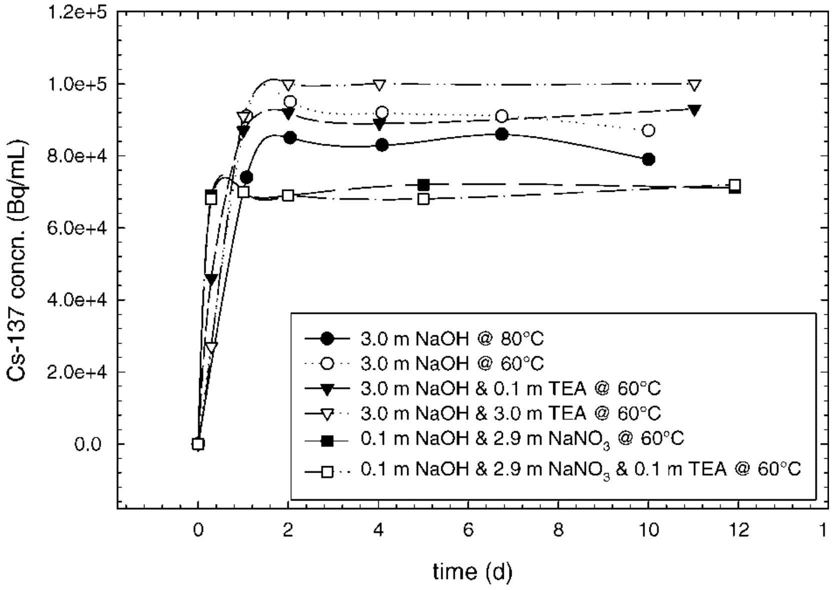

Figure 5. Concentration of ^{137}Cs in leachates of all tests.

part of the ^{137}Cs would have been removed by further washing without using leaching agents. When leaching with 3.0 *m* NaOH alone, increasing the temperature to 80°C decreased the equilibrium concentration of ^{137}Cs compared with the concentrations measured at 60°C. The addition of 0.1 *m* TEA to 3.0 *m* NaOH had no significant effect on the dissolution of cesium at 60°C. The highest concentrations of cesium were measured in the leachate solution with 3.0 *m* TEA. The data indicate that approximately 96% of the ^{137}Cs remaining in the washed sludge was removed. We speculate that breakdown of the sludge particles releases soluble cesium into solution. In all cases, the concentration of cesium rapidly increased and reached equilibrium in approximately 2 days or less.

Cobalt-60 was measured in leachates from two tests in which TEA was present (Fig. 6). The ^{60}Co concentrations are just above the detection limit in samples from the leaching test using 0.1 *m* NaOH–2.9 *m* $NaNO_3$–0.1 *m* TEA. Four of the other five experiments may have resulted in a similar amount of ^{60}Co dissolution. However, differences in the dilution factors during analysis could have reduced the values below the detection limits.

The samples from leaching tests using 3.0 *m* NaOH–3.0 *m* TEA unambiguously show that cobalt is removed from the sludge. The period of most rapid rise in ^{60}Co concentration occurred in the first 2 days, and the concentration continued to rise throughout the 11-day experiment. Approximately 53% of the ^{60}Co was dissolved from the sludge when 3.0 *m* TEA was used. Because

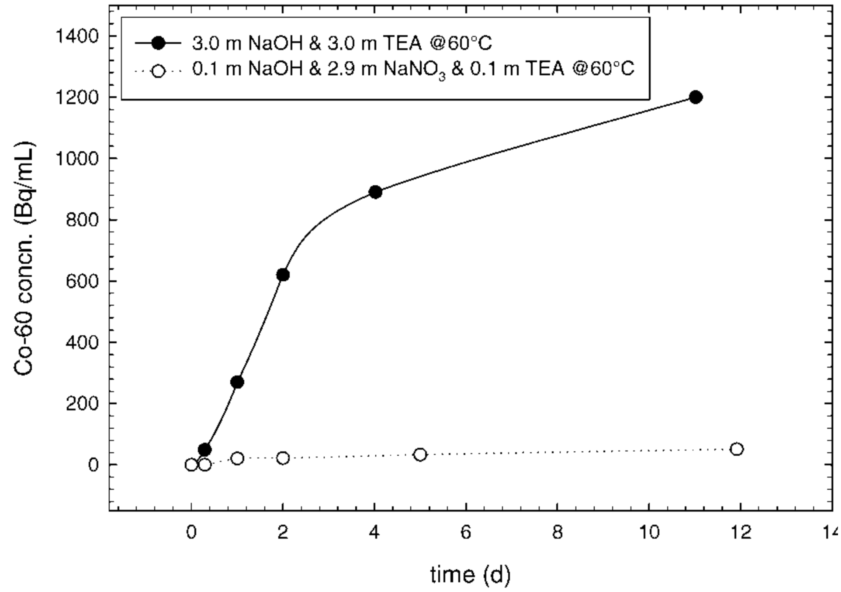

Figure 6. Leaching of ^{60}Co at the higher concentrations of TEA.

the overall cobalt concentration (radioactive and stable) in the sludge is below the detectable limits using ICP-AES analysis, the mass involved is negligible.

A small amount of europium was leached from the sludge when it was treated with a solution of 3.0 *m* NaOH–3.0 *m* TEA. Figure 7 shows the measured ^{154}Eu concentration in the leachates as a function of time. The data indicate that the europium concentration initially rose very rapidly and then decreased to a steady value. One speculative explanation for this early maximum in the concentration profile is that the solubility limit of $Eu(OH)_3$ was reached. An equilibrium concentration of europium was approached in approximately 2 days. At this concentration, only approximately 0.4% of europium present in sludge was dissolved.

CONCLUSIONS

Six tests were performed with leachate concentrations ranging from 0.1 to 3.0 m NaOH, 0 to 3.0 m TEA, and 0 to 2.9 m $NaNO_3$. One test was performed at an operating temperature of 80°C to obtain baseline data, and the remaining five tests were all performed at 60°C. The two tests performed with a leaching solution of 3.0 m NaOH, at 60°C and 80°C,

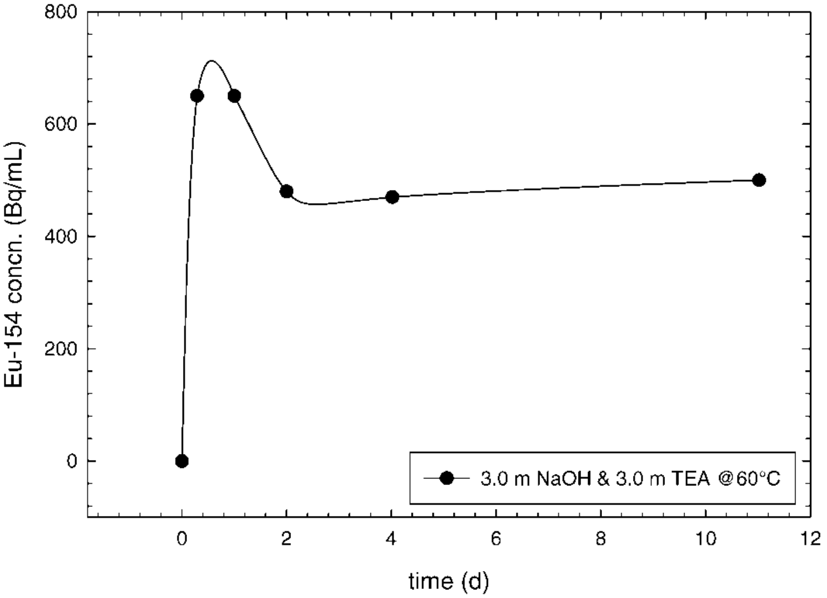

Figure 7. Minor leaching of ^{154}Eu from the sludge at the higher concentration of TEA.

indicated that more aluminum entered the solution at the higher temperature. Equilibrium was achieved at both temperatures within approximately 10 days. The addition of TEA significantly increased the concentration of aluminum in the leachate, and the concentration continued to increase even after 11 days of processing. The fraction of aluminum dissolved at 60°C increased from approximately 35% using 3.0 m NaOH alone to approximately 87% using a combination of 3.0 m NaOH and 3.0 m TEA. Leaching with solvents that had low hydroxide but high nitrate concentrations, either without TEA or with a small (0.1 m) TEA addition, did not significantly dissolve the aluminum.

Use of TEA also increased the solubility of some other sludge components. The fractions of copper, nickel, and iron that were dissolved increased to 72, 13, and 52%, respectively. However, the original fractions of these metals were only 0.055, 0.72, and 3.1%, respectively, of the dry mass of the sludge and therefore represented minor constituents. The presence of nickel in the leachate had a dramatic effect on its color as it changed from light yellow to deep green. By comparison, the baseline leaching with 3.0 *m* NaOH at 60°C removed approximately 14% of the copper; nickel and iron were below the detectable limit.

The TEA also had a measurable effect on the solubility of the radionuclides ^{137}Cs, ^{60}Co, and ^{154}Eu. The presence of ^{137}Cs in the leachates was expected because of the ^{137}Cs-bearing wash solution retained by the wet washed sludge and because of its high solubility in aqueous media. The high nitrate leaches, which were the least effective of the leaching solutions, removed 69% of the ^{137}Cs from the washed sludge, while a combination of 3.0 *m* NaOH and 3.0 *m* TEA removed 96%. Very little ^{60}Co was removed from the sludge except with the use of the 3.0 *m* NaOH–3.0 *m* TEA solution, which removed 53%. This solution was also the only one that dissolved europium; however, only approximately 0.4% of the ^{154}Eu was removed from the sludge.

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