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CAUSTIC LEACHING OF HIGH-LEVEL RADIOACTIVE TANK SLUDGE: A CRITICAL LITERATURE REVIEW

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ABSTRACT

The Department of Energy (DOE) must treat and safely dispose of its radioactive tank contents, which can be separated into high-level waste (HLW) and low-level waste (LLW) fractions. Since the unit costs of treatment and disposal are much higher for HLW than for LLW, technologies to reduce the amount of HLW are being developed. A key process currently being studied to reduce the volume of HLW sludges is called enhanced sludge washing (ESW). This process removes, by water washes, soluble constituents such as sodium salts, and the washed sludge is then leached with 2-3 M NaOH at 60-100°C to remove nonradioactive metals such as aluminum. The remaining solids are considered to be HLW while the solutions are LLW after radionuclides such as ^{137}Cs have been removed. Results of bench-scale tests have shown that the ESW will probably remove the required amounts of inert constituents. While both experimental and theoretical results have shown that leaching efficiency increases as the time and temperature of the leach are increased, increases in the caustic concentration above 2-3 M will only marginally improve the leach factors. However, these tests were not designed to validate the assumption that the caustic used in the ESW process will generate only a small increase (10 Mkg) in the amount of LLW; instead, the test conditions were selected to maximize leaching in a short period and used more water and caustic than is planned during full-scale operations. Even though calculations indicate that the estimate for the amount of LLW generated by the ESW process appears to be reasonable, a detailed study of the amount of LLW from the ESW process is still required. If the LLW analysis indicates that sodium management is critical, then a more comprehensive evaluation of the clean salt process or caustic recycle would be needed. Finally, experimental and theoretical studies have clearly demonstrated the need for the control of solids formation during and after leaching.

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INTRODUCTION

By the end of 1995, the chemical reprocessing of spent reactor fuel and irradiated targets had generated a total of 215,300 m³ of solid high-level waste (HLW) and 158,100 m³ of liquid HLW.¹ The HLW, which is stored in underground tanks, contains the nonvolatile fission products, activation products, residual uranium, plutonium, and other transuranics (TRUs). After the HLW is more than a year old, the radioactivity comes primarily from ¹³⁷Cs in the liquids and ⁹⁰Sr in the solids. The relatively small amount of TRUs can be found in the solids. PUREX reprocessing of spent fuel produces an acidic liquid waste. At Hanford and the Savannah River Site (SRS), this HLW has been neutralized with sodium hydroxide, and sodium nitrite has also been added to prevent corrosion during storage in carbon-steel tanks. Neutralization of the HLW formed hydrated oxides, which precipitated and formed a sludge in the storage tanks. In cases where the neutralized supernatant liquids were concentrated sufficiently by evaporation, sodium nitrite and sodium nitrate crystallized to form salt cakes.

The Department of Energy (DOE) is required by law to treat and safely dispose of its radioactive tank wastes. It is expected that appropriate separation technologies will be used to divide the tank contents into HLW and LLW fractions. At Hanford, the pretreatment or separation steps are expected to generate 78,000,000 kg (78 Mkg) of LLW and 9.3 Mkg of HLW.¹ After these separation or pretreatment steps have been completed, the segregated waste will be immobilized and isolated geologically. After the HLW has been concentrated at Hanford and the SRS, it will be incorporated into borosilicate glass, which is acceptable for permanent disposal in a geologic repository. The LLW will be immobilized in grout or glass and stored on site. Table 1 lists the current volume of tank waste as well as the projected volume and number of HLW canisters.¹

Technology development efforts have focused on volume reductions of the HLW because the costs (per kilogram of waste oxide) of processing, immobilizing, and disposing of HLW are considerably higher than those for its LLW counterpart, as shown in Table 2. The primary incentives to reduce the total volume of HLW glass include a lower overall life-cycle cost and the limited availability of repository space. A process to reduce the volume of HLW should be cost effective if the amount of additional LLW generated is less than 33 times the amount of HLW volume reduction. From Table 2, a doubling of the pretreatment cost for HLW can be paid for with a 1% decrease in HLW volume (i.e., \$28/\$2126).

TABLE 1. TANK INVENTORIES (1995) AND PROJECTED VOLUME OF GLASS CANISTERS

Volume of tank waste in 1995, 1000 m ³	Hanford	SRS
Liquid	89.839	58.700
Solids	143.668	67.800
HLW volume after separations and vitrification	Hanford ^a	SRS ^b
Projected cumulative volume of HLW glass canisters, 1000 m ³	14.277	3.720
Estimated cumulative number of HLW canisters	12,442	5,944

^aBased on assumptions in Reference 1: canister has diameter of 0.61 m and is 4.50 m long (about 2 ft in diam by about 15 ft long). The nominal glass volume is 1.1 m³ with a minimum waste oxide loading of 25 vol % (excluding sodium and silicon).

^bBased on assumptions in Reference 1: canisters are 0.6 m in diam by 3 m long (about 2 ft in diam by about 10 ft long). Each canister is assumed to contain 0.625 m³ of glass made with HLW from the reprocessing of spent nuclear fuel at SRS. The glass incorporates 36 wt % oxides from waste.

TABLE 2. COMPARISON OF LLW AND HLW MANAGEMENT COSTS

	Hanford ^a		International Atomic Energy Agency ^b	
	LLW (\$/kg waste oxide)	HLW (\$/kg waste oxide)	LLW (\$/m ³)	HLW (\$/m ³)
Pretreatment	16	28		
Immobilization	44	728		
Disposal	4	1,370	600–6,800	450,000–1,400,000
Total	64	2,126		

^aReference 4.

^bReference 5.

HLW volume can be reduced by various means, such as loading as much waste into the glass as possible. Technology development efforts are currently under way to improve glass formulations so that maximum waste loadings can be achieved. However, modifications to the glass formulations are expected to be only partially effective. Another approach is to separate the more abundant inert constituents, such as sodium and aluminum, from the radionuclides in the sludges. This process would also remove chromium, sulfate, and phosphate, which can cause vitrification problems.² In 1993, the DOE considered three separation options for the Hanford sludges. The treatment options³ included simple sludge washing, enhanced sludge washing, and advanced separations. Simple sludge washing uses only water or dilute sodium hydroxide with corrosion inhibitors such as sodium nitrite. Enhanced sludge washing (ESW) refers to simple sludge washing that is followed by caustic leaching with 2–3 M NaOH at an elevated temperature. The leached solids are then washed with dilute NaOH to remove the dissolved components and the added NaOH. Advanced separations consist of complete dissolution, if possible, followed by extensive radionuclide separation.

An analysis of the options led to the conclusion that simple sludge washing would result in an unreasonably large volume of HLW and that advanced separation would require extensive technology development and complex facilities. Therefore, ESW was selected as the baseline process for sludge pretreatment. Several assumptions, for example, the minimum wash and leach factors⁴ in Table 3, were made about the ESW process, and verification of these assumptions was required by the DOE. This paper discusses the results of these verification studies as well as other processing issues such as solids formation after leaching.

ANALYSIS OF EXPERIMENTAL RESULTS

Enhanced Sludge Washing

As part of the verification studies, ESW studies^{7–16} have been performed on sludge samples from 34 Hanford tanks by researchers at Pacific Northwest National Laboratory (PNNL), Los Alamos National Laboratory (LANL), and Oak Ridge National Laboratory (ORNL). In the ESW study at ORNL, multiple tests were performed on sludge samples from a few Hanford tanks to evaluate the effects of temperature, leaching time, and caustic concentration. In contrast, the PNNL and LANL researchers have tested numerous sludge

TABLE 3. PERCENTAGE REMOVAL OF KEY ELEMENTS BY SLUDGE WASHING^a

Minimum goal (%)	Test results on a weighted basis (%)			Overall wash and leach factor	
	Wash	Leach	Total	1996	1997
Al	68	14	74	88	60
Cr	64	44	32	77	40–65
PO ₄ ³⁻	74	55	35	90	70

^aReference 6.

samples under a single set of conditions, which were periodically modified as new results were obtained. While the ESW concept appears to be relatively simple, the ESW test procedure is quite complicated. In 1995, the ESW procedure consisted of 18 steps.¹⁰ It should be noted that a small portion of each sludge sample undergoes extensive water washing without caustic leaching and that the remainder of the sample is only slightly washed before being subjected to the caustic leach tests. Therefore, none of the samples were subjected to the entire ESW process.

While the PNNL and LANL procedures, as well as the ORNL tests, were quite reasonable for bench-scale experiments, they were not designed to mimic the expected full-scale operations. For example, the researchers used excessive amounts of water and caustic, as shown in Table 4. Theoretical calculations based on phase equilibria¹⁷ have determined the minimum volumes of water and caustic that must be used to remove the soluble salts and key nonradioactive metals from sludge in Hanford tank S-101. In the test with real waste,¹³ the volumes of water and caustic were seven and five times larger than required by the calculations, respectively; these excessive liquid volumes permitted much shorter wash (0.5–1 h) and leaching (5 h) times. During large-scale operations, the additions of water and sodium should be kept to a minimum so that a reasonable amount of LLW will be generated, and the wash and leaching times can be expected to be longer than those used in the laboratory tests. It is also important to remember that the sludge samples that were leached were not washed extensively first. Note in Table 4 the large increase in the cumulative wash and leach

TABLE 4. AMOUNTS OF WATER AND SODIUM ADDED DURING THE ESW PROCEDURE

Tank	Wet sludge (g)	Dried sludge (g)	Water added, initial wash (mL/g dried sludge)	Water added, last wash (mL/g dried sludge)	Sodium added (g Na/g dried sludge)	Initial sodium (g Na/g dried sludge)
B-106 ^a	8.80	3.53	55	11	1.70	0.49
BX-103 ^a	7.41	5.24	29	41	3.61	0.11
BY-110 ^b	6.23	4.01	28	7	0.98	0.46
C-104 ^a	9.17	4.22	45	32	4.17	0.37
C-105 ^a	2.97	2.58	22	45	3.98	0.0061
S-107 ^b	5.71	3.40	32	33	4.28	0.26
SX-108 ^b	21.06	20.22	21	22	1.13	0.24
SX-113 ^a	3.25	1.63	39	51	6.50	0.034
S-104 ^c	5.14	4.85	18	31	2.16	0.019
S-101 ^c	6.27	4.00	45	18	2.35	0.28
S-101 ^d	10.0	6.4	4.7	7.1	0.32	0.28

^aReference 16.

^bReference 11.

^cReference 13.

^dBased on theoretical calculations using equilibrium constants (Reference 17).

factors from 1996 to 1997, even though only 8 of the 34 tanks were tested in 1997.^{13,16} It is possible that additional ESW tests on other tanks in 1998 can change these factors further. While results of past bench-scale experiments have demonstrated that the ESW will probably exceed the minimum goals for removing key constituents, as shown in Table 3, the tests have not established that the ESW can meet the removal goals and generate an acceptable amount of LLW. It is unwise to draw conclusions about the contents of an entire tank based on a single test with a few grams of sludge.

Sludge Washing Optimization

Several DOE researchers are currently performing parametric studies on ESW to optimize the process for particular sludges and to provide a much more reasonable estimate of the LLW that will be generated by the washes and leaches. During these studies, the effects of process variables such as NaOH concentration, temperature, and leaching time on the efficacy of the caustic leaching process will be determined. The test conditions for these parametric studies are shown in Table 5. The goal of these tasks is to minimize the overall system cost by optimizing the leaching of the HLW to produce the appropriate amounts of wastes. As a starting point, researchers are using the aluminum concentration in the sludge to determine the solid/liquid ratio.

Table 6 shows the effect of leach behavior when the caustic concentration, the total volume of leach solution, the temperature, and the leaching time are increased.⁸ It is important to note that the caustic concentration and leach time in Case 1 were higher than those in the typical ESW tests. In Case 2, the NaOH concentration, the solid/liquid ratio, the temperature, and leach time were increased significantly from the values in Case 1.⁸ Table 7 shows the results of a second comparison, which involved only variations in the caustic concentration.⁸ An analysis of the results indicates that time and temperature play a large role in the increased leaching efficiency with this sludge. However, increases in the caustic concentration will only marginally improve the leach factors. Further support for these observations was obtained through simulations using equilibrium constants on sludge from Hanford tank S-101.¹⁷ The volume of NaOH that would be needed to treat a 1-g sample of S-101 sludge was calculated for the following temperatures and caustic concentrations: (1) 25°C and 1 M NaOH (60 mL), (2) 70°C and 1 M NaOH (20 mL), (3) 25°C and 3 M NaOH (17.3 mL), and (4) 70°C and 3 M NaOH (5.7 mL). It is interesting to note that as the temperature was increased, the number of NaOH moles per gram of sludge was decreased by a factor of 3. However, an increase in the caustic concentration resulted in only a slight decrease in the number of NaOH moles per gram of S-101 sludge. It is important to remember that the caustic will eventually report to the LLW unless other treatment steps are taken to recycle this stream.

Sodium Management

During the initial evaluation of the ESW process, it was assumed in the reference flowsheet¹⁸ that only 10 Mkg of sodium would be added during the ESW process. The total

TABLE 5. CURRENT PARAMETRIC STUDIES FOR
SLUDGE LEACHING

Parameter	Proposed test points
Caustic concentration, M	1, 3
Temperature, $^{\circ}\text{C}$	60, 80, 100
Leaching time, h	5, 24, 72, 168

TABLE 6. EXAMPLES OF EFFECT OF INCREASING NaOH
CONCENTRATION, SOLUTION/SLUDGE RATIO,
LEACH TIME, AND LEACH TEMPERATURE
ON S-104 SLUDGE DISSOLUTION^a

Parameter	Original sludge conc. (mg/g)	Percent removed	
		Case 1	Case 2
Sludge wt., g		1.49	1.10
NaOH conc., M		3.99	6.33
NaOH vol., mL		15	30
Leach temp., $^{\circ}\text{C}$		70	80
Leach time, h		21	126
Component		Percent removed	
		Case 1	Case 2
Al	140	21	96
Cr	3.1	98	99
P	2.5	43	96

^aReference 8.

TABLE 7. EFFECT OF NaOH CONCENTRATION ON
CAUSTIC LEACHING OF S-104^{a,b}

Component	Percent removal	
	3.99 M NaOH	6.33 M NaOH
Al	21	27
Cr	98	97
P	43	49
Cs	97	99

^aReference 8.

^bSludge weight = 1.5 g, NaOH volume = 15 mL; leach temp. = 70°C; leach time = 21 h.

amount of sodium in the sludges in the Hanford single-shell tanks is approximately 8 Mkg.^{6,19} Therefore, the reference flowsheet assumes a 125% increase in the amount that is from the ESW process. However, all of the bench-scale tests have used much larger amounts of caustic than the reference flowsheet assumed, and the average increase in sodium was approximately 1300%. In the case of sludge from Hanford tank S-101, the equilibrium calculations indicated that an increase of 114% in the amount of sodium would be needed to leach all of the aluminum, while the ESW test on the S-101 sludge increased the amount of sodium by 840%. While the initial assumption appears to be reasonable based on the S-101 calculation, the 125% assumption must still be validated. Each 10% increase in the amount of caustic used in the ESW adds \$64,000,000 to the estimated cost to process and dispose of LLW, and a sodium increase of 1300% would more than double the amount of LLW. It must be reiterated that the ESW test conditions were chosen to perform the leaches quickly and efficiently; no implication is made that the full-scale operations would use these same conditions.

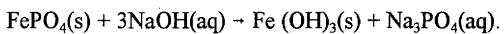
The large amount of caustic that may be required raises the issue of sodium management. DOE researchers have developed two technologies that can limit the amount of sodium to be immobilized as LLW. The first technology involves the clean salt process, which uses

multiple fractional crystallizations of sodium nitrate to produce a decontaminated salt product from liquid waste or supernate, which is primarily sodium nitrate. Most of the radioactivity in the supernate is due to ^{137}Cs . In a test with supernate from Hanford tank AW-101,²⁰ an average cesium decontamination factor (DF) of 21 was obtained for each stage, and a cumulative DF of 4×10^6 was obtained after five stages. No additional separation process was used except for filtration of the initial acidified waste feed to remove undissolved solids. However, before this technology can be used to reduce the amount of sodium to be immobilized, the Environmental Protection Agency must set limits that will permit free release of clean sodium nitrate. The second promising technology involves the use of electrochemical processes, which can generate clean sodium nitrate or sodium hydroxide.^{21, 22}

Awareness of Solids Formation and Control

The chemistry of sludge dissolution and leachate handling is complex. At the end of the ESW process, the remaining solids are considered to be HLW, while the potentially saturated solutions are defined as LLW after cesium has been removed. However, solids will form in the solutions as they are permitted to cool or as they are mixed with other solutions. The leachates can result in the formation of crystalline solid precipitates and gels, which can cause significant processing problems. Therefore, a controlled precipitation may be required since the amount of caustic needed to prevent solids formation, as shown in Figure 1,²³ is unacceptably large at 200 L of 3 M NaOH per kg of aluminum. Clearly, the treatment of LLW liquid and LLW solids from the ESW process will be necessary.

Sludges and supernates containing phosphates offer additional challenges. Phosphate is typically present in the form of insoluble compounds and must be removed by the metathesis of water-insoluble metal phosphates to insoluble hydroxides and soluble phosphates. An example of this reaction is shown for iron phosphate in the following equation:



However, additional problems are generated when an effort is made to solubilize phosphate, since phosphate solubility is very sensitive to temperature. After the leach at elevated temperatures, the phosphate may reprecipitate as a sticky gel as the liquid is cooled. The presence of fluoride in the tank can also greatly increase the complexity. Gels of natrophosphate, $\text{Na}_7(\text{PO}_4)_2\text{F} \cdot 19\text{H}_2\text{O}$, have been observed in leached solutions of sludge from

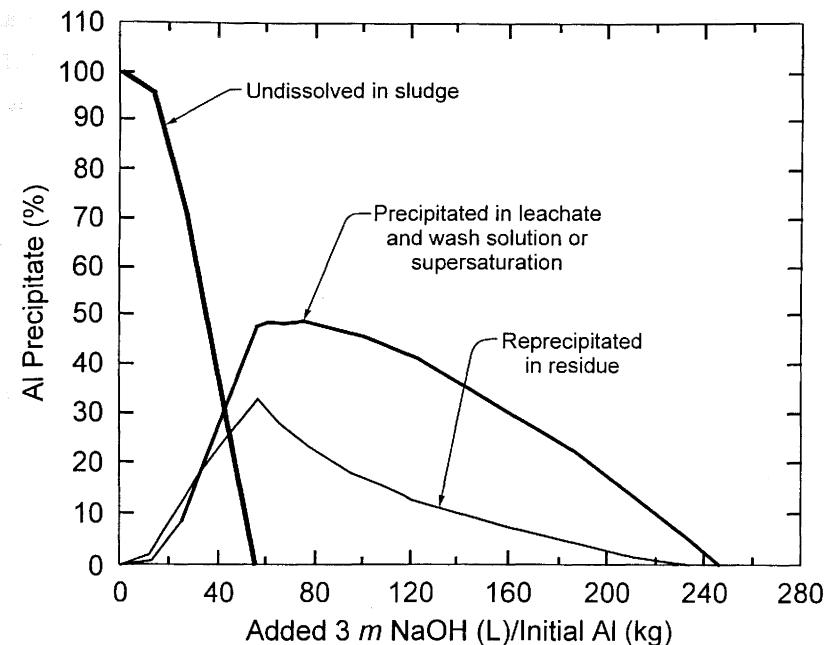


FIGURE 1. Calculation of T-104 caustic leaching: OH^- , initially 3 m at 75°C, ambient temperature, 25°C.

tank T-104. When this substance was wet, it resembled a gel; it was soft and stuck to the sample container walls. On drying, it appeared as a white mass.

Additional solids formations have been observed during sludge washing tests of sludge samples from tanks C-105, C-107, C-108, SX-113, B-202, and C-104.⁸ Upon processing and cooling, solids formed in 7 out of a total of 8 filtered sludge washing solutions (from different tanks) that were tested. With C-105 and C-107 tests, a clear gel-like material formed in the leachates. Wash solutions from the C-105 and C-107 tests also had some clear masses of sorbents. The leachate from the C-108 test developed a mass of gel-like material, as well as some material that appeared to be more crystalline. The wash solutions from the C-108 test developed a small amount of filmy fibrous material. The filtered leachate from the SX-113 test generated a significant amount of particulate material that appeared to be semigelatinous

when suspended. Scanning electron microscopy with energy-dispersive X-ray analysis showed that these particles contained sodium and silicon. These particles could be any of a host of sodium silicates in the $\text{NaOH-SiO}_2\text{-H}_2\text{O}$ system. The test with sludge from B-202 also produced particulate material containing bismuth, a principal component of this sludge.²⁴

Additional evidence of the problems with solids formation is the fact that several cross-site transfer lines are plugged at Hanford. This pluggage, which occurred following the transfer of hot, saturated solutions that were allowed to cool during transfer, has resulted in the abandonment of these transfer lines.

As a result of these observed problems, an alternative flowsheet²³ has been proposed, based on the Bayer process in the aluminum industry. In this flowsheet, the sludge is leached at elevated temperatures and the aluminum, phosphate, and silicates in the leachate LLW stream are intentionally precipitated. This process may be aided by the addition of lime (to precipitate the anions) and flocculent, as shown in Figure 2. The resulting solids will be transported to the low-activity waste stream for immobilization. Although this refinement is not part of the current Hanford flowsheet, it is under consideration.

CONCLUSIONS

Numerous bench-scale tests have shown that the ESW will probably remove the required amounts of aluminum, phosphate, and chromium. Experimental and theoretical results have shown that leaching efficiency improves as time and temperature are increased while increases in the caustic concentration will only marginally improve the leach factors. However, it has also been assumed that the caustic added during the ESW process will generate only a small increase (10 Mkg) in the amount of LLW. The bench-scale experiments were not designed to validate this assumption; rather, the test conditions were selected to maximize leaching in a short period. Theoretical calculations indicated that the amount of LLW from the ESW process appears to be reasonable. However, a more detailed study on the amount of LLW from the ESW process is needed. The findings should, at least, encourage remediation personnel to limit their use of caustic. If the LLW analysis indicates that sodium management is critical, then a more comprehensive evaluation of the clean salt process or caustic recycle would be needed. Finally, experimental and theoretical studies have clearly demonstrated the need for the control of solids formation. Solids formation can be particularly difficult for

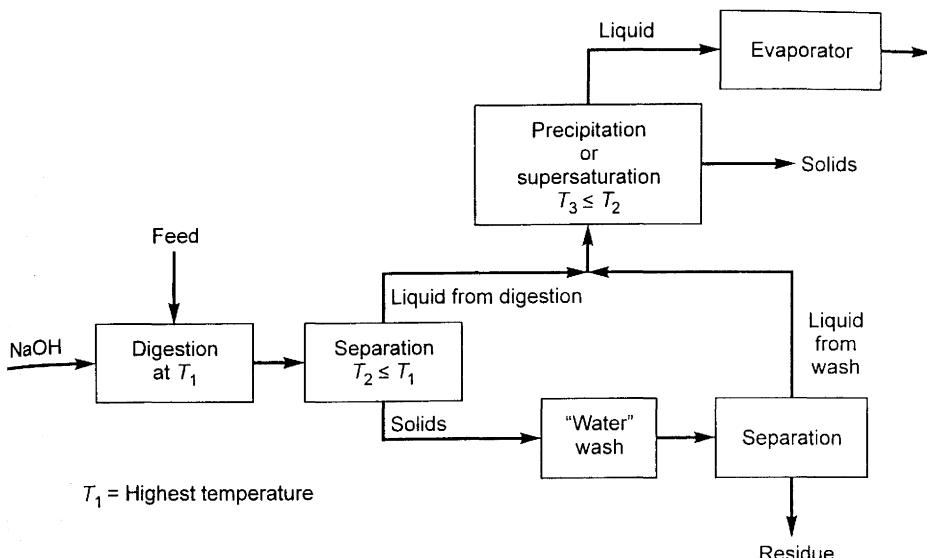


FIGURE 2. Simplified diagram of enhanced sludge washing and the Bayer process.

wastes containing significant concentrations of phosphate. Solids can hamper downstream treatment steps such as cesium removal and, in the extreme cases, could plug lines. Methods to address these issues are needed.

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