

LEACHING METHODS FOR PRETREATING HANFORD TANK SLUDGES

G. J. Lumetta^(a) and B. M. Rapko
Pacific Northwest National Laboratory^(b)
P.O. Box 999; MSIN P7-25
Richland, Washington 99352

P. Sylvester, L. A. Rutherford, Jr., A. Gonzalez-Martin, and J. Kim
Lynnntech, Inc.
7610 Eastmark Drive, Suite 202
College Station, Texas 77840

ABSTRACT

Hanford tank sludges need to be pretreated to reduce their volume before vitrification. Caustic leaching is the baseline method for pretreating these sludges. The caustic leaching process is designed to remove aluminum, chromium, phosphate, and sulfate. Removing these nonradioactive components will reduce the immobilized high-level waste (IHLW) volume. Aluminum is removed by dissolving aluminum oxides/hydroxides as sodium aluminate. We presume that phosphate and sulfate are removed by insoluble phosphates and sulfates metathesizing to the corresponding metal hydroxides and soluble sodium phosphate and sulfate. Earlier we presumed that chromium would be removed by dissolving Cr(III) as $\text{Cr}(\text{OH})_4^-$, but experimental evidence suggests that removing Cr by this mechanism is unfavorable. In this paper, we summarize caustic leaching studies on actual tank waste samples. The behaviors of Al and Cr as a function of waste type and processing conditions will be discussed.

Recent studies suggest that for some Hanford sludges, the Cr content will define the IHLW volume; therefore, removing Cr from these sludges would further decrease the IHLW volume. We have been investigating oxidative leaching methods for removing Cr from Hanford wastes. In these methods, Cr(III), which has low solubility under alkaline conditions, is converted to soluble chromate ion. Chromium removal has been demonstrated at a laboratory scale for several different Hanford tank sludges. The most effective leaching agent investigated to date is permanganate, which readily oxidizes Cr(III) to Cr(VI) under alkaline conditions. In many cases, this method removes more than 90% of the Cr. Other oxidants, including oxygen, ozone, and ferrate ion, have also shown promise for removing chromium. This paper will also summarize the status of the oxidative chromium leaching technology.

INTRODUCTION

During the past few years, the primary mission at the U.S. Department of Energy's (DOE's) Hanford Site has changed from producing plutonium to restoring the environment. Large volumes of high-level radioactive wastes (HLW), generated during past Pu production and other operations, are stored in underground tanks onsite. DOE plans to remediate the Hanford tank farms by retrieving, pretreating, immobilizing, and disposing of the wastes. The tank wastes will be partitioned into high-level and low-level fractions. The low-level waste (LLW) will be processed to remove ^{137}Cs and ^{99}Tc (and possibly other radionuclides), and then it will be immobilized in a glass matrix and disposed of by shallow burial onsite. The HLW will be immobilized in a borosilicate glass matrix cast inside metallic canisters; the resulting glass canisters will then be disposed of in a geologic repository (1). Because of the expected high cost of HLW immobilization and geologic disposal, pretreatment processes will be implemented to reduce the volume of immobilized high-level waste (IHLW). The volume of IHLW is dictated by the waste oxide loading limits and in some cases on the presence of certain waste components (e.g., Cr) that

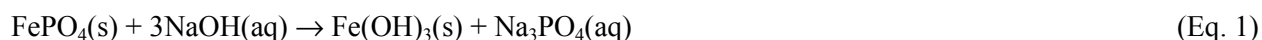
WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

are not well tolerated in the vitrification process. Pretreatment processes are designed to reduce the amount of waste oxides and remove the problematic components when required.

Dilute hydroxide washing is the minimum pretreatment for Hanford tank sludges. The sludge is simply mixed with dilute (0.1 M or less) NaOH, and then some sort of solid/liquid separation is performed. This is meant to remove water-soluble sludge components (mainly sodium salts) from the HLW stream. Dilute hydroxide is used rather than water to maintain the ionic strength high enough that colloidal suspensions are avoided.

Caustic leaching is the baseline method for pretreating Hanford tank sludges. The sludges are leached with aqueous NaOH solution, and then the leached sludge is washed with dilute NaOH to remove excess Na and any dissolved waste components. Leaching with NaOH is expected to remove a large fraction of the Al, which is present in large quantities in Hanford tank sludges. The Al will be removed by converting aluminum oxides/hydroxides to sodium aluminate. The chemistry involved in this process is well known, as it has formed the basis for the Bayer process in the aluminum industry (2).

A significant portion of the P is also expected to be removed from the sludge by metathesis of water-insoluble metal phosphates to insoluble hydroxides and soluble Na₃PO₄. An example of this is shown for iron(III) phosphate in the following equation.



Similar metathesis reactions can occur for insoluble sulfate salts, allowing the removal of sulfate from the HLW stream.

Based on its known amphoteric behavior (3), Cr(III) is expected to be removed by caustic leaching according to the following equation:



However, recent studies conducted at the Pacific Northwest National Laboratory (PNNL) have suggested that the behavior of Cr in the caustic leaching process is more complex (4). Chromium(III) hydroxide does dissolve appreciably in high-caustic solutions at room temperature, but heating such solutions causes gytanaite, syn-(CrOOH), to precipitate. This precipitate does not readily redissolve in aqueous caustic media. This observation is consistent with previous reports of low solubility of Cr(OH)₃ in acidic and near-neutral solutions at elevated temperature (3). The precipitation of CrOOH might explain the low removal efficiencies observed for chromium during caustic leaching tests with actual tank waste and the failure to observe any Cr(III) in the leachates (4–6). Because caustic leaching has not removed chromium very well from certain Hanford tank sludges, methods are being investigated to improve chromium removal from these sludges. These methods have primarily focused on oxidation to Cr(VI), which is soluble in NaOH solutions.

The paper discusses recent investigations of both caustic leaching of Hanford tank sludges and oxidative leaching of Cr from Hanford sludges.

CAUSTIC LEACHING

A vast amount of caustic leaching data have been accumulated, with sludges from over 30 Hanford tanks investigated (4, 5, 7-14). Due to the limited scope of this paper, a comprehensive review of all these data

is not possible. The behavior of the specific sludge components Al, Cr, P, Na, and radionuclides when subjected to a standard set of leaching conditions has been summarized elsewhere (5, 7). In this paper, we will focus on the results of parametric leaching studies that were recently performed on several Hanford tank sludges. This discussion is organized in terms of the behavior of Al and Cr.

Aluminum

Aluminum is a major constituent of Hanford tank sludges, and removing this element from the HLW stream will significantly reduce the IHLW volume. Aluminum exists in numerous different chemical phases in the Hanford sludges (7). Because of this, it is very difficult to predict the response of Al to caustic leaching. Parametric leaching studies are required to determine the optimal process conditions needed to reach the desired Al removal target for the specific sludge being treated.

We have previously reported the increase in Al removal from Hanford REDOX sludges by increasing the duration of the caustic leach (7). This was attributed to boehmite (AlOOH) being the predominant Al-containing phase in these particular sludges. The boehmite in the REDOX sludges behaves in a consistent manner when subjected to caustic leaching. Thus, the response of Al in the caustic leaching of REDOX sludges is reasonably predictable. Unfortunately, most of the other types of Hanford tank sludges contain mixtures of Al phases. The Al in these mixtures can behave in unpredictable ways. Furthermore, sludges from tanks alleged to contain the same waste type can behave differently. This is illustrated with the cases of sludges from Tanks BX-110 and BX-112, which contain waste from the first cycle of the bismuth phosphate process for Pu separation and evaporator bottoms.

Figure 1 shows the Al concentration as a function of time during the caustic leaching of BX-110 sludge. As expected, the Al concentration generally increased with time. This is especially evident for samples leached with 1 M NaOH. When leached with 3 M NaOH, Al dissolved rapidly with the Al concentrations reaching >90% of their final values within the first 5 h of leaching. These results indicate that, provided sufficient time is allowed, leaching the BX-110 sludge with 1 M NaOH at 60°C is as effective at removing Al as leaching with 3 M NaOH at 95°C. Caustic leaching effectively removed Al from the BX-110 solids; in every case, $\geq 95\%$ of the Al was removed after leaching for 168 h.

Figure 2 shows the Al concentration as a function of time during the caustic leaching of BX-112 sludge. For the most part, the hydroxide concentration dependence is as expected. That is, Al removal improves with increasing hydroxide concentration, although the increases are small. However, the Al concentration *decreased* as a function of time, which is opposite to the behavior observed for the BX-110 sludge. Furthermore, Al removal decreased with increasing temperature—a trend contrary to what was expected. We hypothesize that this is caused by the formation of aluminosilicate minerals. Two pieces of evidence support this hypothesis. First, the general trends for Si removal parallel those for Al. That is, the Si removal decreases with increasing temperature. Second, the Al and Si concentrations in the leach solutions both decrease over time. This was observed for virtually all conditions examined. The decrease in Al concentration occurred more rapidly at 80 and 100°C; the decreases seen at 60°C were more gradual. An increased rate of formation of aluminosilicates at the higher temperatures would explain these observations. In all cases investigated, the Si removal was poor, but it was especially so at 100°C. The Al/Si molar ratios in the leached solids were very similar for each leaching condition investigated. The average Al/Si molar ratio was 0.78, with a standard deviation of 0.1. Transmission electron microscopy analysis of the BX-112 solids remaining after leaching with 3 M NaOH at 100°C for 168 h revealed the presence of sodium aluminosilicate minerals. Caustic leaching was less effective at removing Al from the BX-112 sludge than from the BX-110 sludge, with 50 to 75% being removed from the BX-112 solids.

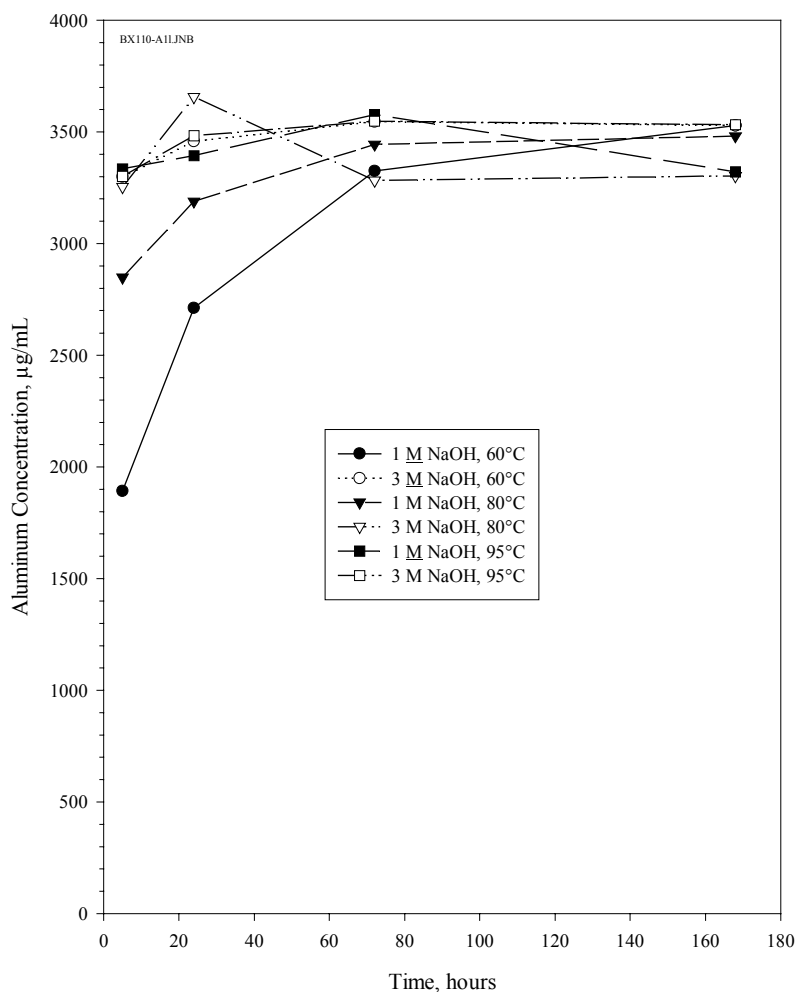


Figure 1. Aluminum Concentration as a Function of Time During the Caustic Leaching of Tank BX-110 Sludge. The sludge was washed with 0.1 M NaOH before leaching with caustic.

Chromium

The presence of Cr in the HLW solids might adversely affect the HLW immobilization process, producing an unacceptable mass of IHLW. Indeed, recent projections for the mass of IHLW produced in processing the Hanford tank wastes suggest that Cr could be the limiting constituent. That is, improved removal of Cr might be needed to further reduce the mass of IHLW. All the parametric caustic leaching tests performed to date indicate that Cr removal increases with increasing leaching time, temperature, and caustic concentration, although the actual fraction of Cr removed varies widely from one sludge to another.

Figures 3 and 4 show the Cr concentration as a function of time during the caustic leaching of BX-110 and BX-112 sludges, respectively. The general trend of increasing Cr concentration with increasing temperature, caustic concentration, and time is consistent between these two sludges. The fraction of Cr removed is also consistent between these particular sludges. Approximately 50% of the Cr was removed from the solids at 1 M NaOH and 60°C, and 80 to 90% was removed at 3 M NaOH and 95 to 100°C.

We have observed during the caustic leaching tests with actual tank waste that the Cr in the NaOH leach solutions is predominantly in the +6 oxidation state. This is true even when the sludge has been washed to remove Cr(VI) from the interstitial liquid before the caustic leaching step. Thus, the Cr removal is likely sensitive to the redox chemistry (i.e., the presence of oxidants) of the waste itself and of the washing or leaching solutions.

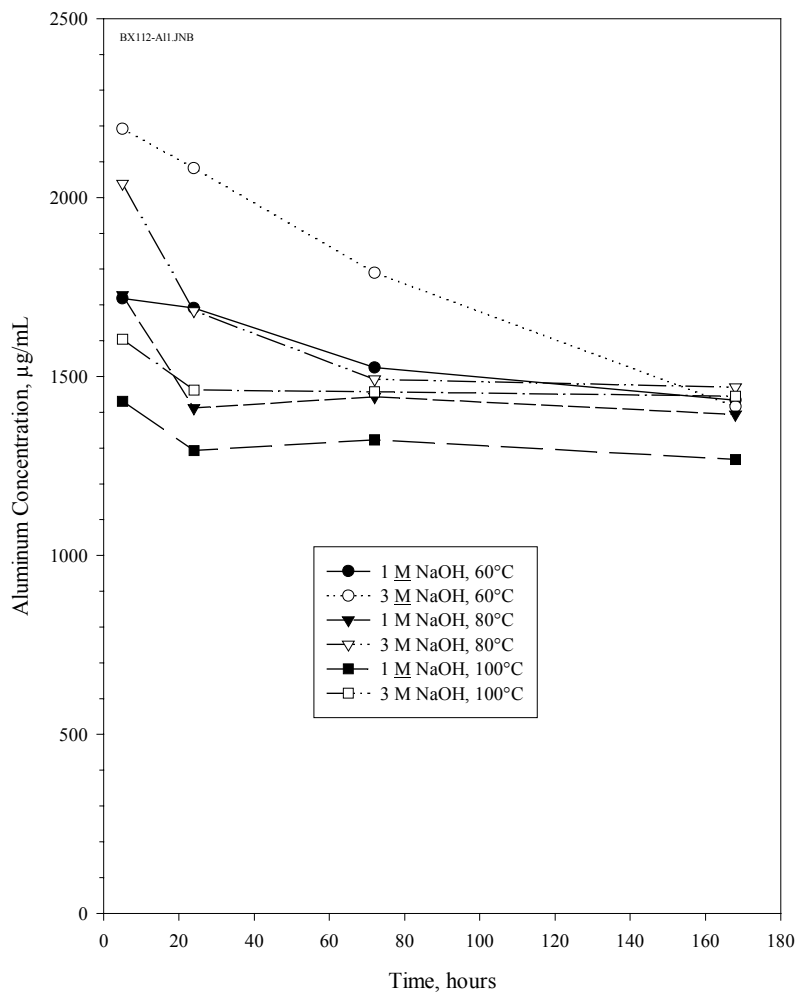


Figure 2. Aluminum Concentration as a Function of Time During the Caustic Leaching of Tank BX-112 Sludge. The sludge was washed with 0.1 M NaOH before leaching with caustic.

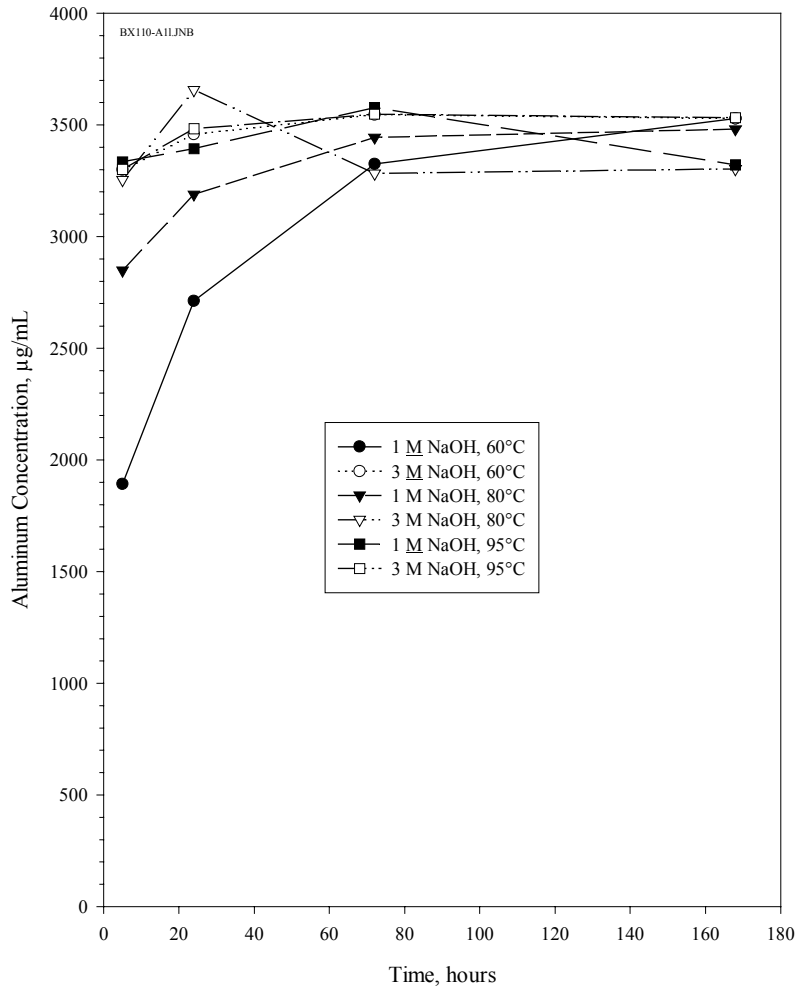


Figure 3. Chromium Concentration as a Function of Time During the Caustic Leaching of Tank BX-110 Sludge. The sludge was washed with 0.1 M NaOH before leaching with caustic.

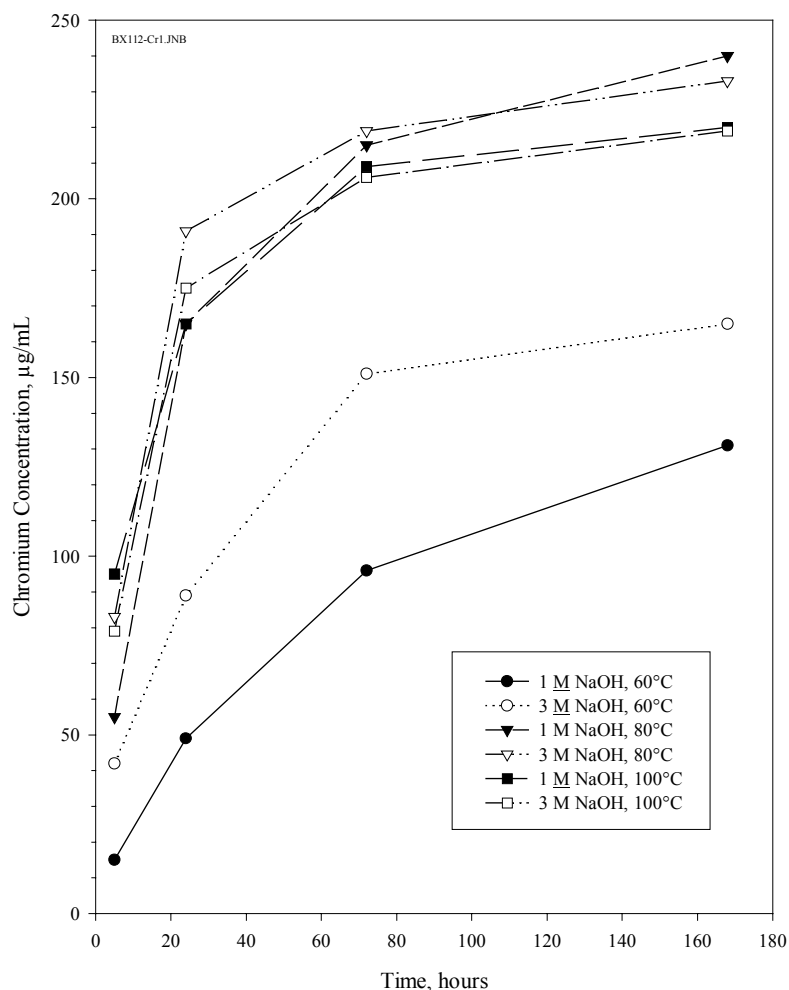


Figure 4. Chromium Concentration as a Function of Time During the Caustic Leaching of Tank BX-112 Sludge. The sludge was washed with 0.1 M NaOH before leaching with caustic.

OXIDATIVE CHROMIUM LEACHING

We have been investigating oxidative leaching methods for removing Cr from Hanford tank sludges for several years. These methods rely on converting insoluble Cr(III) to Cr(VI), which is soluble under alkaline conditions.



Although a number of oxidants have been screened for this purpose, testing with actual tank waste has focused on MnO_4^- , oxygen (O_2), and ozone (O_3). Results of tests conducted through FY 1997 were summarized in Reference 15. This discussion will focus on more recent tests (16). Very recently, we have also investigated the efficacy of ferrate ion at leaching Cr, which we will also discuss in this paper. The FeO_4^{2-} used in the latter tests was generated electrochemically using proprietary technology developed at Lynntech, Inc.

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

Table I summarizes the results of recent oxidative Cr leaching tests. The samples used were all pretreated in some manner (e.g., washed or washed and leached) before the oxidative leaching test. The Cr-removed values given in the table represent the fraction of Cr removed from the pretreated sample. Because some Cr was removed during the pretreatment of the samples, the values in the table represent the minimum Cr removed from the original sludge samples. It should also be noted that the specific reaction conditions varied between different sets of tests, so direct comparison of results is not always straightforward. The test conditions used were not necessarily the optimal conditions for the process; further work is needed to optimize process conditions.

As was observed in previous tests (15), MnO_4^- was generally effective at removing Cr from Hanford tank sludges. For example, treatment of the U-108 and U-109 solids (which contain considerable Cr) resulted in nearly quantitative removal of Cr. The oxidation of Cr(III) with MnO_4^- is rapid—the reaction is generally complete within a matter of hours. The efficacy of MnO_4^- is generally improved by increasing the NaOH concentration.

Ferrate ion was essentially as effective as MnO_4^- at removing Cr for the S-107 sludge, with ~80% removal. Varying the temperature and the FeO_4^{2-} :Cr ratio had no effect on the results with FeO_4^{2-} . The same fraction of Cr was removed at 50°C and a 5-fold molar excess of FeO_4^{2-} as was achieved at 70°C and a 10-fold molar excess. Like MnO_4^- , oxidation of Cr(III) with FeO_4^{2-} is rapid.

One disadvantage of using MnO_4^- and FeO_4^{2-} is that reaction of these oxidants results in addition of Mn and Fe, respectively, to the HLW sludge solids. Although these elements are better tolerated in the HLW immobilization process, it could be advantageous to use oxidants that add no mass to the HLW solids. Ozone can be used to leach Cr from Hanford sludges, with Cr removals comparable to those achieved with MnO_4^- . However, processing with ozone might be problematic due to its corrosive nature. Interestingly, O_2 shows significant promise in removing Cr from the Hanford sludges. Striking examples of this are the U-108 and U-109 solids. About 95% of the Cr can be removed from these solids by leaching with O_2 at 3 M NaOH at 80°C. This Cr removal is nearly equal to that for MnO_4^- . The Cr(III) oxidation with O_2 is very dependent upon the NaOH concentration, with much better removals being achieved at 3 M NaOH compared to 1 M.

The major advantages of using O_2 to oxidize Cr are 1) no mass added to the HLW solids and 2) process simplicity. One major disadvantage is that the oxidation is much slower than that of MnO_4^- and FeO_4^{2-} . As mentioned above, during caustic leaching tests, the amount of Cr removed increases with increasing leaching time (Figures 3 and 4). This is likely caused by adventitious O_2 in the leaching system. The rate of Cr removal can likely be improved by intentionally adding O_2 into the system. Thus, modification of the baseline caustic leaching process by sparging with CO_2 -free air during leaching would likely significantly improve process performance. This modification would likely have little or no impact on the overall sludge pretreatment costs.

Some transuranic (TRU) element dissolution was observed in most of the oxidative leaching tests, especially when MnO_4^- was used as the oxidant. However, the resulting low-level waste form (assuming 20 wt% Na_2O) would generally be within the NRC Class A LLW TRU criteria of 10 nCi/g. The only exception to this was for leaching of the U-109 solids with MnO_4^- at 3 M NaOH. In that case the TRU content of the LLW was projected to be ~11 nCi/g, which is essentially at the Class A limit, but well within the Class C limit of 100 nCi/g. For leaching with O_2 or FeO_4^{2-} , the projected TRU content of the LLW form was less than 1 nCi/g in all cases.

CONCLUSIONS

Leaching methods are being implemented to reduce the volume of IHLW resulting from the processing of Hanford tank sludges. Caustic leaching represents the baseline method. The efficacy of caustic leaching for removing Al, Cr, and P varies from one sludge to another. Parametric process testing should be performed on individual wastes to be processed so that the optimal process conditions can be established.

For sludges with high Cr content, oxidative leaching methods can be implemented. Permanganate and ferrate are very effective at removing Cr from Hanford sludges, and these reagents react rapidly with Cr(III) to form the soluble chromate ion. Air sparging during caustic leaching is a potentially promising alternative to using permanganate or ferrate. Although some TRU elements are solubilized during oxidative leaching, this should have no adverse effects on the LLW form.

ACKNOWLEDGEMENTS

This work was funded by the U.S. Department of Energy (DOE) through the Office of Science and Technology (Tanks Focus Area, TFA) and the Tank Waste Remediation System (TWRS). Additional funding was provided by DOE through Contract No. DF-FG03-97ER82421 with Lynntech, Inc. The authors thank the following individuals from the TFA organization: D. Geiser, C. P. McGinnis, J. Hennig, M. C. Vargas, and T. L. Stewart. The authors thank the following individuals from the TWRS organization for their assistance and guidance: R. A. Kirkbride, G. T. MacLean, R. M. Orme, and K. M. Eager.

The authors gratefully acknowledge the technical assistance of J. J. Wagner, L. R. Greenwood, R. T. Steele, and F. V. Hoopes and other personnel within the PNNL Radiochemical Processing Group. We also thank W. C. Cosby and C. H. Delegard for reviewing this document. Finally, the authors thank W. F. Bonner and E. J. Hirschi for their project-management support.

Table I. Summary of Oxidative Chromium Leaching

Tank	Initial Chromium Concentration (wt%)	Sample Pre-treatment	Oxidant	Oxidant:Cr Molar Ratio	[NaOH], M	Time/Temp	Chromium Removed, %		
S-107	1.3	(a)	MnO ₄ ⁻	2	0.1	(d)	58		
		(a)	MnO ₄ ⁻	2	3	(d)	80		
		(a)	O ₃	Excess	0.1	(d)	84		
		(a)	O ₃	Excess	3	(d)	87		
		(a)	O ₂	Excess	0.1	(d)	8		
		(a)	O ₂	Excess	3	(d)	65		
		(a)	Blank (Ar)	N/A	0.1	(d)	5		
		(a)	Blank (Ar)	N/A	3	(d)	(k)		
		(b)	FeO ₄ ²⁻	5	2.4	(e)	77		
		(b)	FeO ₄ ²⁻	7.5	2	(e)	75		
		(b)	FeO ₄ ²⁻	10	1.7	(e)	60		
		(b)	Blank	N/A	3	(e)	28		
		(b)	FeO ₄ ²⁻	5	2.4	(f)	76		
		(b)	FeO ₄ ²⁻	7.5	2	(f)	75		
		(b)	FeO ₄ ²⁻	10	1.7	(f)	77		
		(b)	Blank	N/A	3	(f)	39		
		SX-108	0.58	(a)	MnO ₄ ⁻	1	0.1	(g)	29
				(c)	MnO ₄ ⁻	1	3	(g)	64
				(a)	MnO ₄ ⁻	2	0.1	(d)	22
				(a)	MnO ₄ ⁻	2	3	(d)	45
(a)	O ₃			Excess	0.1	(d)	42		
(a)	O ₃			Excess	3	(d)	33		
(a)	O ₂			Excess	0.1	(g)	4		
(c)	O ₂			Excess	3	(g)	71		
U-108	12.6			(c)	MnO ₄ ⁻	1	0.1	(h)	97
		(c)	MnO ₄ ⁻	1	3	(h)	100		
		(c)	O ₂	Excess	0.1	(i)	5		
		(c)	O ₂	Excess	3	(i)	94		
		(c)	Blank (Ar)	N/A	0.1	(i)	1		
		(c)	Blank (Ar)	N/A	3	(i)	5		
U-109	20.5	(c)	MnO ₄ ⁻	1	0.1	(h)	96		
		(c)	MnO ₄ ⁻	1	3	(h)	99		
		(c)	O ₂	Excess	0.1	(j)	14		
		(c)	O ₂	Excess	3	(j)	97		
		(c)	Blank (Ar)	N/A	0.1	(j)	2		
		(c)	Blank (Ar)	N/A	3	(j)	5		

(a) Washed with 0.01 M NaOH, leached with 3 M NaOH solution, and again washed with 0.01 M NaOH
 (b) Washed with 0.1 M NaOH
 (c) Washed with 0.01 M NaOH
 (d) 24 h at room temperature, then 7 h at 80°C
 (e) 24 h at 50°C
 (f) 24 h at 70°C
 (g) 24 h at room temperature, then 300 h at 80°C
 (h) 24 h at room temperature, then 140 h at 80°C
 (i) 24 h at room temperature, then 70 h at 80°C
 (j) 24 h at room temperature, then 130 h at 80°C
 (k) Could not be calculated due to analytical difficulties.

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

REFERENCES

1. R.M. Orme, A.F. Manuel, L.W. Shelton, and E.J. Slaathaug. *Tank Waste Remediation System Privatization Process Technical Baseline*. WHC-SD-WM-TI-774, Westinghouse Hanford Company, Richland, Washington (1996).
2. K. Wefers and C. Misra. *Oxides and Hydroxides of Aluminum*. Alcoa Technical Paper No. 19, Revised. Alcoa Laboratories, Pittsburgh, Pennsylvania (1987).
3. D. Rai, M. Sass, and D.A. Moore. "Chromium(III) Hydrolysis Constants and Solubility of Chromium(III) Hydroxide," *Inorg. Chem.*, Volume 26, pp. 345-349 (1987).
4. G.J. Lumetta, B.M. Rapko, M.J. Wagner, J. Liu, and Y.L. Chen. *Washing and Caustic Leaching of Hanford Tank Sludges: Results of FY 1996 Studies*. PNNL-11278, Rev. 1, Pacific Northwest National Laboratory, Richland, Washington (1996).
5. G.J. Lumetta, I.E. Burgeson, M.J. Wagner, J. Liu, and Y.L. Chen. *Washing and Caustic Leaching of Hanford Tank Sludges: Results of FY 1997 Studies*. PNNL-11636, Pacific Northwest National Laboratory, Richland, Washington (1997).
6. N.G. Colton. *Status Report: Pretreatment Chemistry Evaluation—Wash and Leach Factors for the Single-shell Tank Waste Inventory*. PNNL-11290, Pacific Northwest National Laboratory, Richland, Washington (1996).
7. G.J. Lumetta, B.M. Rapko, J. Liu, and D.J. Temer. "Enhanced Sludge Washing for Pretreating Hanford Tank Sludges," in *Science and Technology for Disposal of Radioactive Tank Wastes*, W. W. Schulz and N. J. Lombardo, eds., Plenum Press, New York, pp. 203-218 (1998).
8. G.J. Lumetta and B.M. Rapko. *Washing and Alkaline Leaching of Hanford Tank Sludges: A Status Report*. PNL-10078, Pacific Northwest Laboratory, Richland, Washington (1994).
9. B.M. Rapko, G.J. Lumetta, and M.J. Wagner. *Washing and Caustic Leaching of Hanford Tank Sludges: Results of FY 1995 Studies*. PNL-10712, Pacific Northwest Laboratory, Richland, Washington (1995).
10. G.J. Lumetta, M.J. Wagner, F.V. Hoopes, and R.T. Steele. *Washing and Caustic Leaching of Hanford Tank C-106 Sludge*. PNNL-11381, Pacific Northwest National Laboratory, Richland, Washington (1996).
11. D.J. Temer and R. Villarreal. *Sludge Washing and Alkaline Leaching Tests on Actual Hanford Tank Sludge: A Status Report*. LAUR-95-2070, Los Alamos National Laboratory, Los Alamos, New Mexico (1995).
12. D.J. Temer and R. Villarreal. *Sludge Washing and Alkaline Leaching Tests on Actual Hanford Tank Sludge: FY 1996 Results*. LAUR-96-2839, Los Alamos National Laboratory, Los Alamos, New Mexico (1996).
13. D.J. Temer and R. Villarreal. *Sludge Washing and Alkaline Leaching Tests on Actual Hanford Tank Sludge: FY 1997 Results*. LAUR-97-2889, Los Alamos National Laboratory, Los Alamos, New Mexico (1997).
14. G.J. Lumetta, B.M. Rapko, J. Liu, D.J. Temer, R.D. Hunt. *Washing and Caustic Leaching of Hanford*

WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

Tank Sludges: Results of FY 1998 Studies. PNNL-12026, Pacific Northwest National Laboratory, Richland, Washington (1998).

15. G.J. Lumetta and B.M. Rapko. "Removal of Chromium from Hanford Tank Sludges," *Sep. Sci. Technol*, Volume 34, pp. 1495-1506 (1999).

16. B.M. Rapko. *Oxidative Alkaline Dissolution of Chromium from Hanford Tank Sludge: Results of FY 98 Studies*. PNNL-11908, Pacific Northwest National Laboratory, Richland, Washington (1996).

FOOTNOTES

(a) To whom correspondence should be addressed; e-mail: gregg.lumetta@pnl.gov

(b) Pacific Northwest National Laboratory is operated for the U.S. Department of Energy by Battelle under Contract DE-AC06-76RLO 1830.