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SEPARATIONS/PRETREATMENT CONSIDERATIONS FOR HANFORD PRIVATIZATION PHASE II

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ABSTRACT

Pretreatment technologies developed to support the privatization effort by the Department of Energy are reviewed. Advancements in evaporation, solid-liquid separation, sludge treatment, solids controls, sodium management, and radionuclide removal are considered.

I. INTRODUCTION

The Tank Focus Area (TFA) is funded by the Department of Energy (DOE) Office of Science and Technology (OST) to develop, demonstrate, and deploy technologies that will assist in the treatment and closure of its nuclear waste tanks. These activities have assumed increased importance as the DOE has eliminated site funding for technical support. The TFA activities are driven by the site needs. As the Hanford site moves to privatization, the need for an understanding of the past work to support the TFA goals has been recognized, and follow-on work to support solving the DOE problems is under way.

The TFA technical team is organized by four line functions of retrieval, pretreatment, immobilization, and closure. Two overarching technical teams of characterization and safety support the four line functions. The interfaces between pretreatment and retrieval, immobilization, safety, and characterization are recognized as being key to understanding the overall remediation system.

Pretreatment involves assisting retrieval in preparation of retrieved waste for transport and storage, filtering the retrieved waste, processing sludge to reduce the mass of high-level waste (HLW) to be immobilized, concentrating the treated sludge, separating the radionuclides such as cesium, strontium, and technetium from the liquid low-level waste (LLW) fraction, and limiting the addition of chemicals to minimize the increase in LLW and HLW to be disposed of. Pretreatment is necessary to reduce overall remediation costs. As shown in Table 1, the cost of processing and disposing of HLW is estimated to be 33 times the cost of processing and disposing of LLW.¹ A doubling of the pretreatment costs for HLW can be offset by just a 1% reduction in the cost of immobilization and disposal. This paper discusses the role that pretreatment has in delivering cost-effective technologies.

II. PRIVATIZATION SUPPORT

The DOE is issuing a contract to private industry to treat wastes from its tanks. For Phase I of this process, the DOE management and integration contractor is responsible for retrieval, for limited waste pretreatment, and for staging of the waste transfer to the contractor. The privatization contractor will treat approximately 7 to 10% of the Hanford wastes. In Phase II, the contractor is responsible for the retrieval, pretreatment, and immobilization of the rest of the waste.

Table 1. Comparison of LLW and HLW management costs

	Hanford	
	LLW (\$/kg waste-oxide)	HLW (\$/kg waste-oxide)
Pretreatment	16	28
Immobilization	44	728
Disposal	4	1,370
Total	64	2,126

The DOE is responsible for preparation of the request for proposals, the bid process, and monitoring of the progress of the cleanup. Phase II is scheduled to be awarded in FY 2002.

While potential bidders for the Phase II effort have experience with nuclear waste remediation, the Hanford tank wastes are unique, and the contractors can benefit from the technology development work done by the TFA. Previous work has certified that enhanced sludge washing (ESW) will work, confirmed that solid-liquid processing can be adequately achieved, and demonstrated with real waste that cesium can be removed from the LLW supernatant and wash liquor. The DOE has also studied technetium removal and methods for separation of sodium salts for potential recycle or release, which would cut the volume and processing cost of the LLW. An understanding of past and present work will benefit the DOE and the private contractor in Phase II.

III. CESIUM REMOVAL STUDIES

The DOE has been instrumental in developing, demonstrating, and deploying technologies to remove cesium from tank supernatants. The Hanford CPU was designed as a mobile facility to treat a tank of Hanford supernatant using remote techniques. The DOE funded the development of the crystalline silicotitanate (CST) by Sandia and Texas A&M. CST was commercialized by UOP. Over the past several years flow studies with cesium sorbents have been done at Pacific Northwest National Laboratory (PNNL), Oak Ridge National Laboratory (ORNL), Westinghouse Hanford, and Savannah River. Cesium removal enables the LLW fraction to be handled with less radiation field. Figure 1 shows the loading of the sorbents with actual Melton Valley tank

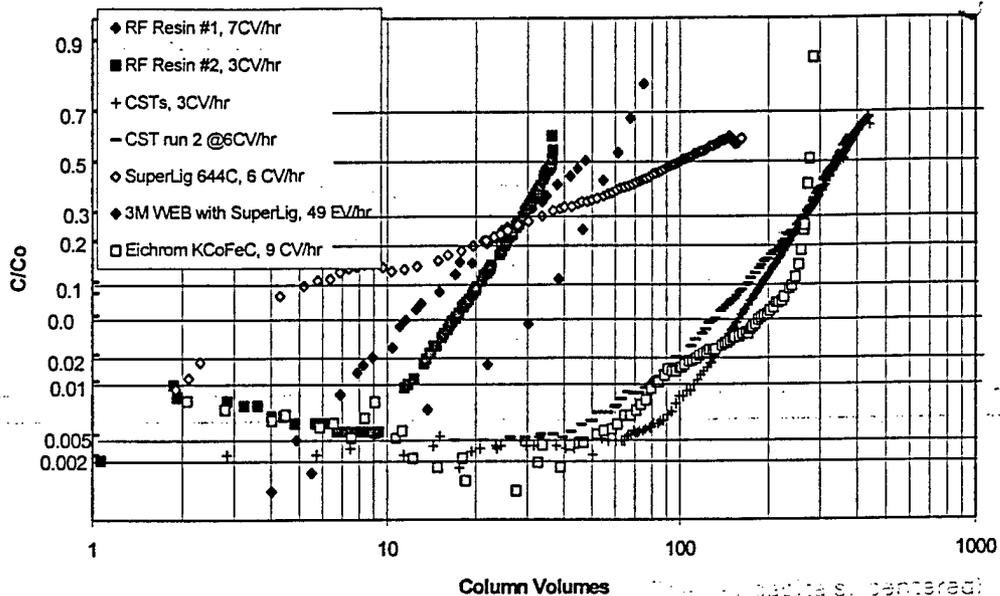
supernatant,² which is similar to the Hanford supernatant. Note that for these tests the CST from UOP provided the best decontamination factors. With the CST, 50% breakthrough occurred at approximately 400 column volumes (CV). These studies have led to a full-scale demonstration of a CPU at ORNL to treat supernatant.³

The studies at ORNL were used to develop a similar set of experiments at Hanford where several sorbents were evaluated on two types of Hanford supernatants, which were double shell slurry feed (DSSF) and complex concentrate (CC). With CST, 50% breakthrough occurred at 700 CV for the DSSF supernatant and about 1100 CV with the CC supernatant. Resorcinol-formaldehyde yielded 15 CV at 50% breakthrough with the DSSF supernatant. The better performance for the Hanford waste compared with the ORNL waste is due to the higher nitrate in the ORNL waste. The CST is not regenerable; our tests have shown that regeneration of the organic resins causes degradation of the resin.

IV. OTHER SUPERNATANT TECHNOLOGIES

The TFA has also supported development and demonstration of a number of other technologies. They included evaporation to reduce supernate volume, cross-flow filtration to separate solids from liquids, countercurrent decantation to increase the efficiency of sludge leaching, technetium removal, and sodium management technologies to minimize LLW and to recycle sodium.

Figure 1. Comparison of Ion-Exchange Materials Run in ORNL Cell C Continuous System Using MVST W-27 @ pH 13



Name (last, first, middle initial)
 Title
 Department/organization
 E-mail address (or box number)
 Phone (state, city)
 Telephone number
 Fax (if available)

Technetium Removal. Technetium removal was listed as a critical need in FY 1996 and FY 1997. The TFA funded removal flow studies and laboratory development studies.^{4,5} Per technetate (valence of +7) can be extractable using anion exchangers, but a portion of the technetium at Hanford was nonpertechnetate and was not extractable. This phenomenon was investigated at Los Alamos National Laboratory (LANL).⁶

Strontium/Transuranic (TRU) Removal. Strontium (Sr) was removed from Hanford and ORNL supernatant.^{4,7,8} For the bulk of the supernatants, Sr and TRU are below the level of regulatory concern. Careful filtration removes the Sr and TRU by an order of magnitude, indicating the Sr and TRU are suspended as a particulate. For the CC tanks, the concentrations of Sr and TRU in the supernatants are sufficiently high to require treatment.

Saltcake Dissolution. Feed staging for Phase I requires an understanding of saltcake dissolution so ESP modeling and experimental studies are under way. Personnel from Hanford, ORNL, Mississippi State University, and AEA Technology are involved in this study.

Evaporator. TFA has had an active evaporator effort for the past several years. Argonne National Laboratory has a small-scale evaporator, which was tested on simulants of Savannah River (SR) and ORNL supernatants. Oak Ridge did a full-scale demonstration in FY 1996, in which 25,000 gal was concentrated. TFA is now assisting SR in installing an evaporator to concentrate salts associated with the incinerator facility. Hanford engineering has expressed interest in this evaporator work in the past.

Sodium Management. The DOE has been funding work on caustic recycle⁹ and clean salt.^{10,11,12} Caustic recycle is a method to recover sodium hydroxide, which can be used to condition the sluicing solution and to leach sludge during ESW. The Hanford flowsheet shows that 18% increase in waste is added by new sodium hydroxide requirements, so there is a cost driver to reduce the amount of added caustic. Clean salt is a selective crystallization process where pure sodium nitrate is removed from the supernate. These technologies have been reviewed by Richland and SR, and TFA is considering a technology procurement to demonstrate these technologies.

Table 2 describes some of these technologies which have been demonstrated or have advanced designs.

Table 2. Description and key data related to some of the more developed TFA pretreatment technologies

Technology	Description	Characteristics/Parameters/Data	Applicability
Evaporation	Out-of-Tank Evaporator Demonstration: A single-stage, subatmospheric evaporator, skid mounted to process radioactive, high-salt - content liquid waste. Remotely operated	Distillate rating: 90 gph Feed tank capacity: 400 gal Decontamination factor for Cs: ~ 5 x 10 ⁶ Volume reduction: ~ 25%	-Minimize LLLW volumes -Create space in double-shell tanks
Cesium Removal	Cesium Removal Demonstration: Large-scale demonstration producing a small-volume solid waste form Modular, transportable, remotely operated design	Ion-exchange material: crystalline silicotitanate Waste form meets NTS WAC Waste form is compatible with vitrification Feed tank capacity: 500 gal Max. feed rate: 5 gpm Nominal feed rate: 3-6 CV/h	-Alkaline supernates
Countercurrent Decantation	Countercurrent solids washing using thickeners and fluidic reverse flow-diverter pumps Flocculants used as a settling aid	Feed tanks: Two, 10,000 gal Thickeners: Six, 7-ft diam, cone bottom Sludge feed rate: 3 gpm Wash water rate: 5 gpm	Sludge washing Release tanks for other uses Requires less wash water

V. SLUDGE TREATMENT

The volume of immobilized HLW, which will be primarily determined by the sludges, 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 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. In 1993, the DOE considered three separation options for the Hanford sludges. The treatment options¹³ included simple sludge washing, ESW, and advanced separations. Simple sludge washing uses only water or dilute sodium hydroxide with corrosion inhibitors such as sodium nitrite. ESW refers to simple sludge washing followed by caustic leaching with 2 to 3 M NaOH at an elevated temperature. The leached solids are then washed with the dilute NaOH to remove the dissolved components and the added NaOH. Advanced separations consist of complete dissolution, if possible, followed by extensive radionuclide separation such as TRUEX^{14 15 16 17} and SREX.^{18 19} After a cost analysis of the options, it was concluded that the 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 such as the minimum wash and leach factors^{20 21} were made

about the ESW process, and verification of these assumptions was required by the DOE.

VI. ENHANCED SLUDGE WASHING

As part of the verification studies, ESW studies^{22 23 24 25 26 27 28 29 30 31 32 33 34} have been performed on sludge samples from 34 Hanford tanks by researchers at PNNL, LANL, and ORNL. The ORNL ESW study attempted to maximize the effectiveness of the ESW process through increases in temperature, leaching time, and caustic concentration. In contrast, the LANL and PNNL researchers have tested numerous sludge samples with a single set of test conditions, which were periodically modified as new results were obtained. As shown in Table 3, tests have shown that the ESW will most likely remove the required amounts of aluminum, phosphate, and chromium.

Researchers at LANL, ORNL, and PNNL are currently performing parametric studies on ESW to optimize the process for a particular sludge and to provide a much more reasonable estimate of the LLW that will be generated by the washes and leaches. During the current studies, the effects of process variables such as NaOH concentration (1 and 3 M), temperature (60, 80, and 100°C), and leaching time (5, 24, 72, and 168 h) on the efficacy of the caustic leaching process will be determined. The goal of these tasks is to minimize the overall system cost for the total quantity of HLW and LLW 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 3. Percentage removal of key elements by sludge washing

Ions	Minimum goal (%)	Test results on a weighted basis			Overall wash and leach factor	
		Wash	Leach	Total	1996	1997
Al	68	14	74	88	60	92
Cr	64	44	32	77	40-65	86
PO ₄ ³⁻	74	55	35	90	70	95

VII. CONTROL OF SOLID FORMATION

At the end of the ESW process, the remaining solids are considered HLW while the potentially saturated solutions are defined as LLW. However, solids will form in the LLW 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 is unacceptably large (200 L of 3 M NaOH per kilogram of aluminum).³⁵

Tests to monitor solid formation during the ESW process were performed on sludge samples from eight Hanford tanks. Solids in filtered solutions were observed with seven of the eight sludge samples^{23 35} which were from Hanford tanks T-104, C-105, C-107, C-108, SX-113, B-202, and C-104. Sludges that contain phosphates are likely to form solids because phosphate solubility is very sensitive to temperature. Phosphate is typically present as insoluble phosphates and must be removed by metathesis of water-insoluble metal phosphates to insoluble hydroxides and soluble phosphates. After the leach at elevated temperatures, the phosphate can reprecipitate as a sticky gel as the liquid is cooled. For example, after the sludge sample from Hanford

tank T-104 was leached with caustic, gels of natrophosphate, $\text{Na}_7(\text{PO}_4)_2\text{F}\cdot 19\text{H}_2\text{O}$, were observed in filtered leach solutions.

Other components in the filtered leach solutions can also form precipitates as the hot solutions are allowed to cool. The filtered leachate from the SX-113 test produced a significant amount of particulate material, which appeared to be semigelatinous when suspended. Chemical analysis indicated that these SX-113 particles contained sodium and silicon. The test with sludge from Hanford tank B-202 also produced a particulate material, which contained bismuth.³⁶

As a result of these observed problems, an alternative flowsheet³⁵ has been proposed based on the Bayer process in the aluminum industry. In this flowsheet, the sludge is leached at elevated temperature, and 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. The resulting solids will be transported to the low-activity waste stream for immobilization.

VIII. SUMMARY TABLE

Table 4 lists key references by pretreatment subject area.

Table 4. Pretreatment unit operations and documents relevant to waste remediation at Hanford

	Unit Operations	Key References
Sludge pretreatment	Enhanced sludge washing	13, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34
	TRUEX (advanced sludge separation)	14, 15, 16, 17
	SREX (advanced sludge separation)	18, 19
	Control of solid formation and thermodynamic modeling	35, 36
	Countercurrent decantation	37, 38, 39, 40
Supernatant pretreatment	Cesium removal	2, 3, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50,
	Strontium removal	7
	Technetium removal	6
	Evaporation	51
	Sodium management	10, 11, 12
Solid-liquid Separation		49, 50

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