

OVERVIEW OF LABORATORY TREATMENT STUDIES FOR DECONTAMINATION OF LIQUID LOW-LEVEL WASTE AT OAK RIDGE NATIONAL LABORATORY

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ABSTRACT

Oak Ridge National Laboratory (ORNL) is taking a multifaceted approach to solving its radioactive waste problems. Liquid low-level waste (LLLW), which usually contains radioactive contaminants at millicurie-per-liter levels, is generated at a rate of over 400,000 gal/year. At present, most of this waste is collected in a centralized treatment facility, where the waste is made alkaline and evaporated to a specific gravity of 1.25. The resulting evaporator concentrate, which contains most of the radioactive constituents, is 4-5 M NaNO₃. It is stored in underground tanks along with more than 350,000 gal of other complex wastes from past nuclear energy research activities. Methods are being developed to dispose of these wastes and to determine more efficient methods of treatment (source and centralized) for future wastes. The aim of the development studies is to devise treatment methods that will concentrate the contaminants of the waste into small solid waste packages while allowing discharge of the treated liquids to the environment or reducing the hazard class of the liquids for solidification and disposal as a solid waste.

To reduce the amount of radioactive materials in future LLLW, the major contributors to LLLW have been identified, and evaluations are under way to determine if source treatment, centralized treatment, or a combination of pretreatment at the source, followed by centralized treatment, will be the most appropriate. Methods for removing the contaminants at the source facilities are being proposed and tested. The choice of treatment method depends not only on the chemical nature of the waste but also on the physical aspects of the facility where the waste is to be treated. Therefore, current development efforts are being tailored for the particular facility and waste composition. For existing legacy waste, potassium-cobalt hexacyanoferrate and sodium titanate are being tested for selective removal of radioactive cesium and strontium from simulated waste solutions. These same two materials have also been found to be effective for treatment of simulated LLLW with the composition predicted for future generated LLLW collected in the centralized system. Studies are being performed using resorcinol-formaldehyde-based ion-exchange resins and tailored inorganic exchangers in the form of microspheres produced using the Sol-Gel process for source treatment/pretreatment at the major LLLW-generating sites. In some cases, these treatment studies are critical to the continuity of certain ORNL operations. On the whole, these efforts will result in improved radioactive waste management at ORNL, enhanced waste treatment technology base for U.S. Department of Energy facilities, and strengthened viability of nuclear energy research.

INTRODUCTION

Oak Ridge National Laboratory's (ORNL's) approach to development of liquid low-level waste (LLLW) treatment technology involves plans for both centralized treatment of combined wastes and treatment of selected wastes at the generator facility. LLLW, which usually contains radioactive contaminants at millicurie-per-liter levels, is generated at a rate of over 400,000 gal/year. At present, many of these wastes are combined in a centralized treatment facility and evaporated to a concentrated salt solution for storage in underground tanks along with complex wastes from past nuclear energy research. Storage capacity in these tanks, however, is limited, and methods for minimization of LLLW and disposal of the existing salt solutions are being developed. At present, the underground storage tanks, known as the Melton Valley Storage Tanks (MVSTs) contain 350,000 gal of a complex mixture of contaminated sludges and 4-5 M NaNO₃ liquid. Various treatment options are being evaluated for this waste. In a systems analysis performed in 1989 (1), the current major contributors of LLLW were identified, and methods for minimizing LLLW and treating the waste at the generator facility are being evaluated. Where possible, it is desired to remove the bulk of the radioactive contaminants at the generator facility while allowing treated liquids containing nonhazardous salts and trace contaminants to be discharged to a central-

ized treatment plant. Characteristics of waste feed for the centralized LLLW treatment system will depend on future ORNL programs and waste generation activities. The composition for future generated LLLW has been predicted, and treatability studies are being performed using laboratory simulated wastes.

TREATMENT OF WASTES IN MELTON VALLEY STORAGE TANKS

At the present time, about 350,000 gal of concentrated LLLW salt solution is stored in eight 50,000 gallon underground storage tanks known as the MVSTs. The waste is composed of solid (sludge) and liquid (supernate) phases. The supernate composition is principally a highly alkaline 4-5 M NaNO₃ solution with up to 6 mCi/L of ¹³⁷Cs and 0.2 mCi/L ⁹⁰Sr along with lesser quantities of radioactive cobalt and europium. The solid phase is principally precipitated sodium, potassium, and calcium compounds with transuranic contaminants at levels up to 1 mCi/kg. Three treatment options are being considered for this waste. The options are designed to accommodate potential disposal requirements for the treated wastes. The primary choice for treatment involves mobilization of the solid phase, mixing with the supernate phase, and evaporating a uniform mix of the two phases into a solid salt cake for disposal at the Waste Isolation Pilot Plant (WIPP) in

Carlsbad, New Mexico (2). An alternative to this method involves treating and removing contaminants from the liquid supernates and discharging the treated liquids to the environment. The sludges would be solidified in a separate operation for disposal at the WIPP. A modification of this alternative involves removing the contaminants from the supernates and evaporating the treated supernates to a Class LII salt cake for disposal on the Oak Ridge Reservation (ORR). This option also involves treating the sludges separately for disposal at the WIPP. If decontamination of the MVST supernate requires methods similar to those expected for future LLLW, it may be feasible to mix the two wastes and treat them together.

Methods for selectively removing strontium and cesium from the supernates have been studied extensively in bench-scale experiments using simulated and actual wastes (3). The two most effective ion exchangers are the transition-metal hexacyanoferrates such as potassium-cobalt hexacyanoferrate, $K_2CoFe(CN)_6$ (abbreviated KCCF), for cesium and sodium titanate (abbreviated NaTi), developed and produced for strontium removal at the Sandia National Laboratory (4). Cesium distribution coefficients with KCCF were as high as 1.9×10^5 ml/g at alkaline pH values. Strontium distribution coefficients with NaTi were as high as 1.9×10^5 ml/g at pH 13. Both exchangers are effective enough for treatment of supernates for solidification to a Class LII waste; however, nitrates must be removed if liquids are to be discharged to the environment.

After removal of the strontium and cesium, the MVST supernates might be solidified using a grout formulation which, from the standpoint of radiological composition, would meet the draft waste acceptance criteria for Class LII disposal of the waste on the ORR. The disadvantage of this approach is the large volume of Class LII waste that would be produced. The estimated total volume of solidified supernates is about 1500 m^3 . The supernates could be evaporated to a salt cake and produce a much smaller volume of Class LII waste; however, the salt cake waste form is undesirable due to the lack of leach resistance. If the nitrates can be destroyed, other alternatives such as further treatment and discharge of liquids to the environment can be considered. Two promising methods are being evaluated for nitrate destruction. One method, being developed by ORNL, involves reduction of the nitrate to ammonia by reaction with aluminum powder (5). This produces a solid, stable secondary waste form that tends to coprecipitate and trap other contaminants as an additional benefit. The estimated volume of this secondary waste is about 640 m^3 . Since the levels of nitrate are large, the amount of ammonia generated is too large for gaseous discharge to the environment. Methods for dealing with the ammonia are being investigated. Another nitrate treatment method, being demonstrated on a pilot scale by Bradtec-US, Inc., involves electrochemically converting the nitrate to N_2 , O_2 , and water. This method would produce essentially no secondary solid waste. The N_2 , O_2 , and water could be discharged to the environment, and a secondary liquid waste consisting of slightly contaminated sodium hydroxide solution might possibly be used for pH adjustment in other waste treatment operations or be neutralized, treated to remove trace contaminants, and discharged to the environment. Results of the initial Bradtec study (6) at Rocky Flats indicate good conversion of nitrate to ammonia; however, the conversion of ammonia to N_2 , O_2 , and water at the anode compartment of the cell was incomplete. Bradtec is continuing the studies.

Alternatives for treatment of the MVST sludges are being developed. Most of the development efforts center on methods of mobilizing and removing the sludges from the tanks. Enhanced sludge treatment options which involve dissolving the sludge in an acid solution and using a solvent extraction process such as TRUEX (7) for selective removal of transuranic components of the sludge are also being tested.

TREATMENT OF NEWLY GENERATED LIQUID LOW-LEVEL WASTE

The future composition of LLLW is likely to change as a result of new research and development (R&D) activities and changes in waste treatment and disposal operations. The systems analysis performed in 1989 (1) has supplied data to allow estimating the possible composition of the newly generated LLLW (NGLLLW). NGLLLW is likely to contain even higher concentrations of radioactive strontium and cesium and will also contain high levels of radioactive ruthenium. The estimated future generation rate is about 55,000 gal/year. The waste will likely be more dilute than MVST supernates and be dominated by sodium carbonates and hydroxides with smaller amounts of nitrates. Table I compares the composition of NGLLLW with that of MVST supernate. Bench-scale treatability studies are being performed to determine methods for removing the principal contaminants from simulated NGLLLW. Among the many exchangers considered and tested for contaminant removal, KCCF and NaTi were once again found to be superior sorbents for cesium and strontium respectively. KCCF was tested as a granular solid, a dry powdered solid, a wet preformed slurry, and an in situ slurry (formed in the waste solution by adding the two liquid KCCF constituents). Of all the application methods, the wet preformed slurry and in situ methods were the most effective. These methods, however, will require settling and filtration operations to separate the loaded sorbents from the wastewater. Sodium cobalt hexacyanoferrate, $Na_2CoFe(CN)_6$ (abbreviated NaCoCF), which was comparable to KCCF in cesium removal effectiveness, was prepared in the form of microspheres using the internal gelation process. The internal gelation, or Sol-Gel process, was developed to prepare uranium oxide microspheres for fast breeder reactor fuels (8). The microspheres can be used in columns for liquid waste treatment, thereby eliminating solid/liquid separation equipment. The physical stability of the microspheres was excellent. Cesium removal effectiveness, though not as good as the KCCF slurry, was superior to the KCCF granular material. Table II compares cesium removal effectiveness for the various KCCF application methods. The optimum pH for cesium removal with KCCF was 8, and the cesium/KCCF reaction product was stable while in contact with waste liquids for extended times.

For removal of strontium from NGLLLW, NaTi was most effective at pH 13, when applied in granular, powder, or slurry forms. It was found that NaTi obtained from different suppliers and different lots from the same supplier sometimes performed inconsistently. Most products, however, were sufficiently effective for removal of strontium from NGLLLW. Decontamination factors (DFs) as high as 20 were achieved using 500 ppm NaTi in the NGLLLW. Strontium removal was further enhanced by precipitation of aluminum hydroxide in the pH range of 10 to 8. Dissolved calcium greatly interfered with sorption of strontium by the NaTi. Alternative strontium removal methods are being evaluated.

TABLE I
Composition of Newly-Generated LLLW and MVST Supernate Solutions

Component	New-Generated LLLW		MVST Supernate ^a	
	mol/L	mCi/L	mol/L	mCi/L
NaOH	0.335		0.24	
Na ₂ CO ₃	0.587		0.14	
NaNO ₃	0.061		3.9	
NaCl	0.034		0.10	
NaAlO ₂	0.0117			
KNO ₃			0.25	
LiCl	0.025			
Al(NO ₃) ₃			0.005	
¹³⁷ Cs	8.8 × 10 ⁻⁶	99.5	5.0 × 10 ⁻⁷	5.94
⁹⁰ Sr	1.6 × 10 ⁻⁶	19.4		
¹⁰⁶ Ru	3.2 × 10 ⁻⁷	132		
pH	13.1		12.7	
Density	1.074 g/mL		1.242 g/mL	

TABLE II
Comparison of Sodium/Potassium-Cobalt Hexacyanoferrate Preparations for Removal of Cesium from Newly-Generated LLLW^{*}

Exchanger Preparation	Decontamination Factor
Potassium-cobalt hexacyanoferrate, (KCCF) granular form	24
KCCF preformed slurry	2440
KCCF in situ slurry	2472
NaCoCF/HTiO microsphere ^{**}	535

^{*} Exchanger concentration was 100 mg/L and contact time was 20 hr.
^{**} NaCoCF/HTiO is sodium cobalt hexacyanoferrate incorporated into a hydrous titanium oxide microsphere.

Ferric sulfate and KCCF were more effective than manganese dioxide or cobalt sulfide for ruthenium removal. However, the required ruthenium DF for discharge of treated NGLLLW liquids to the environment is 1.7×10^7 . None of the materials were effective enough to reach this DF. Table III shows test results from evaluation of the these sorbents. If, however, the treated NGLLLW were to be evaporated and solidified for Class LII disposal, the ruthenium sorbents could be used to reduce the gamma exposure rate associated with ^{103/106}Ru radiations in the final waste form. If solidified, it is estimated that about 80 m³ of Class LII solid waste would be produced each year. Improved methods for removal of ruthenium continue to be pursued. Literature (9) indicates that ruthenium speciation in a waste solution is likely to vary and that some species are more likely to sorb than others. For improved removal from the waste, the ruthenium must be converted to a species that can readily be removed.

Although the concentration of nitrates in the NGLLLW is small compared with that in the MVST supernates, reduction in nitrate level would be necessary before discharge to the

environment. The same methods as those described for MVST supernate treatment are being investigated for NGLLLW.

The studies so far indicate that similar methods will be effective for treating both the MVST supernates and the NGLLLW. Additional studies will evaluate modifications necessary for treating mixtures of MVST supernate and NGLLLW.

TREATMENT OF LIQUID LOW-LEVEL WASTE AT GENERATOR FACILITIES

There are several reasons why it is important to have the capability for removing contaminants at source facilities instead of allowing the wastes to combine and undergo treatment at centralized facilities: 1) the contaminant is more likely to be in a less complex matrix and, therefore, easier to selectively remove from the solution; 2) the resultant secondary waste volume is likely to be smaller; and 3) the processes used to treat the waste may be smaller in scale and less complex. Another driving force for treatment of waste at the generator facility involves requirements set forth in the Federal Facility Agreement (FFA) (10). The FFA includes new requirements for construction and maintenance of underground liquid waste storage and transfer systems. Due to inadequate secondary containment and system design, a significant portion of the ORNL LLLW collection and transfer (CAT) system is either being upgraded or taken out of service. As a result, certain facilities have lost or are at risk of losing access to the LLLW CAT system. To allow R&D activities to continue, these facilities are developing methods for removing, stabilizing, and solidifying radiological contaminants at the source so that the resultant liquid waste can be bottled and transferred by truck to the LLLW evaporator system. The activity of the liquid waste must be low enough to meet U.S. Department of Transportation regulations and limit the dose rate at the container surface to less than 50 mrem/hr. The high-activity solid waste can be dewatered, packaged, shielded, and safely transferred to storage facilities.

Source Treatment at the Radiochemical Engineering Development Center

The Radiochemical Engineering Development Center (REDC) is an important producer of transuranic radioisotopes. The waste produced at this facility is extremely complex and contains higher levels of contaminants than that produced at any of the other ORNL facilities. The processing at this facility involves dissolution of irradiated curium-amerium targets and complex chemical operations to separate and purify the radioisotopes of interest (11). The combined waste from this facility is a mixture of alkali metal salts and organic extractants. An effort is currently under way to develop treatment methods for removal of specific radioactive contaminants from processing solutions before they are combined with other waste materials. The treatment equipment must be relatively small due to limited hot cell space and must also be very simple to allow remote operation with mechanical manipulators. Currently, the removal of cesium from target decladding solutions and the removal of Cs, Sr, and Ru from Cleanex raffinate solutions are being investigated. Table IV gives the composition of these solutions. The decladding so-

lution is a highly concentrated solution of sodium hydroxide and sodium nitrate used to dissolve aluminum cladding from the irradiated target. After decladding, this waste contains a high concentration of sodium aluminate along with sodium hydroxide, sodium nitrate, and curie-per-liter levels of radioactive cesium and strontium. In studies to date, only resorcinol-formaldehyde-based ion-exchange resins have proved to be effective in removing cesium from highly alkaline solutions. Other exchangers would likely be applicable if pH adjustment were possible; however, the large inventory of aluminum remains dissolved only in highly basic solutions. The resorcinol-formaldehyde resin made at the Savannah River Laboratory (SRL resin) for a similar application (12) has been found to be the most effective in this case. Distribution coefficients ranging from 200 ml/g (at process levels of 1.5 mM cesium) to 10,000 ml/g (with trace cesium levels) have been measured, and it is estimated that 95% or more of the cesium activity can be removed from the decladding solution by using a small quantity of this material. The proposed treatment involves installation of a small column (2 in. ID x 2 ft long) containing 1 L of the resin for the processing of a batch

TABLE III
Comparison of Various Sorbents for Removal of Ruthenium from Newly-Generated LLLW

Sorbent	Sorbent Conc. (mg/L)	Optimum pH	Decontamination Factor
Potassium cobalt hexacyanoferrate (KCCF) in situ slurry	100	11	9.9
KCCF in situ slurry	1000	11	48
Ferric sulfate	70 (as Fe)	10	28
Cobalt sulfide	500	11	9.7
Manganese dioxide	500	11	6.7
Alkaline precipitation only	None	10	7.3

TABLE IV
Major Constituents in REDC Fission Product (non-Rare Earth) Streams

Isotopic Constituent	Dejacketing Solution (Ci/L)	Cleanex Raffinate (Ci/L)
^{137}Cs	2-9	2.5-3.0
^{90}Sr	0.02-0.14	0.5-7.5
^{106}Ru	0.03-0.5	3.4-62
<u>Chemical Constituent</u>	<u>(millimol/L)</u>	<u>(milliomol/L)</u>
Cs	0.2-1.5	0.2-0.5
Rb	0.01-0.18	0.015-0.06
Ba	0.1-0.14	0.13-3.4
Sr	0.006-0.04	0.065-1.0
Ru	0.001-0.07	0.9-7.0
NaOH	2.2 M	
NaAlO ₂	1.25 M	
NaNO ₃	0.94 M	0.75 M
NaNO ₂	0.14 M	
HNO ₃		< 0.1
LiNO ₃		0.1

TABLE V
Comparison of Inorganic Ion-exchangers for Decontamination of Cleanex Raffinate*

Exchanger	Cesium Distribution After 4 hours (mL/g)	Strontium Distribution After 4 hours (mL/g)
Natural clinoptilolite zeolite, granular	396±4	36±4
Natural chabazite zeolite, granular	120±10	<15
Titanium monohydrogen phosphate, (TiHP) powder	126±3	<6
Zirconium monohydrogen phosphate, (ZrHP) powder	135±5	4.8±0.6
Synthetic mordenite zeolite, granular	650±30	19±2
Polyantimonic acid (HAP), granular	<13	590±60
Ammonium molybdophosphate, (AMP) powder	7700 (30 min)	
Sodium cobalt hexacyanoferrate (NaCoCF) powder	44000±2000	<2
NaCoCF powder	5100 (30 min)	
NaCoCF/TiHP microspheres**	439 (30 min)	

* Solution to exchanger ratio was 200.
** Solution to microsphere ratio was 113.

of decladding solution. The loaded resin is then eluted with a small volume of hydrochloric acid, which is evaporated and stored in the hot cell for later treatment.

The Cleanex process is a solvent extraction process in which di(2-ethylhexyl) phosphoric acid (HDEHP) is used to extract trivalent actinide and lanthanide elements from dilute acids or salt solutions. The Cleanex raffinate is a dilute hydrochloric acid solution used to strip impurities such as Cs, Ru, and Ba from the HDEHP solvent. Several ion-exchange materials have been tested for removal of cesium and strontium from this solution. Materials tested for this waste include several inorganic zeolites; titanium monohydrogen phosphate, $\text{Ti}(\text{HPO}_4)_2 \cdot \text{H}_2\text{O}$ (abbreviated TiHP), and hydrous zirconium phosphate, $\text{Zr}(\text{HPO}_4)_2 \cdot \text{H}_2\text{O}$ (abbreviated ZrHP), prepared as microspheres; polyantimonic acid, $\text{Sb}_2\text{O}_5 \cdot n\text{H}_2\text{O}$ (abbreviated HAP); a synthetic mordenite zeolite; sodium cobalt hexacyanoferrate, $\text{Na}_2\text{CoFe}(\text{CN})_6$ (abbreviated NaCoCF); and ammonium molybdophosphate, $(\text{NH}_4)_3\text{PO}_4 \cdot 12\text{MoO}_3 \cdot n\text{H}_2\text{O}$ (abbreviated AMP). In bench-scale scoping studies using simulated Cleanex raffinate, AMP and NaCoCF were the most effective for cesium removal. HAP was the only sorbent effective for removal of strontium from this solution. Table V compares cesium removal effectiveness for the exchangers. To be used, these materials must be made into a granular material or otherwise fixed onto a support matrix such that they may be used in a small, flow-through column. There is not enough space in REDC hot cells to allow for application of powdered or slurried exchangers which require solid/liquid separation equipment. The NaCoCF microspheres, used in NGLLW treatment tests, were tested for cesium removal from Cleanex raffinate. The formulation for the microspheres can contain up to 30% NaCoCF and still maintain physical integrity in aqueous solutions with a wide pH range. The composite microspheres can also be converted to the phosphate form to make them usable in strong acids. The spheres can be produced in sizes up to 1.2

mm and as small as 0.1 mm in diameter. Similar microsphere development is being performed for the AMP.

Source Treatment for the Fission Product Release Program

In another ORNL facility, the Sol-Gel microspheres are being used successfully to remove $^{137/134}\text{Cs}$, ^{154}Eu , and ^{60}Co from waste produced as a result of the Fission Product Release Program (FPRP). The FPRP is an experimental program designed to test high-burnup spent fuel from commercial light water reactors under severe accident conditions. The goal of the program, which is sponsored by the Nuclear Regulatory Commission, is to establish a fission product release data base to be used in modeling analysis of these hypothetical accidents. As a consequence of FFA directives, the singly contained LLLW tanks, formerly used to collect and transfer LLLW from this facility to the centralized treatment system, were taken out of service. In order to continue FPRP activities, the wastes had to be pretreated to meet the requirements for collecting and trucking the waste to the centralized LLLW evaporator. The FPRP wastes consist of aqueous ammonium hydroxide/hydrogen peroxide leach solutions containing 4-8 Ci/L of cesium and 50-300 mCi/L of europium and cobalt. A very simple hot cell apparatus was constructed for contacting these wastes with a mixture of ZrHP and hydrous titanium oxide, $\text{TiO}_2 \cdot n\text{H}_2\text{O}$ (abbreviated HTiO) microspheres. Fifty grams of each type of microsphere is loaded into a stainless steel mesh basket, which is placed into a 1.5-L stainless steel container with the LLLW. Air is sparged into the container to strip the ammonia from the solution and to enhance exchanger/solution contact. The exchanger and waste are allowed to react for at least 1 day, after which the solution is transferred to a second container outside of the hot cell where the solution is contacted with 50 grams of ZrHP to remove a small amount of residual $^{137/134}\text{Cs}$. The resultant waste effluent is bottled and transported by truck to the LLLW treatment system. Once the inorganic exchangers have

been loaded to capacity, they will be dried with air within the stainless steel containers. Afterwards, they will be heat treated in the cell to 200°C to remove any free water or water of hydration. Once sealed, the container is adequate for waste storage. The loaded inorganic exchangers are extremely stable and are an excellent matrix for long-term storage or disposal. Acid leachates (4 M HNO₃) used to remove fission products from experimental apparatus will be treated in a similar fashion. First, these solutions will be diluted with water to 1 M HNO₃, then contacted with a mixture of ZrHP and HTiO microspheres containing about 20% (by weight) NaCoCF.

SUMMARY

In general, all of the above described activities are designed to reduce the volume of radioactive waste at ORNL to small solid concentrated waste packages while allowing treated liquids either to be discharged to the environment after further treatment or to be solidified and stabilized into a lower-hazard-class waste form which can be safely and permanently disposed of on the ORR. Selective ion exchangers are being applied where possible for removal of major contaminants such as ¹³⁷Cs and ⁹⁰Sr from complex legacy wastes and wastes to be treated in the future. Nitrate destruction techniques are also being developed for these wastes. Source treatment methods are being developed for removing contaminants from simple, well-characterized solutions where contaminant concentrations are highest as opposed to allowing the contaminants to disperse into large-volume, very complex waste mixtures. Innovative methods are being developed for source treatment using inorganic exchangers in the form of microspheres. In some cases, these methods are critical to the continuing progress of research activities. The combination of source treatment and centralized treatment will result in the optimum overall approach to LLLW management at ORNL. These efforts will result in improved radioactive waste management at ORNL, enhanced waste treatment technology base for U.S. Department of Energy facilities, and enhanced viability of nuclear energy research.

REFERENCES

1. T. J. ABRAHAM, S. M. DePAOLI, A. B. WALKER, and H. L. JENNINGS, "Preliminary Analysis of the Oak Ridge National Laboratory Liquid Low-Level Waste System," ORNL/TM-11250, Oak Ridge National Laboratory (September 1989).
2. J. B. BERRY, D. O. CAMPBELL, D. D. LEE, and T. L. WHITE, "Process Development for Remote-Handled Mixed-Waste Treatment," presented at the 1990 AIChE Summer National Meeting, San Diego, CA (August 1990).
3. D. O. CAMPBELL, D. D. LEE, and T. A. DILLOW, "Development Studies for Treatment of Low-Level Liquid Waste," ORNL/TM-11798, Oak Ridge National Laboratory (November 1991).
4. R. G. DOSCH, "Final Report on the Application of Titanates, Niobates, and Tantalates to Neutralized Defense Waste Decontamination — Materials, Properties, Physical Forms, and Regeneration Techniques," SAND80-1212, Sandia National Laboratory (1981).
5. A. J. MATTUS, D. D. LEE, T. A. DILLOW, L. L. FARR, S. L. LOGHRY, W. W. PITT, and M. R. GIBSON, "A Low-Temperature Process for the Denitration of Hanford Single-Shell Tank, Nitrate-Based Waste Utilizing the Nitrate to Ammonia and Ceramic (NAC) Process," ORNL/TM-12245, Oak Ridge National Laboratory (1992).
6. K. J. ATKINS, D. BRADBURY, G. R. ELDER, and S. M. SCRIVENS, "Electrochemical Ion Exchange Treatment of Plutonium-Bearing Nitrate Waste, Demonstration Study," prepared for EG&G Rocky Flats, Inc., by Bradtec US, Inc. (August 1992).
7. R. A. LEONARD, G. F. VANDEGRIFT, D. G. KALINA, D. F. FISCHER, R. W. BANE, L. BURRIS, E. P. HORWITZ, R. CHIARIZIA, and H. DIAMOND, "The Extraction and Recovery of Plutonium and Americium from Nitric Acid Waste Solutions by the TRUEX Process — Continuing Development Studies," ANL-85-45, Argonne National Laboratory (1985).
8. J. L. COLLINS, M. H. LLOYD, and R. L. FELLOWS, "The Basic Chemistry Involved in the Internal-Gelation Method of Precipitating Uranium as Determined by pH Measurements," *Radiochimica Acta*, 42,121-134 (1987).
9. W. W. SCHULZ, S. G. METCALF, and G. S. BARNEY, "Radiochemistry of Ruthenium," NAS-NS-3029 (rev.), Rockwell Hanford Operations, Richland, Washington (1984).
10. S. M. ROBINSON, S. M. DePAOLI, and A. B. WALKER, "Federal Facility Agreement Contingency, Upgrade, and Replacement Plans for the ORNL Active Low-Level Radioactive Waste Tank System," ORNL/TM-11795, Oak Ridge National Laboratory (June 1991).
11. L. J. KING, J. E. BIGELOW, and E. D. COLLINS, "Experience in the Separation and Purification of Transplutonium Elements in the Transuranium Processing Plant at Oak Ridge National Laboratory," pp 133-147 in *Transuranium Elements — Production and Recovery*, J. D. Navratil and W. W. Schulz (eds.), ACS Symposium Series 161, American Chemical Society, Washington, D.C. (1981).
12. J. P. BIBLER and L. A. BRAY, "Testing a New Cesium-Specific Ion Exchange Resin for Decontamination of Alkaline High-Activity Waste," American Chemical Society I&EC Special Symposium, Atlanta, GA (September 1992).