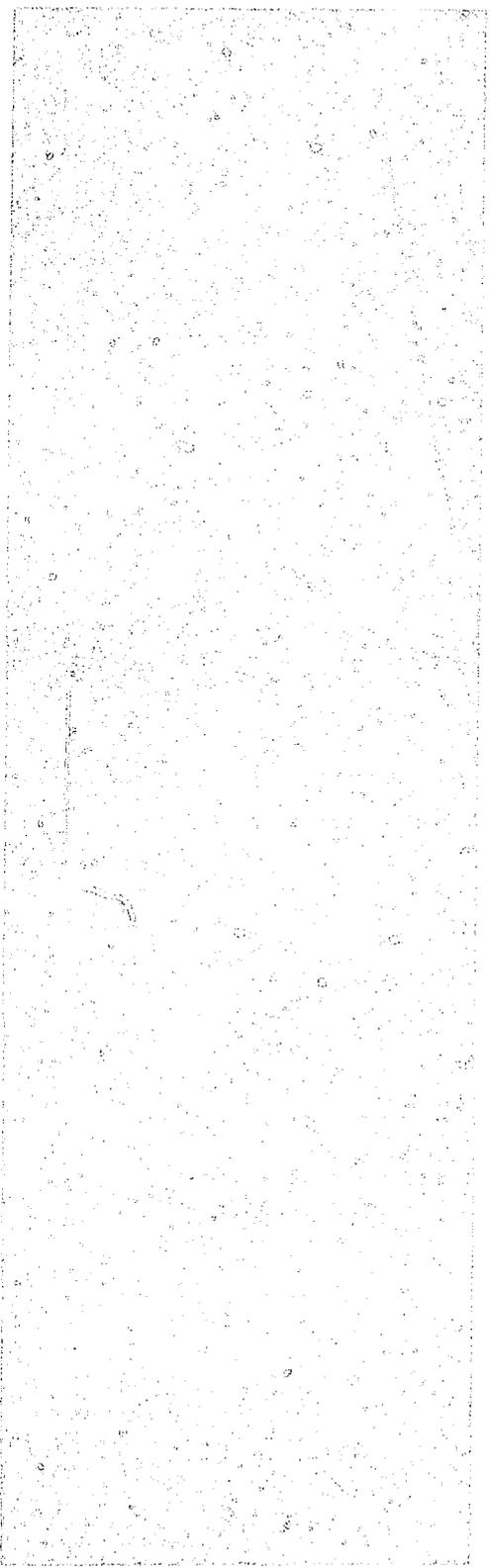


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*Partitioning High-Level Waste from Alkaline  
Solution: A Literature Survey*

**Los Alamos**  
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*Partitioning High-Level Waste from Alkaline  
Solution: A Literature Survey*

*S. Fredric Marsh*

**MASTER**

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# PARTITIONING HIGH-LEVEL WASTE FROM ALKALINE SOLUTION: A LITERATURE SURVEY

by

S. Fredric Marsh

## ABSTRACT

Most chemical partitioning procedures are designed for acidic feed solutions. However, the high-level waste solutions in the underground storage tanks at US Department of Energy defense production sites are alkaline. Effective partitioning procedures for alkaline solutions could decrease the need to acidify these solutions and to dissolve the solids in acid, which would simplify subsequent processing and decrease the generation of secondary waste. We compile candidate technologies from our review of the chemical literature, experience, and personal contacts. We recommend several of these for evaluation.

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## I. INTRODUCTION

Although most partitioning procedures are designed for acidic solutions, a few are compatible with alkaline feed streams. If selected radioactive species or nonradioactive components of high-level waste (HLW) can be removed directly from alkaline waste solutions, significantly less radioactive and/or bulk materials would remain for additional processing. These remaining solids could be dissolved in smaller quantities of acid, yielding more concentrated acidic feed streams and decreasing the quantity of secondary waste.

To evaluate the possibilities for partitioning the hazardous components of HLW from alkaline solution, we examined the literature in *Nuclear Science Abstracts* (1948-1976), *Energy Science and Technology* (1974-present), *INSPEC* (1969-present), and *Chemical Abstracts* (1967-present). Our search was intentionally broad to reduce the chance of missing significant references.

We reviewed nearly all of these literature references and also pertinent information from personal contacts and experience. The most promising techniques and processes are summarized here. We recommend several of these for further evaluation.

## II. ION EXCHANGE

Ion exchange is the reference technology for removing cesium and technetium from alkaline HLW solution. Although specialized resins are required for cesium, conventional anion exchangers can remove technetium. Hirsch and Portock<sup>1</sup> measured the distribution of 35 different ionic species on strong-base anion exchange resin from 0.10, 1.00, and 10.0 M sodium hydroxide solution. The metal ions significantly sorbed from 1 M NaOH (and the log of their distribution

coefficients) were rhenium (4.06), technetium (3.29), molybdenum (1.49), chromium (1.34), thallium (0.18), and selenium (0.11).

### **A. Technetium**

Although technetium is readily sorbed from basic solution on strong-base anion exchange resin, its elution normally requires large volumes of 4 M nitric acid. Walker and coworkers<sup>2</sup> have shown that pertechnetate ion can be removed from this eluate by resorption on a weak-base resin, which allows the nitric acid to be recycled. Technetium can then be eluted from the weak-base resin with a small volume of 1 M sodium hydroxide solution.

### **B. Cesium**

Cesium was initially recovered and purified at Hanford using a zeolite; however, this was later replaced by Duolite ARC-359, a phenolic resin.<sup>3</sup> A resorcinol/formaldehyde resin recently developed at Savannah River can selectively sorb 0.25 mM cesium from supernatant solutions that contain as much as 6 M sodium.<sup>4</sup> The sorbed cesium can be eluted with 1 M formic acid or 0.5 M nitric acid.

### **C. Cesium and Strontium**

Because the basic supernatant tank solutions usually contain not only cesium, but also a significant amount of the strontium, there is interest in resins that might remove both elements. Varadarajan et al.<sup>5</sup> synthesized a polycondensate phenolic resin with iminodiacetic acid functional groups and demonstrated that it removes both cesium and strontium from alkaline reprocessing-plant waste.

### **D. Inorganic Exchangers**

Inorganic ion exchangers have been extensively studied for treating aqueous waste because they usually resist radiation and nitrate better than organic resins and because they often are directly compatible with final waste forms such as grout or glass. Recent studies involve molecular sieve zeolites,<sup>6</sup> zeolite-based anion exchangers,<sup>7</sup> and sorption of fission products by a variety of inorganic exchangers.<sup>8</sup> Sorption of actinides and selected fission products has been measured on antimonite, hydrous titanium oxide, manganese dioxide, zirconium phosphate, sodium copper ferrocyanate, and titanium phosphate.<sup>9</sup>

Selected fission products and activation products have been sorbed on very fine particles of sodium nickel ferrocyanate, manganese dioxide, hydrous titanium oxide, and ferric hydroxide, which were subsequently removed by ultrafiltration.<sup>10</sup> The sorption of plutonium and selected fission products and activation products from basic solution by more than 20 inorganic absorbers was independently measured.<sup>11</sup>

Schulz<sup>12</sup> reported that sodium titanate had a high affinity and capacity for multivalent cations from alkaline Hanford tank waste solutions. Schulz et al.<sup>13</sup> then demonstrated that sodium titanate could remove low concentrations of plutonium and americium from neutralized (pH 11-12) Hanford Site Plutonium Recovery Facility waste.

Ying et al.<sup>14</sup> studied sodium titanate for sorbing actinides and fission products from liquid wastes. They determined the physical and chemical properties of sodium titanate prepared with

different sodium/titanium ratios. Sodium titanate performed best when the sodium/titanium ratio was 1.5. The ion exchange capacity of material hydrolyzed in a water-acetone mixture was 5 to 6 times greater than when hydrolyzed in pure water.

Distribution coefficients of strontium, cerium, cobalt, plutonium, americium, and curium on sodium titanate from pH 2 solutions all exceeded  $10^3$ , whereas that of cesium was only 24. High concentrations of sodium suppressed the sorption of cesium. High concentrations of iron or calcium suppressed the sorption of strontium. Even millimolar concentrations of complexants such as NTA, EDTA, or DTPA significantly lowered the sorption of multivalent metals but had little effect on cesium.<sup>14</sup>

Sandia National Laboratories (SNL) has proposed sodium titanate for removing strontium and transuranics from Hanford aqueous wastes. This sorbent reportedly requires a pH of at least 3 and performs substantially better in more basic solutions. SNL claims it is "extremely efficient for removal of strontium and transuranics in highly caustic Hanford tank wastes."<sup>15</sup> More recently, collaboration between SNL and Texas A&M has produced crystalline silico-titanate exchangers that are claimed to be selective for cesium.<sup>16</sup>

## E. Composite Absorbers

Inorganic ion exchangers are sparingly soluble compounds that often, when compared with synthetic organic ion exchangers, offer higher thermal and radiation stability, and good chemical stability even in strong acids. Unfortunately, the granular and mechanical properties of many inorganic exchangers are not suited to column applications. Recently, however, inorganic sorbents have been incorporated into porous particles that are suitable for column applications.<sup>17</sup> Examples include Ionsiv<sup>TM</sup> TIE-96, a titanium oxide-loaded zeolite used to process basic waste solutions at the West Valley Reprocessing Plant.<sup>18</sup>

Composite absorbers have recently been developed by Narbutt et al. in Poland<sup>19-21</sup> and by Sebesta et al. in the Czech Republic.<sup>22-25</sup> The Polish composites incorporate hexacyanoferrate salts of titanium, nickel, and cobalt, and also hydrous antimony pentoxide in a matrix of phenolsulfonic-formaldehyde resin. The Czech composites include ammonium molybdophosphate, potassium nickel hexacyanoferrate, sodium titanate, manganese dioxide, magnesium dioxide, calcium-activated barium sulfate, and synthetic mordenite, each in a matrix of polyacrylonitrile.

Laboratory tests<sup>22</sup> with these composites have shown that sorption properties are not impaired by the binding polymer. Sorption of radionuclides on composite absorbers from actual acidic, neutral, and alkaline waste solutions has been successfully demonstrated.<sup>23</sup>

Suzuki<sup>26</sup> prepared hydrous oxides of cerium or zirconium in porous polymer beads by incorporating the selected metal alkoxide into the beads, after which the alkoxide was hydrolyzed with acid. These composites removed fluoride from solutions of pH 2 to 8.

## F. Chelating Resins

Chelating resins offer the possibility of removing selected elements that exist at trace levels in low-level waste. Ruthenium, which exists in tank waste as a nitrosyl species, has historically resisted complete removal by ion exchange. Recent work with somewhat flexible sulfur-containing copolymers<sup>27</sup> gives hope that specially designed functional groups may remove these nitrosylruthenium complexes from nuclear waste. A study of many organic copolymers,

functionalized inorganic phases, and sulfur-containing minerals identified eight sorbents that removed ruthenium from simulated basic waste solution with distribution coefficients greater than 5000.<sup>28</sup>

Sakaguchi and Nakajima have shown that tannin, which has multiple adjacent hydroxy groups, is a strong sorbent and is highly selective for uranyl ion; the reported sorption order from near-neutral solutions is  $\text{UO}_2^{2+} \gg \text{Cu}^{2+} \gg$  other ions.<sup>29-32</sup> Chinese gallotannin (tannic acid) has been immobilized on agarose gel, using the epichlorohydrin or cyanuric chloride coupling procedure.<sup>29</sup> These same investigators also found that Chinese gallotannin immobilized on various cellulosic matrices<sup>30</sup> or on matrices with multiple active amino groups<sup>31</sup> resulted in effective sorbents. Uranium and thorium subsequently were sorbed by Sakaguchi and Nakajima, who immobilized the extracted juice of unripe astringent persimmon fruit, using various aldehyde compounds.<sup>32</sup>

Mitsubishi Nuclear Fuel Co. has developed a fixed tannin sorbent to remove actinides from processing-waste water.<sup>33</sup> Another Mitsubishi process uses finely divided tannin to remove ionic and colloidal substances from alkaline nuclear-waste solution.<sup>34</sup> Mitsubishi claims that this material strongly sorbs uranium from alkaline solution, and plutonium from nitric acid. Mitsubishi also states that their sorbent removes (1) uranyl ion from basic ammonium fluoride solutions or nitric acid, (2) plutonium from nitric acid, and (3) the other transuranium (TRU) ions, after the solution pH is adjusted.

### III. PRECIPITATION

The chemical similarity of the trivalent actinides and lanthanide fission products makes their group separation quite difficult in most systems. Americium, the most abundant transplutonium element in aged waste, is usually trivalent in moderate concentrations of acid; however, Am(IV), Am(V), and Am(VI) are readily obtained in basic solution. Am(VI) is known to form a soluble complex in sodium bicarbonate.<sup>35</sup> Am(VI) also exhibits amphoteric behavior and will redissolve in ozonated 1 M NaOH.<sup>36</sup>

Koch et al. have patented an americium-lanthanide separation process, wherein Am(V) remains dissolved in 1 M NaOH, while the lanthanide elements precipitate.<sup>37</sup> Although this technique may not be selected for partitioning, we should recognize that any process that uses ozonation of basic solutions could dissolve americium.

### IV. MAGNETIC SEPARATION

Magnetic separation might be used to physically segregate precipitates, based on differences in magnetic susceptibility. Because all actinide compounds are paramagnetic, they can be separated from the diamagnetic components, which include most matrix elements. This technique was demonstrated on recycle plutonium and process residues by Avens and coworkers.<sup>38</sup> These investigators also proposed magnetic separation for soil decontamination<sup>39</sup> and remediation of Hanford tank waste.<sup>40</sup> Although complete separation is unlikely, an enriched actinide fraction could be segregated from most bulk matrix components, providing a concentrated fraction for processing and a lean fraction for disposal as low-level waste.

Harding<sup>41</sup> suggested that magnetite,  $\text{FeO} \cdot \text{Fe}_2\text{O}_3$ , could be intentionally precipitated as the coagulant for removing actinides from solution, after which a high-gradient magnetic separation could rapidly collect the actinide-containing magnetite.

## V. SOLVENT EXTRACTION

Reilly et al.<sup>42</sup> studied cobalt(III) dicarbollide,  $[(C_2B_9H_{11})_2Co]^-$ , and proposed it for extracting cesium and strontium from acidic solutions. Although little has been done with this reagent in the United States, the open literature reveals extensive investigations in the former Soviet Union and Czechoslovakia, with indications that large-scale applications were planned.

Actinide elements can dissolve, even in alkaline solution, in the presence of certain complexing ligands. Such actinides can be extracted from selected alkaline complexant solutions. Mahajan et al.<sup>43</sup> report the extraction of Pu(IV) from tartrate-containing alkaline solutions with 20% Aliquat-336 in xylene. Plutonium extraction increased with increasing tartrate concentration and was greater than 98% when the tartrate and alkali concentrations were equal.

Several Soviet investigations have studied related systems. Myasoedov et al.<sup>44</sup> studied the extraction of U(VI), Pa(V), Np(V), Th(IV), Pu(IV), lanthanides, and trivalent actinides from alkaline solutions with Aliquat-336 or alkylpyrocatechols, using oxycarbon acids or aminopoly-carboxylic acids to keep the hydrolyzable ions in solution. A later study of the extraction of transplutonium elements in similar systems<sup>45</sup> demonstrated that alkylpyrocatechols formed coordination-saturated internal complex compounds, whereas Aliquat-336 formed ionic associates. High extraction of trace levels and macro quantities of trivalent actinides was observed over a wide range of alkali concentrations.

Although these investigators evaluated only a few complexants, the underlying mechanism may be sufficiently general that other complexants could be suitable synergists. If so, this concept might have application to the complexant concentrate tanks and might allow many actinides to be extracted directly from the tank solutions.

Because some alkaline tank solutions have been ventilated with air for many decades, it is likely that these solutions contain high levels of carbonate. One study of the extraction of actinides from carbonate solution by primary amines<sup>46</sup> is representative of many others not cited here.

Lanthanides and trivalent actinides have been partitioned into two separate groups from actual HLW by a reverse TALSPEAK process,<sup>47</sup> which extracts these elements into di-(2-ethylhexyl)phosphoric acid (HDEPA) from aqueous nitrate media. Japanese researchers<sup>48</sup> have replaced HDEPA with di-isodecylphosphoric acid, which achieves similar group separations from higher concentrations of nitric acid.

Because much of the solid phase in Hanford tanks could be nitrate salts, an intriguing possibility is the extraction of selected components directly from these salts. The distribution coefficients of Co(II), Eu(III), Nd(III), Am(III), Cm(III), Np(V), Np(VI), and U(VI) into TBP, from a  $LiNO_3$ - $KNO_3$  eutectic mixture at 150°C, were 100 to 1000 times higher than those from aqueous nitrate systems.<sup>49</sup> French investigators<sup>50</sup> studied the extraction of transplutonium elements and Eu(III) from molten  $LiNO_3$ - $KNO_3$  into a series of diphenyldiphosphine-dioxides and found Eu(III)/Am(III) separation factors greater than  $10^4$ .

Pure sodium nitrate melts at 306°C. Aluminum nitrate, another major component of tank waste, melts at 70°C. It is possible that some of the nitrate salt mixtures in Hanford waste tanks may be low-melting eutectics that allow multivalent metal ions to be extracted in highly concentrated form. Even moderate warming of the mixed hydrated salts might yield a solution with very high nitrate concentration. Both options offer the possibility of highly selective separations between the lanthanide fission products and the trivalent actinide elements.

Crown ethers and related compounds have been proposed for partitioning various components of HLW tanks. Yakshin<sup>51</sup> studied more than 50 different structures of cyclic and acyclic

polyethers containing aromatic, aliphatic, and cycloaliphatic fragments, and showed how specific substituents can increase the extraction selectivity and separation of chemically similar elements.

The ORNL Chemistry Division<sup>52</sup> has systematically studied the separate effects of the extractant structure and of synergistic combinations of macrocyclic (crown ether) ligands and organic (sulfonic, carboxylic) acids. For example, detailed equilibrium measurements were made on the extraction of alkali metals and alkaline earths by crown ethers that also incorporate a sulfonic acid or a carboxylic acid.

## VI. RECOMMENDATIONS

1. The direct solvent extraction of actinides and some fission products from complexant-containing alkaline waste solutions at Hanford is an attractive possibility. Solutions containing U(VI), Pu(IV), and Am(III) could be prepared to simulate the composition of these wastes. The simulated solutions could be contacted with Aliquat-336 and other potentially useful extractants to determine how actinides distribute in their most common oxidation states. Systems that show promise with simulated solutions could then be evaluated with actual solutions from Hanford tanks.

2. Extracting multivalent metal ions (with thermally stable extractants and solvents) from molten nitrate salt systems might be used to remove selected components from the bulk salt materials and should be evaluated. Specifically, the composition and melting point of nitrate salt combinations in Hanford tanks (mixtures of salt cake and sludge layers) should be determined. Safety issues involved with heating tank solids must, of course, be given the highest priority before and during any such investigations.

3. Samples of the composite sorbents now being produced in Poland and the Czech Republic should be obtained and evaluated. Dr. J. Narbutt, of the Institute of Nuclear Chemistry and Technology in Warsaw, and Dr. Ferdinand Sebesta, of the Technical University of Prague, who are developing these new materials, have provided samples for evaluation at Los Alamos.

4. The insoluble tannin sorbent from Mitsubishi Nuclear Fuels should also be evaluated with simulated Hanford tank waste solution. If successful, the tests should be repeated with actual Hanford tank waste. The fact that a US DOE delegation learned of this sorbent during an information-gathering trip to Japan in July of 1992 demonstrates one of the benefits of such international contacts.

## ACKNOWLEDGEMENT

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