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Introduction

Over 100 commercial providers with mixed-waste treatability capabilities exist in the USA. The maturity level of these technologies varies from a bench scale to a pilot or a commercial scale. The techniques include deactivation, chemical oxidation, recovery of metals, stabilization, vitrification, incineration, biodegradation, and chemical extraction [INEL-96/0249]. This report focuses on the use of resins to remove actinides and heavy metals from aqueous waste streams. Only the literature that described resins with high removing efficiency are presented here. The majority of the literature reviewed are proceedings and national or international reports ordered through the Berkeley Lab library. Some of the reports that we requested have not yet arrived. Only a few papers were found in the open literature (journals or magazines). Although this report does not include all existing references, it provides an accurate assessment of efficient resins to be considered for waste minimization procedures.

Literature Review

A screening study was performed by Marsh et al. (1994) in a collaboration between Los Alamos and Sandia National Laboratories. The study evaluates 63 commercially available or experimental absorber materials for their ability to remove hazardous components from high-level waste (HLW). The absorbers included cation and anion exchange resins, inorganic exchangers, composite absorbers, and a series of liquid extractants sorbed on porous support-beads. Synthetic inorganic ion exchangers have higher selectivity for sorbing certain ions compared with synthetic organic ion exchangers; they also have greater thermal and radiation stability, and good chemical stability even in strongly acidic media. Their disadvantage is their unsuitable granular and mechanical properties that limit their use in column applications, but there are several methods to modify the inorganic ion exchanger for use in packed-bed columns. When the ionic exchanger is embedded in a binding polymer, the resultant materials is called a composite absorber. Its main application is the treatment of liquid radioactive wastes (Sebesta and John, 1995). Composite absorbers allow the treatment of various types of waste and underground waters contaminated with heavy and toxic metals.

Marsh et al. (1994) tested the 63 absorbers with simulated acid-dissolved sludge and alkaline supernate from underground storage tank 102-SY at the Hanford site. Table 1 summarizes the highest distribution coefficients (K_d) obtained with a variety of absorbers. In a typical experiment, 250 mg of air-dried absorber was contacted with 6 mL of the solution of interest. Radioactive tracers were added and gamma spectrometry was selected for measuring the distribution of elements. The contact time varied from 30 minutes to 6 hours and did not seem to affect

significantly the results. K_d 's reported in Table 1 are all obtained with contact time of 6 hours. K_d is defined as follows:

$$K_d = ((Pr - Po)/Po)(S/A),$$

where Pr = measured precontact activity

Po = measured postcontact activity

S = milliliters of solution contacted, and

A = grams of dry absorber contacted.

Table 1: Distribution Coefficients (Kd) Obtained with a Variety of Absorbers

Absorber	Kd for selected elements						
	Cs	Sr	Tc	Co	U	Pu	Am
SNL-HTO, b	1.4	>4200	<0.1	1.0	>17,000		779
SNL/CST 111	>5000, a	>4600, b					
SNL/CST 84	>11K, a						
AMP-PAN	>5000, a						
Aliquat™ 336			572, a 651, b				
Cyanex™ 923						199, a	
LANL-TiP					79, b		908, b
MnO-PAN		>8000, b			15, b		>1000, b
JSK-3						1173, a	
Bone Char		>11K, b					3084, b

a: acid solution (pH=0.6); b: basic solution (pH=13.9)

SNL/HTO: amorphous hydrous titanium oxide (Sandia National Laboratories)

SNL/CST 111: crystalline silico-titanate (Sandia National Laboratories)

SNL/CST 84: crystalline silico-titanate (Sandia National Laboratories)

AMP-PAN: ammonium molybdophosphate (polyacrylonitrile matrix, Czech Republic)

Aliquat™ 336: Los Alamos synthesized cobalt dicarbollide

Cyanex™ 923: trialkylphosphonic oxide (Los Alamos)

LANL TiP: titanium phosphate (Los Alamos)

MnO-PAN: manganese dioxide (polyacrylonitrile matrix, Czech Republic)

JSK-3: anion resin

A complementary study by Marsh et al. (1995) showed that the presence of irradiation-degraded organic materials such as EDTA in Hanford simulated solutions significantly decreases the K_d values for Sr sorption, while the sorption of Cs and Tc was not affected.

Haefner, from Lockheed Idaho Technology Company, Idaho Falls, reported the use of ion exchange resins for removal of cesium, cobalt, lead and mercury from the waste streams accumulated at the Idaho National Engineering Laboratory. The study indicated that Purolite S-920 resin (a hydrogen-form, chelating-type resin with thiouronium functional groups) could effectively remove mercury, while Rohm and Haas's Amberlite 200-CH (a strong acid, cationic- hydrogen-form resin) was used for lead and radionuclide removal (Cs and Co). Mercury was removed to below 0.4 microgram/L. Lead was removed to below 0.036 mg/L. These two resins could also remove chromium, cadmium, and barium. The presence of other elements in the waste-water (such as Ca, Cu, Fe, Mn, Mg, P, K, Na) did not seem to affect the removal of the metals of interest. A large scale design and treatment was performed. 129,000 L of acidic waste water (pH = 2.8) were treated and all Resource Conservation and Recovery Act (RCRA) criteria were met. Resins from the spent Amberlite columns were sent for toxic characteristic leaching procedure (TCLP) metal analysis. All concentrations were below disposal limits.

A Diphonix^R resin has been developed recently by the Separation Chemistry Group of the Chemistry Division of Argonne National Laboratory (ANL) in collaboration with the Department of Chemistry, University of Tennessee. The resin contains diphosphonic and sulfonic acid groups chemically bonded to a styrenic-based polymer matrix. The resin exhibits a strong affinity for actinides, especially in the tetra and hexavalent oxidation states (Chiarizia et al., 1997). Actinide uptake by the resin is more efficient in acidic solutions. It also has high affinity for multivalent ions such as Al(III), Cr(III) and especially Fe(III) from acidic and neutral solutions. The higher distribution coefficients (D) for Am(III), Np(IV), Pu(IV) and U(VI) in HNO_3 solutions are described in Table 2. D is defined as the ratio between the metal concentrations in the resin phase and in the solution in equilibrium with the resin.

Table 2. Distribution Coefficients (D) for Selected Metals Using Diphonix^R Resin

Actinide	[HNO ₃] M	D (mL/g)
Am(III)	0.1	1.5 x 10 ⁶
Np(IV)	0.4	9.0 x 10 ⁴
Pu(IV)	1.0	1.0x 10 ⁵
U(VI)	0.01	1.0 x 10 ⁶
Fe(III)	1.0	2.0x 10 ⁵
Cr(III)	0.04	6.0 x 10 ³
Ba(II)	0.02	9.0 x 10 ⁴
Pb(II)	pH = 7	4.0 x 10 ⁴
Ag(I)	0.02	8.0 x 10 ⁴

Apatite minerals (calcium phosphate) was used to precipitate lead and other heavy metals. Metal sequestered in apatite have great durability and leach resistance that significantly exceeds other chemically stabilized waste forms. Apatite in nature acts like a chemical filter for metals. The reactions between the apatite and metals are rapid and therefore the treatment is effective immediately (Wright et al., 1995). Wright et al. conducted studies on the sorption and desorption of heavy metals on and from North Carolina mineral apatite, specifically for Pb, Cd, and Zn. Approximately 100% of the Pb was removed from solutions, representing a capacity of 151 mg of Pb/g of apatite, while 49% Cd and 29% Zn were removed.

Studies by Robinson et al. at Oak Ridge National Laboratory have shown that chabazite-zeolite, an inorganic aluminosilicate, has an extremely high capacity for removal of Cs-137 and is very efficient in removing Sr-90 in the presence of calcium and magnesium. The contaminated zeolite must be disposed of as solid radioactive waste because no methods have been developed yet for its regeneration. Several pilot-scale and near-full-scale zeolite column were tested.

A full-scale application for the use of cesium-selective synthetic mordenite (zeolite) and organic ion exchange materials was described by Jacob et al. (1987). The zeolite was synthesized with sodium

exchangeable cation and with a high silica-to-alumina ratio of at least 10/1. The organic ion-exchange resins selected to be mixed with zeolite were a sulfonated styren-divinylbenzen gel (strong acid cation) and a Type I quaternary on a styren-divinylbenzene gel (strong base anion). The pH of the feed and effluent waste ranged between 8 and 5.5.

Disposal of Spent Resins

When the resins are spent, they can be disposed of as low level radioactive wastes. Disposal regulations require that low level wastes contain less than 1 volume % free-standing liquid in the disposal container. Water can be removed by centrifugation of the resins or pumping on the resins. Rockwell International Corporation has developed a volume reduction system using a heated-air spray dryer (Gay et al., 1984). Full-scale tests were conducted in which slurries of sodium sulfate, boric acid, and powdered ion exchange resin were dried completely. This treatment led to a significant cost savings by reducing the volume and weight of waste. Another technique to reduce the spent ion-exchange resins was used by JGC Corporation in Japan (Kuribayashi and Yamanaka, 1984). The technique is based on a wet oxidation method using hydrogen peroxide at 100 °C. Incineration is one of the techniques that can also be used to minimize the volume of spent resins. An extensive survey of the available literature on LLW incineration was conducted by S. W. Long (1990). Specific types of incineration technologies including designation of the kind of wastes that can be processed, the magnitudes of volume reduction that are achievable in typical operation, and the requirements for ash handling and off-gas filtering and scrubbing are described. The reader is referred to that report for details.

Availability and Cost of Resins

This area of search is still in progress. The information about the resins cost were obtained by telephone conversations. Diphonix^R resin and apatite seem to be the more available and cost-effective resins. The price of Diphonix^R resin is \$ 510 per 500 grams. If, for example, we have a Liter of an acid waste solution that contains 0.1 mM Pu(IV), only 10 mg of the resin will be needed to extract Pu(IV). Apatite is less expensive than Diphonix^R resin but we do not have the corresponding distribution coefficients for actinides.

Conclusion

The nature of wastes at the different DOE sites vary significantly leading to variations in waste treatment methods. For all the full scale applications that we reviewed, preliminary bench-scale studies have been performed to select the best method for treating a specific waste.

The higher distribution coefficients for actinide were obtained using Diphonix^R resin. Apatite has also high potential for actinide removal. We suggest that we perform few experiments with these two resins to examine their efficiency in treating Berkeley Lab wastes. If results from these experiments are encouraging, we will develop the appropriate treatment scheme. A full-scale treatment can be carried out that has the objectives of 1) reducing the volume of liquid waste and its contamination level, 2) minimizing the amount of secondary waste generated, and 3) achieving large cost savings.

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