2

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TREATMENT OF CONTAMINATED WASTEWATER AT OAK RIDGE NATIONAL LABORATORY

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Introduction

Oak Ridge National Laboratory (ORNL), an energy research and radioisotope production facility, operates two centralized liquid waste treatment systems, one for liquid low-level waste (LLLW) system and the other for process waste (PW). New regulatory and waste minimization requirements have led ORNL to consider zeolite ion exchangers for removing cesium and strontium from LLLW and PW streams for their economic advantages, selective molecular sieve properties, and ease of disposal. Natural and synthetic zeolites have been compared with inorganic and organic ion exchangers for these applications.

Waste Stream Definition

The streams tested include: (1) Melton Valley Storage Tank (MVST) supernate, legacy LLLW that was concentrated by evaporation, (2) newly-generated liquid low-level waste (NGLLLW), (3) Cleanex raffinate solution, and (4) newly-generated PW. LLLW is produced as a result of reactor and research operations. ORNL has approximately 500,000 gal of legacy LLLW stored in the MVST, and an additional 20,000 gal of NGLLLW is generated each year. Cleanex raffinate, a major NGLLLW contributor, is generated from production of transuranic isotopes. The PW system collects and treats 70,000,000 gal/yr of slightly contaminated dilute wastewater, including laboratory wastewater, once-through cooling water, and groundwater.

Simulated wastewaters have been used to test ion exchange materials. The MVST supernate contains 4-5 $\underline{N}$ $NaNO_3$, 5 x 10^{-7} $\underline{N}$ Cs, and 1 x 10^{-4} $\underline{N}$ Sr, and significant quantities of potassium, calcium, and magnesium salts. NGLLLW is more dilute and contains primarily 0.3 $\underline{N}$ NaOH, 0.6 $\underline{N}$ Na_2CO_3, 9 x 10^{-6} $\underline{N}$ Cs, and 2 x 10^{-6} $\underline{N}$ Sr. It does not contain significant quantities of potassium,

cobalt, calcium, or magnesium salts. Both waste streams have a pH of approximately 13. The Cleanex raffinate has a composition of 0.03 N HCl, 0.04 N LiCl, 0.10 N NaCl, 9.2 X 10^{-4} N Cs, and 2.1 X 10^{-3} N Sr with a pH of 1.5. Process wastewater contains 2 x 10^{-3} N Ca, 6 x 10^{-4} N Mg, 2 x 10^{-4} N Na, 2 x 10^{-6} N Sr, and 1 x 10^{-12} N Cs. The pH of PW is near neutral. Trace amounts of Cs-137 and Sr-85 were added to the simulated wastewaters for analytical purposes.

Results

Sorption of strontium and cesium from MVST waste and NGLLLW was tested using several chelating ion-exchange resins, such as a resorcinol-formaldehyde resin, and inorganic exchangers, such as zirconium phosphate, titanium phosphate, sodium titanate, hexacyanoferrate compounds, and zeolites. Natural clinoptilolites and natural and synthetic chabazites were not effective for removing cesium or strontium from simulated MVST supernate or NGLLLW over a pH range of 8 to 13. Sodium titanate was the most successful material for removing strontium, and hexacyanoferrate compounds were most effective for removing cesium. The latter compounds are under further consideration for both waste streams.

Simulated Cleanex raffinate solution was tested with natural clinoptilolites, natural and synthetic chabazites, titanium monohydrogen phosphate, zirconium monohydrogen phosphate, hexacyanoferrate compounds, polyantimonic acid and molecular sieves. The clinoptilolite removed some of the cesium, but none of the zeolites removed strontium. The polyantimonic acid was the most successful material for removing strontium, and hexacyanoferrate compounds were most effective for removing cesium. Zeolites are no longer being considered for this application.

Natural clinoptilolite, natural and synthetic chabazites, erionite, ferriorite, mordenite, and type A zeolites, strong-acid and weak-acid cation resins, and glass-based resins were tested for treating PW. The chabazite zeolites were the only materials which removed both cesium and strontium. Natural and synthetic chabazites with high sodium contents performed best. A new treatment facility is being designed to treat PW using a natural chabazite.

Summary

Zeolites have not been effective for removing radionuclides from ORNL waste streams which contain high concentrations of nonradioactive salts. Chabazite zeolites are the most effective material for removing trace quantities of cesium and strontium from ORNL process wastewater. The existing PW treatment plant will be replaced with a system consisting of filtration and natural chabazite ion exchange columns with estimated secondary waste generation rates approximately 50% of those from the existing plant. Pilot-scale studies are underway to design a 300-gal/min treatment facility.