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TESTING OF HEXACYANOFERRATES FOR DECONTAMINATION OF RADIOACTIVE WASTEWATERS AT OAK RIDGE NATIONAL LABORATORY

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ABSTRACT

Liquid low-level radioactive waste (LLLW) and slightly contaminated process wastewater at Oak Ridge National Laboratory has been tested for removal of ^{137}Cs using several transition-metal hexacyanoferrate compounds. The method being developed is scavenging precipitation, in which ion exchange particles are mixed with the solutions and allowed to settle. Decontamination factors of 35 for simulated LLLW and 12 for process wastewater were achieved.

INTRODUCTION

Oak Ridge National Laboratory (ORNL), in Oak Ridge, Tennessee, is operated by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy (DOE). Many of the major ORNL activities involve research and development that supports the DOE's programs in fusion, fission, and other energy technologies. In carrying out these programs, a wide variety of liquid wastewater streams are generated. Liquid low-level waste (LLLW), generated at radiochemical production facilities and laboratories, is generally highly contaminated with fission products and transuranium elements. A more dilute contaminated stream, process wastewater, is also generated. Improved treatment methods are being investigated for both of the above waste streams.

BACKGROUND

The LLLW system currently treats over 400,000 gal of waste per year, reducing the volume by evaporation to approximately 12,500 gal of waste concentrate. The waste concentrate is stored in eight 50,000 gal vaulted underground storage tanks known as the Melton Valley Storage Tanks (MVSTs). The LLLW concentrate has been accumulating in the storage tanks since 1984, and storage space is now very limited. To create storage space for near-term programs, the LLLW supernate liquids are being treated by solidifying in concrete.(1) It was anticipated that the solidified waste could be disposed of in a proposed new DOE Class L-II solid waste storage area on the Oak Ridge Reservation. However, the preliminary waste acceptance criteria (WAC) for the new storage area indicate that the LLLW supernate must be treated to remove some of the radionuclides before it can be solidified and accepted as Class L-II waste. Table 1 lists the proposed Class L-II WAC and the decontamination factors required to meet the criteria.

Previous study has indicated that transition-metal hexacyanoferrate compounds have been successful in selective removal of cesium from simulated LLLW supernate.(2) A study was undertaken to determine the feasibility of applying these hexacyanoferrate compounds directly to the MVSTs for in-situ removal of cesium. If successful, the use of this treatment method could greatly reduce the quantity of greater-than-Class II waste which might otherwise be produced during the solidification campaign.

Process wastewater generated at ORNL is chemically similar to groundwater contaminated with low levels of radionuclides. The primary radioactive contaminants of the wastewater are ^{90}Sr and ^{137}Cs at typical concentrations of 20.3 μCi/m^3 and 1.9

TABLE 1. DECONTAMINATION REQUIREMENTS FOR MVST SUPERNATE RADIONUCLIDES TO MEET PROPOSED CLASS-II WASTE ACCEPTANCE CRITERIA

Radionuclide	Supernate Composition[a]		Proposed Class L-II WAC[a]	Decontamination Factors[b]	
	Tank W-29	Tank W-30		Tank W-29	Tank W-30
Gr. Alpha	2.7E+01	2.7E+01	1.04E+06	0	0
^{14}C	2.45E+03	2.38E+03	2.01E+02	8	8
^{60}Co	1.68E+04	1.32E+04	1.02E+12	0	0
^{137}Cs	5.94E+06	5.07E+06	2.53E+05	16	13
^{90}Sr	1.9E+05	1.82E+05	8.88E+05	0	0

[a] Concentrations are reported in μCi/m^3

[b] Decontamination factors assume a 50% volume increase in waste during solidification.

μCi/m^3, respectively. The Process Waste Treatment Plant (PWTP) currently uses a chemical water softening process followed by treatment with an organic ion-exchange resin for removal of ^{90}Sr from process wastewater at an average flow rate of 8.8 L/s (140 gal/min). The ^{137}Cs concentration of process wastewater varies by a significant amount and periodically approaches the limit of 3 μCi/m^3 set in DOE Order 5400.5.(3) An exhaustive study performed in 1986 and 1987 to determine improved methods for removal of both ^{90}Sr and ^{137}Cs from process wastewater.(4) An inorganic natural chabazite zeolite was determined to be the best material for treatment. More recent study, however, has indicated that hexacyanoferrates may be used to remove ^{137}Cs from process waste streams.(5) It was decided to test these materials and compare performance with the zeolites.

LLLW TESTING

Following evaporation and cooling in the MVSTs, the LLLW waste concentrate separates into sludge and supernate phases. The supernate is 4 to 5 M sodium nitrate

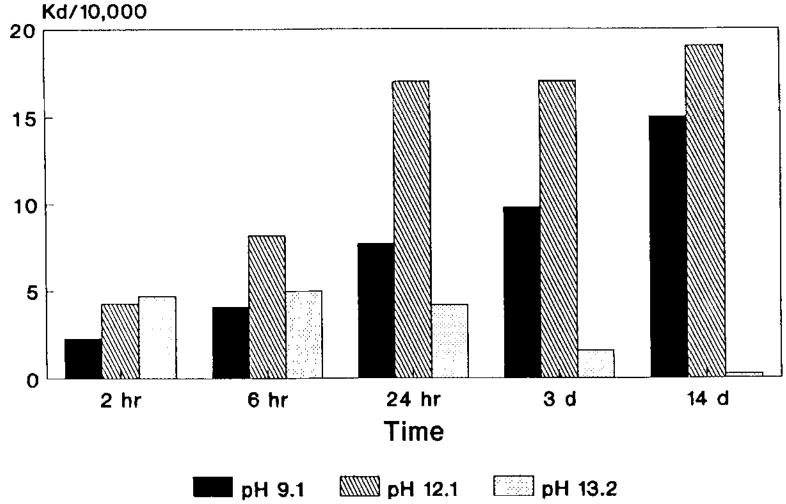

FIGURE 1. Cesium distribution coefficients using granular KCFC at three pH levels.

contaminated with soluble radionuclides, primarily ^{137}Cs and ^{90}Sr, while the sludge consists of precipitated carbonates and hydroxides of metals, and transuranic elements. The Waste Handling and Packaging Plant (WHPP) has been proposed as a FY 1994 project to process this waste for shipment to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM.(6) Scoping studies performed in support of WHPP design have indicated that hexacyanoferrate ion exchangers could result in decontamination factors of 10,000 for cesium.(2) Cesium distribution coefficients obtained in a related development study(7) are shown in Figure 1. They indicate that granular potassium-cobalt hexacyanoferrate (KCFC) used in a simulated MVST supernate solution is effective at high pH for extended time periods. Based on the results of this study, it was decided to evaluate the feasibility of adding KCFC exchangers directly to the MVSTs for in situ decontamination prior to the next solidification project. Bench-scale tests were performed to evaluate the effects of supernate composition, supernate pH, hexacyanoferrate production process, tank mixing rates, particle settling rates, and filtration methods.

TABLE 2. COMPOSITION OF SIMULATED SUPERNATES

Component	Concentration (mol/L)		
	High-Salt	Tank W-29	$NaNO_3$
$NaNO_3$	5.98	3.9	3.9
NaOH	0.01	0.24	0.65
Na_2CO_3	0.20	0.14	
NaCl	0.08	0.10	
KNO_3	1.37	0.24	
$CaCO_3$		0.0001	
$Ca(OH)_2$	0.19		
$Mg(OH)_2$	0.07		
$As(NO_3)_3$		0.005	
$Zn(NO_3)_2$		0.001	
pH	13.05	12.7	13.05, 10.7

The waste solutions stored in the MVSTs were sampled and characterized in 1989 to provide information for design of the WHPP and LLLW solidification projects.(8) This information was used to prepare four simulated supernate solutions to use for sorption measurements. The compositions of the simulated wastes are shown in Table 2. The high-salt supernate represented the "worst case" waste solution in that it contained a higher level of soluble salts than any of the wastes in the MVSTs. The Tank W-29 simulated waste was formulated to match the composition of waste in MVST Tank W-29. Two additional solutions containing 3.9 mol/L $NaNO_3$ were adjusted to pH 13.0 and 10.7 and used to determine the effect of potassium and pH on sorption of cesium by the hexacyanoferrates. The cesium concentration of the solutions was adjusted with CsCl to 0.069 mg/L, which is equivalent to the ^{137}Cs concentration of 5.94×10^6 $\mu Ci/m^3$ reported for Tank W-29.(8) The solutions were also traced with ^{137}Cs.

Four samples of KCFC were tested. Samples A and B were obtained from chemical suppliers, and samples C and D were prepared at ORNL. The ORNL samples are two batches prepared by the method of Prout, Russell, and Groh.(9) This method was also used to prepare the KCFC used in earlier ORNL studies.(7) The granular KCFC samples were prepared in the size range of 20 to 50 mesh (840 to 297 µm). The particles broke into much smaller pieces as soon as they came in contact with the strongly alkaline, pH-13, simulated supernate solutions, but not with the $NaNO_3$ solution at pH 10.7.

LLLW Testing Procedure

Cesium sorption tests were performed using the batch equilibration method. For these tests, weighed amounts of granular KCFC samples were added to centrifuge tubes containing simulated supernate solutions at a solution/solid ratio of 1000/1. The solids and solutions were mixed for a predetermined period of time and allowed to settle for a measured time before aliquots were removed for counting. Centrifugation was performed for 30 min at 5000 rcf, and filtration was through 0.20-µm membranes using a syringe filter. Cesium removal was measured by counting the ^{137}Cs in the treated solutions and comparing the count rate with that of the untreated supernate. The results were calculated as the decontamination factor, $DF = C_i / C_f$, where C_i is the initial count rate and C_f is the final count rate.

LLLW Test Results

The cesium decontamination factors were affected by the solution pH and composition and by the source of the KCFC. KCFC from each of the four different sources was tested in all four of the different simulated waste solutions. Figure 2 shows the average decontamination factors obtained in the solutions after a mixing time of 1 d. Each KCFC sample behaved differently. The commercially available KCFC sample A was almost completely ineffective with all except the pH 10.7 $NaNO_3$ solution, where a decontamination factor of 23 was measured. KCFC sample B had decontamination factors of about 11 for the pH 13.0 $NaNO_3$ solution and about 55 for the pH 10.7 $NaNO_3$ solution. These compare to decontamination factors of 9 and 27.6 respectively with KCFC sample C, and 3.7 and 17.6 respectively with KCFC

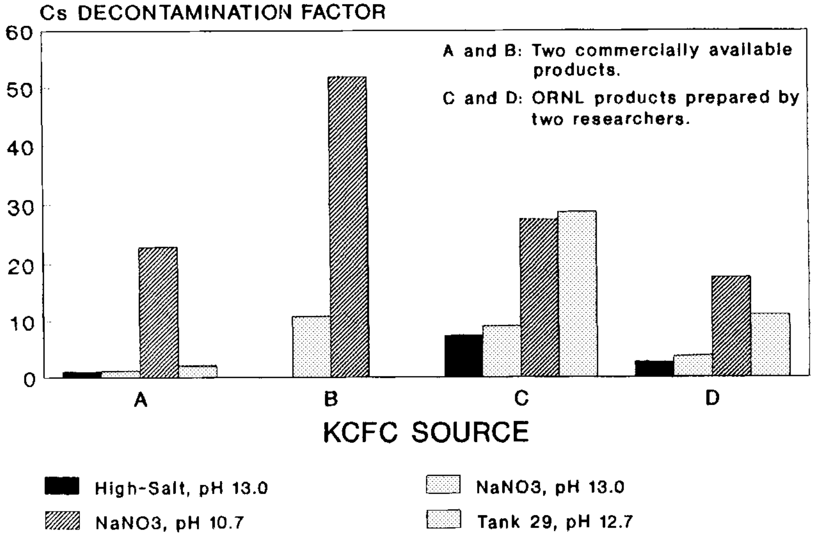

FIGURE 2. Effects of KCFC source and supernate composition on cesium decontamination.

sample D for the same solutions. Although samples C and D were prepared by the same method, decontamination factors for all four solutions were lower with sample D than with sample C. It is obvious from these tests that one cannot expect good cesium removal performance from all sources or batches of KCFC. W. E. Prout, et al,(9) showed that the KCFC is typically a mixture of several different hexacyanoferrates with varying molar ratios of potassium and cobalt. The method of preparation may have an effect on the chemical nature, crystalline structure, and particle size of the KCFC all of which may influence the cesium exchange properties.

Cesium removal for the pH 13.0 $NaNO_3$ solution was similar to that of the high-salt solution even though the solution compositions are significantly different. Cesium removal was generally better for the pH-10.7 $NaNO_3$ solution and the Tank W-29 solution (pH 12.7), indicating that the pH has a greater effect on cesium removal than does solution composition (i.e., the cations present and their concentrations).

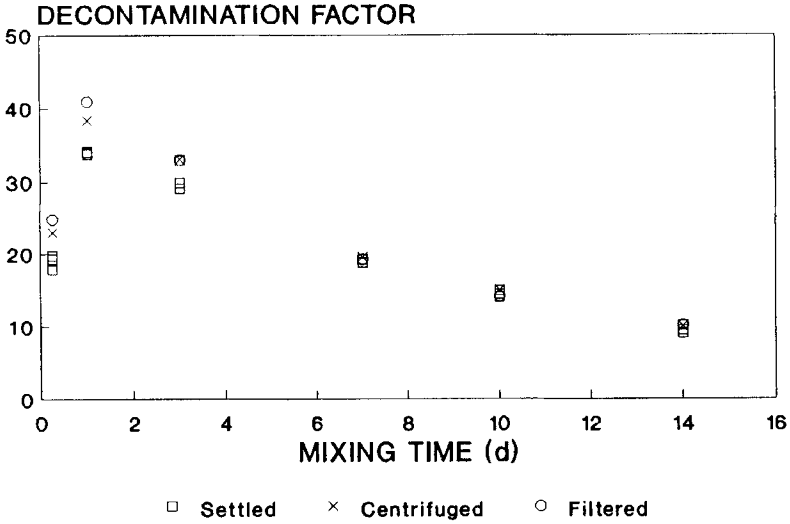

FIGURE 3. Effect of mixing time on cesium decontamination.

For in situ application of the KCFC, it will be important that the cesium, once removed from the solution, remain stable in the settled sludge for several months. To determine the effect of mixing time on cesium decontamination, simulated Tank 29 supernate was mixed with KCFC sample C for up to 14 d. Aliquots of the supernate for each mixing time were measured for cesium content after a 1.5-h settling period, after 30 min centrifugation, and after filtration through a 0.2-μm membrane. As shown in Figure 3, the decontamination factors were at a maximum after 1 to 2 d of mixing and then decreased with longer mixing times. Decontamination factors are slightly higher in centrifuged samples than in settled samples after 1 d of mixing. There was no difference between the settled, centrifuged, or filtered samples after 6 d or more of mixing. This indicates that settling might provide adequate solid/liquid separation. The results show that the KCFC is effective for only a limited time when it is in contact with highly alkaline solutions and that the optimum mixing time appears to be about 1 d. The decrease in the decontamination factor for longer mixing times is possibly due to decomposition of the KCFC to cobalt hydroxide and potassium hexacyanoferrate as a result of extended contact with the highly alkaline solution. This behavior was indicated in earlier studies by Campbell(7) which also showed that KCFC is stable for longer times at lower pH levels.

Conclusions and Recommendations for LLLW Tests

The test results indicate that under certain circumstances, MVST supernate could be decontaminated using KCFC to produce a Class L-II waste form after supernate solidification. However, the decontamination cannot be performed in situ by adding KCFC to the tanks prior to a solidification campaign due to the instability of the KCFC at the high pH of the supernate. The KCFC begins to decompose after only a few days in contact with solutions at pH 13 and would lose most or all of its effectiveness before the treated solution could be removed from the storage tanks and solidified. It is not feasible to adjust the pH of the supernates in the storage tanks to increase the cesium decontamination or the KCFC stability because of the possibility of dissolving part of the sludge which, in most cases, contains transuranic wastes. Decontamination of the supernates should be possible if they are removed from the tanks and treated under more-controlled conditions than are possible in the tanks. The envisioned treatment would include adjustment of the pH to an optimum range and batch treatment with KCFC in a stirred tank to remove cesium, followed by separation of solids from the solution by filtration or other means, and ultimate disposal of the treated liquid and the KCFC solids. Cesium sorption with KCFC has been shown in other studies to be more effective at lower pH, and significantly lower amounts of KCFC would likely be required. The pH could be adjusted outside the storage tank without the risk of dissolving any of the sludge. Unit operations such as filtration would be more effective than settling, which is the only phase-separation option available for treatment in the storage tanks. Treatment at lower pH could produce a more stable final product in a smaller volume for ultimate disposal.

Before treatment of the supernate outside the storage tanks, development work would be needed to 1) determine the optimum pH range for cesium removal and the amounts of KCFC needed for adequate removal of cesium from the supernates, 2) work out methods to prepare (or obtain) KCFC that is dependably effective, 3) determine the stability of the Cs-KCFC complex, 4) develop methods for separating the solids from the treated solution, and 5) determine some of the properties and ultimate disposal options for both the solids and the treated liquids. Scaleup studies will be needed also to determine the effects on cesium decontamination on variables such as particle size, mixing time, and power input to the mixer. Some of the storage tanks contain solutions higher in ^{14}C concentration than is allowed by Class L-II limits. Studies of methods to reduce these concentrations to within allowable limits will be needed in addition to the cesium decontamination studies.

PROCESS WASTEWATER TESTING

In an initial scoping study for removing ^{137}Cs from process wastewater,(5) four different hexacyanoferrate compounds (potassium/cobalt, sodium/cobalt, potassium/nickel, and sodium/nickel hexacyanoferrate) were compared. Based on these tests, sodium nickel hexacyanoferrate (NaNiFC) was selected for further evaluation. To determine NaNiFC performance under more realistic conditions, a larger-scale, continuous-treatment system was constructed and operated.

Pilot Scale Tests

A small-scale, continuous-treatment system was prepared that was designed to roughly simulate the PWTP softening and clarification process. A flow diagram of the system is shown in Figure 4. The test system consisted of a relatively simple train of small chemical feed and process tanks. Actual process wastewater was delivered from the plant to the system at an average flow of 0.5 L/min. A metering pump was used to control flow to the rapid-mix vessel where the wastewater combined with the feed chemicals, which included sodium hydroxide for adjusting pH to 11.5, ferric sulfate (2.5 mg/L as Fe in wastewater) and an organic polymer (0.6 mg/L) for flocculation of precipitates, and the NaNiFC slurry. To minimize dilution of the feed wastewater, all chemical feed flow rates were maintained at or below 4 mL/min. To increase sludge contact time, a 5-gal slow-mix vessel was added to the treatment system following the rapid-mix vessel. The slow-mix vessel overflowed to a cone-shaped clarifier. The

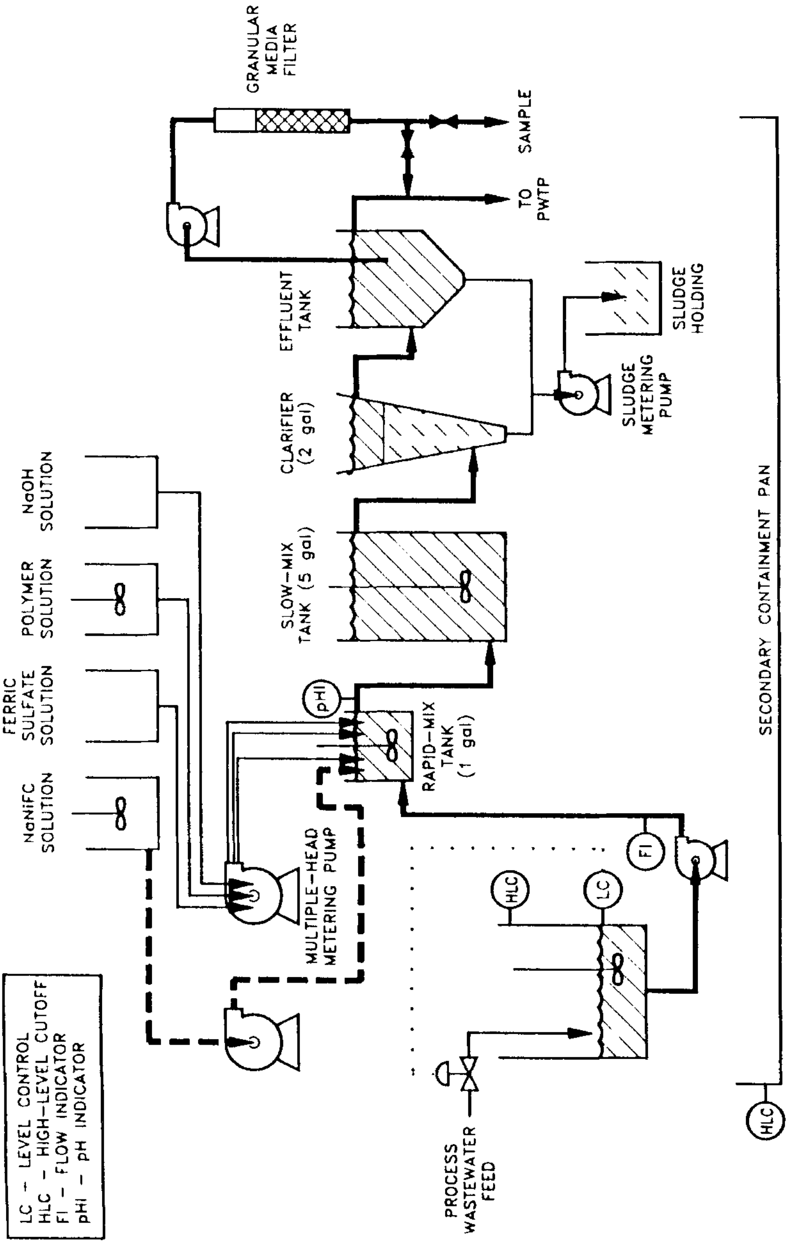

FIGURE 4. Diagram of pilot scale test system.

rapid-mix tank, slow-mix tank, and clarifier allowed for a total wastewater/NaNiFC reaction time of 1 h. The supernate from the clarifier overflowed to the effluent tank. A peristaltic pump was used to periodically remove sludge from the clarifier and effluent tanks. A flow-through filter and pump were used to filter the effluent stream, if desired. The system effluent was discharged to the process wastewater collection system.

To prepare the NaNiFC slurries, reagent-grade sodium ferrocyanide, $Na_4Fe(CN)_6+6H_2O$, and nickel nitrate, $NiNO_3+6H_2O$, were used to prepare 0.3 M solutions of the two reactants. Previous study(7) indicated that the reaction is usually not stoichiometric and that a molar excess of the nickel salt is advisable. In this case, a 70% molar excess of nickel nitrate was used. The sodium ferrocyanide solution was slowly added to the nickel nitrate solution with vigorous stirring. The product was a light-green, evenly-suspended slurry of the NaNiFC. Dilution water was added to the NaNiFC slurry so that it could be metered into the process wastewater stream at a flow rate of 3.5 mL/min. In a typical test, the system was started at the beginning of the week and allowed to run for about 12 h to reach steady state without NaNiFC addition. After sampling to collect baseline data, the addition of the NaNiFC slurry began. Periodic samples of the feedwater and effluent wastewater were collected. The system was allowed to run continuously for 90 h. The extended test duration was considered necessary for data collection and performance evaluation during the inherent changes in process wastewater composition.

The system operated reasonably well during the tests considering problems caused by changing feed wastewater composition. The ^{137}Cs level varied from 1.4 to 4.0 $\mu Ci/m^3$, the pH varied from 8 to 10, and the suspended solids content also changed radically at times due to equipment problems at the full-scale process wastewater equalization tanks. The total hardness of the wastewater was typically reduced by 70% in the test system compared to 90% in the full-scale PWTP softener/clarifier. Control of the sludge blanket in the clarifier proved to be difficult, and carryover into the effluent stream was a common occurrence. To reduce the amount of suspended solids in samples, the periodic samples taken were set aside for 1 h to allow solids to settle. The sample to be submitted for analysis was decanted from the initial sample container. A second sample was also taken and filtered by one

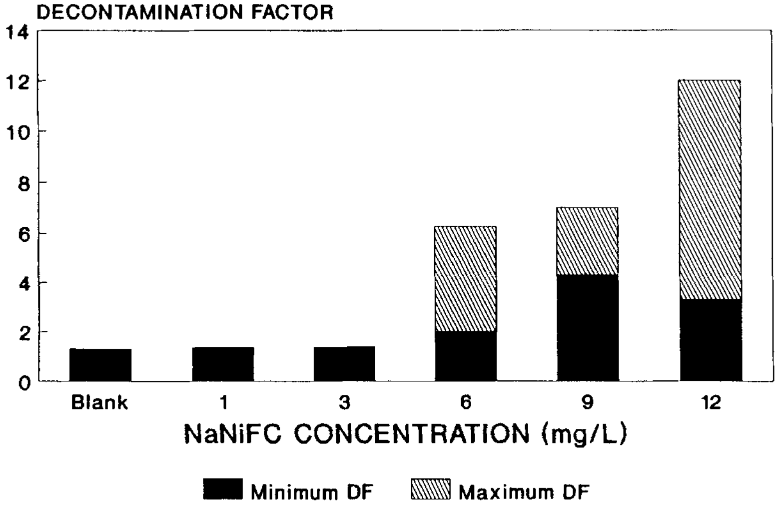

FIGURE 5. Pilot scale testing of NaNiFC for removal of cesium from process wastewater.

of several methods. The filtering methods involved the use of batch gravity filtration using a standard qualitative filter paper and also continuous filtration using the 1-in. dia flow-through column packed with granular anthracite or a combination of sand and anthracite.

Though performance was somewhat erratic, ^{137}Cs removal was reasonably effective in the tests. The test results indicated that a NaNiFC concentration of 12 mg/L was necessary to obtain decontamination factors (DFs) greater than 10. The DFs sometimes improved when filtration of samples was performed. However, the granular media filter column performance was sometimes poor, possibly due to wall effects and channeling from use of a small-diameter test column. Intermittent breakthrough of the cesium-nickel hexacyanoferrate reaction product through the filter may have attributed to the wide variation in the resultant decontamination factor. The test results are summarized in Figure 5.

To ensure that erratic in performance was not due to variation in NaNiFC batch performance, a bench scale uptake test was conducted using two different NaNiFC

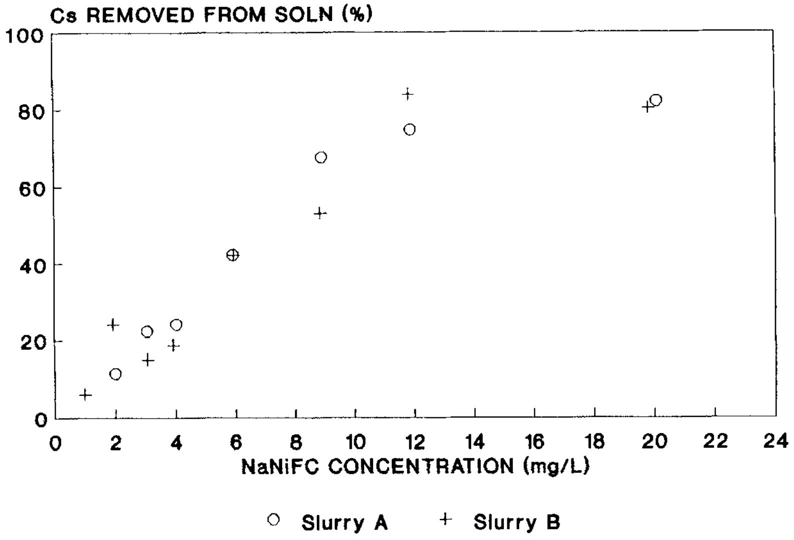

FIGURE 6. Effect of NaNiFC preparation method on cesium removal from process wastewater.

slurry batches prepared by the same researcher. As shown in Figure 6, comparable effectiveness was demonstrated by the two batches of NaNiFC. No further attempts were made to evaluate the influence of preparation method on cesium removal performance.

One of the concerns with regard to the application of NaNiFC is the possibility of dissolved nickel and cyanide compounds entering the wastewater as a result of metal/hexacyanoferrate decomposition. The effluent stream was analyzed on several occasions and submitted for nickel and total cyanide analysis. Levels of dissolved nickel were typically near the detection limit of 0.004 mg/L. Total cyanide, however, was found at levels of 1.7 to 2.1 mg/L when the feed rate of NaNiFC was 6 mg/L. The latest NPDES permit application for ORNL(10) proposes a limit of 0.65 mg/L, therefore, it is anticipated that the wastewater effluent from this process would require a subsequent treatment step to remove residual cyanide. If the cyanide compound is a simple sodium cyanide, the treatment would be a fairly simple operation requiring alkaline pH and agitation of wastewater with the addition of either sodium hypochlorite or ozone for oxidation of the cyanide to less toxic cyanate.(11) If the cyanide is in the form of a ferrocyanide complex, it is far more stable and not destroyed by alkaline chlorination.(11) Ferrocyanide complexes are not considered to be toxic to aquatic life,(11) and a special permit might be arranged with regulatory agencies for discharge of these compounds. Analytical tests were performed to determine the nature of the cyanide compounds found in the effluent stream from the test system. The results from four samples indicate that an average of 0.82 mg/L, or 44%, of the total cyanide in the wastewater effluent is amenable to oxidation. Based on these results, treatment of the effluent stream to oxidize residual cyanide must be considered if hexacyanoferrates are to be used.

Conclusions and Recommendation for Process Wastewater Tests

The results of small-scale pilot tests indicated that a level of 12 mg/L of the NaNiFC was necessary to obtain decontamination factors greater than 10, though this was not achievable on a consistent basis. Analysis of the effluent stream from the test system indicated the presence of both simple and complex cyanides. Though the complex cyanides are not an environmental concern, the simple cyanides must be treated before discharge. There is evidence from previous study(7) that the transition metal hexacyanoferrate compounds are more stable at lower pH, so operating the softener at a lower pH may reduce or eliminate cyanide in the effluent stream. The lower pH may also enhance the effectiveness of ^{137}Cs removal and reduce the hexacyanoferrate requirement. Lowering the wastewater pH will have a detrimental effect on the softening process, however, so a separate wastewater/hexacyanoferrate contactor would be required. The influence of preparation method on hexacyanoferrate effectiveness must also be investigated as well as the stability of the cesium hexacyanoferrate complex in the resultant sludges.

SUMMARY AND CONCLUSIONS

The above studies have shown that under certain conditions, hexacyanoferrates are effective for decontamination of two significantly different wastewaters generated at ORNL. In treatment of simulated LLLW concentrate, effectiveness of cesium removal was most significantly affected by pH, mixing time, and the preparation method used to make the KCFC. Decontamination factors as high as 40 were obtained, though it was determined that in situ treatment of LLLW concentrate in the MVSTs was not feasible. In tests with actual ORNL process wastewaters, pilot-scale testing indicated that NaNiFC could be effective in removal of ^{137}Cs under existing PWTP operating conditions with decontamination factors as high as 12. Residual cyanide compounds were detected in the effluent stream from process wastewater treatment, therefore, additional oxidative treatment of the effluent would be necessary before discharge to the environment.

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