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LABORATORY EVALUATION OF COLLOID TRANSPORT UNDER SIMULATED SUBSURFACE CONDITIONS AT THE WASTE ISOLATION PILOT PLANT (WIPP): 2. LARGE-SCALE-INTACT-CORE COLUMN FLOW EXPERIMENTS.

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INTRODUCTION

The Waste Isolation Pilot Plant (WIPP), located approximately 42 km southeast of Carlsbad, New Mexico, is being developed by the U.S. Department of Energy (DOE) as a disposal facility for transuranic waste produced by defense nuclear weapons programs. Regulations promulgated by the U.S. Environmental Protection Agency (1985) place limits on the cumulative radioactive release to the accessible environment over 10,000 years and require performance assessments to demonstrate WIPP compliance with regulatory standards. The only credible mechanisms for escape of radionuclides to the environment involve a breach of the repository during drilling for natural resources. The brines present in the repository, together with drilling mud and cuttings, may be transported through intrusion boreholes to a overlying transmissive carbonate rock unit, the Culebra Dolomite Member of the Rustler Formation, or directly to the surface, by pressurized fluids in the repository. Performance assessment calculations have shown that thorium, uranium, neptunium, plutonium, and americium are the most significant radionuclides in terms of total release of activity.

Previous work has been performed on soluble actinide sorption and retardation in the Culebra (Lucero et al., 1994). In those tests, uranium has undergone moderate retardation, ($2 < R < 20$) while americium and plutonium have never eluted from the column at measurable activities. Sterile conditions were not maintained in the core or injection solution and measurable bacteria numbers are always present in the column effluent. At least some of those bacteria are believed to be native to the cores themselves. Large concentrations of humic compounds are not present in the cores or brine.

The objective of this work was to quantify possible colloid-facilitated transport of actinides in large, intact Culebra cores under pressure and flow conditions similar to the natural environment. Two types of colloids were tested, a mixture of humic and fulvic acid and live microbes. Since bacteria are naturally present in the core effluent, the microbes

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tests are a measure of the effects of a microbe enrichment. These results can be compared to crushed-rock retardation studies performed by Yelton et al., (1996).

PROCEDURES

Two separate tests were performed, one using humics and the second using a enriched bacteria population (Papenguth and Behl, 1996). Intact cores were collected by horizontal drilling at a depth of approximately 220 meters inside the Air Intake Shaft at the WIPP. The microbe test was performed with Core E, a subsample of core VPX27-7A; while the humics test used core D, a subsample of core VPX25-8A. Both cores were 14.5 cm in diameter and 10 cm long, and oriented in the direction of the current groundwater flow. Several types of porosity which may affect hydraulic and transport properties were present in each, including microcrystalline, porous carbonate interbeds, vugs, fractures, and brecciated zones. The cores, which are shown is in Figure 1, were similar, with the exception that Core D showed more vugs and carbonate interbeds. The dolomite matrix initially formed with interbeds of silty dolomite and large vugs which were later connected by subvertical fractures. Bacteria are generally larger than the microcrystalline and carbonate interbed porosity. However, the vugs, fractures and brecciated zones were visible to the naked eye and could possibly allow bacteria transport. The humic compounds were small enough to fit through all pore features.

A schematic of the test apparatus used is shown in Figure 2. Its major components were column core holders, syringe pumps, brine injection accumulator and effluent collectors. The aluminum column barrel had teflon coated, brass end fittings, with the core held in a flexible sleeve between the two distributor plates. Overburden loads were simulated by applying pressure in the annulus between the column barrel and the sleeve. That pressure was also applied to the top distributor plate which was free to slide up and down, while the bottom plate was fixed. Thus, the core was subjected to the reservoir pressure in all three directions while allowing the effluent to exit at atmospheric pressure. The applied triaxial confining pressure of 725 psi was estimated assuming a rock density of 1 psi/ft above the borehole (Mercer and Snyder, 1990).

Brine was pumped from an accumulator through the core at the rate of 0.1 ml/min, which produced a Darcy flux of 10^{-5} cm/s, the upper range of Culebra flow. Double, high pressure, liquid chromatography syringe pumps drive deionized water into one end of an accumulator, which forces brine out the other. The accumulator, a simple cylinder and piston sealed at both ends, prevents contamination and fouling of the pumps, while the double pump configuration allows automatic filling of the syringe pumps and insures continuous operation. Effluent was collected with a fraction collector set to provide approximately 5 ml per sample.

The humic acid mixture used was equal parts of aliphatic humic (isolated from sediments collected from Lake Bradford, Florida), aromatic humic and fulvic acids (both isolated from the Suwannee River and purchased from the International Humic Substances Society, Golden, Colorado) added to the synthetic brine at 0.1 mg/l concentration. Bacteria used to enrich the injection were WIPP-1A; live, halophilic, rod-shaped cells. They were added to the synthetic brine at 10^6 cells/ml. That concentration was roughly equal to the native population of the synthetic brine. While no specific steps were taken to inoculate the synthetic brine, it was equilibrated with crushed Culebra samples and occasionally left

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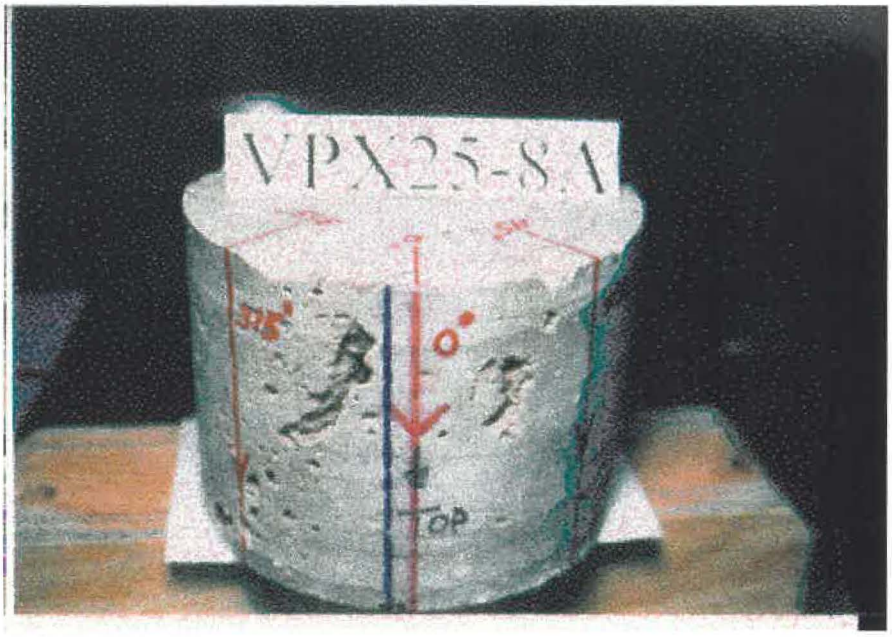
Core D - VPX25-8A



Core E - VPX27-7A



Figure 1. Intact core samples used.

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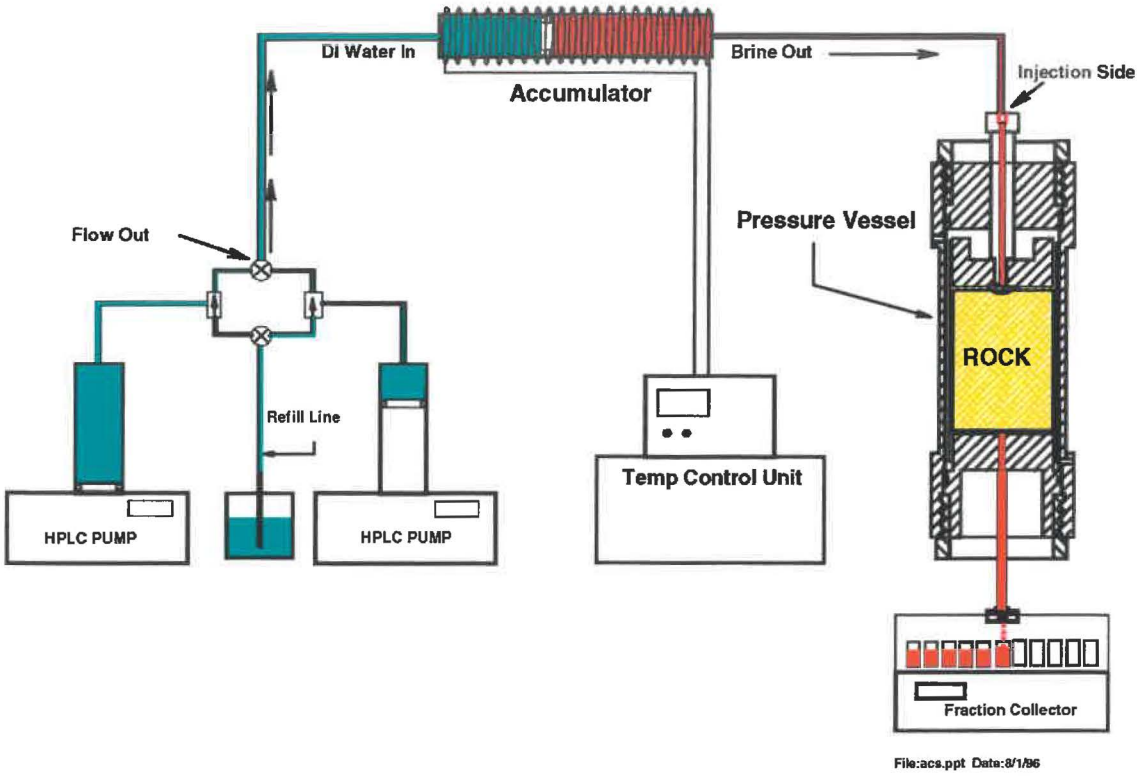

Figure 2. Core column experimental apparatus.

open to the lab air. As a result, bacteria similar to WIPP-1A can be found in the injection fluid. Both colloids were introduced as step injections to allow greater time for colloid-actinide-rock interaction and to enhance the possibility of demonstrating some effect of the colloids. A conservative tracer, ^{22}Na, was also added to the step injections to quantify column porosity and dispersion. Actinides were introduced by two methods. Uranium was introduced with the colloid step injection to maximize its contact colloid time, while americium and plutonium were injected as spikes directly on the rock to eliminate sorption on the experimental apparatus. Test conditions for both injections are listed in Table 1.

The use of live microbes required additional procedures to ensure cells population did not change during testing and analysis. The accumulator that held the brine-microbe mix was insulated and chilled to 10°C to minimize cell growth or mortality. Due to the necessity of maintaining a uniform flow, samples could not be taken from the accumulator once the test began and prevented direct monitoring of the microbes being injected. Therefore, a sample of bacteria enriched brine was placed in a mock injection vessel and maintained under similar conditions. The mock vessel was sampled at several times during the injection and the samples subjected to cell assays along with the effluent.

Most column effluent was analyzed by gamma ray spectrometry for ^{22}Na, ^{232}U, and ^{241}Am and liquid scintillation for ^{232}U and ^{241}Pu at Sandia National Labs following standard methods (Lucero et al., 1994). The radioassay tests are insensitive to colloids or actinide chemical state, and their results indicate only total sample activity. Selected effluent samples from the enriched bacteria test were fixed with formalin solution, chilled and shipped overnight to Brookhaven National Laboratory where they underwent cell assays by direct counting using epifluorescence microscopy. Bacteria length and width was measured for 15 to 53 individuals in each sample and the population volumes computed.

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Table 1. Test conditions and results.

Test	Humics	Bacteria
Colloids	0.1 gm/l	10^6 cells/ml
Core and Brine	VPX25	VPX27
Brine Composition		
Boron (gm/l)	0.026	0.027
Bromine (gm/l)	0.054	0.024
Calcium (gm/l)	0.88	0.86
Inorganic Carbon (gm/l)	0.011	0.17
Chloride (gm/l)	19	19
Magnesium (gm/l)	0.45	0.45
Potassium (gm/l)	0.32	0.32
Sodium (gm/l)	13	13
Sulfate (gm/l)	6.7	6.9
pH	7.7	8.1
Step Injection		
Volume (ml)	4,000	3,900
^{232}U (uCi/ml)	0.020	0.034
^{22}Na (uCi/ml)	0.004	0.004
Initial Actinide Spike		
^{241}Am (uCi)	4.0	13
^{241}Pu (uCi)	16	---
Fitted transport parameters		
Porosity	0.12	0.15
Dispersivity (cm2/s)	0.00079	0.00076
Uranium retardation	12.8	2.7

RESULTS

Figure 3 presents cell concentration and volume as functions of time for both the column effluent and the mock injection brine. The overall number of cells in the mock injection vessel increased after the start of the test to 7 x 10^6 cells/ml at 600ml and then decreased to 2 x 10^6. This would indicate an initial growth followed by a die-off to equilibrium levels. However, cell size in the mock injection vessel was relatively constant throughout the experiment.

Effluent cell counts were an order of magnitude lower than injection and ranged from 1.7 x 10^5 to 7.7 x 10^5. Cell numbers exhibited an erratic behavior with the largest peak at 700 ml. Early effluent, from 0 to 1,100 mls, contained large cells which may have resulted from some enriched cell breakthrough. Bacterial cell volume was greatest at 400 ml, but after 1,500 ml the effluent bacteria were indistinguishable from the normal effluent production. There were many eukaryotic organisms in the effluent, that appear to be generated in the column itself. These flagellated microbes may have eaten a small portion of the injected bacteria.

Figures 4 and 5 show the breakthrough curves for the humics and bacteria enriched tests. Each graph shows the measured effluent curves for ^{22}Na and ^{232}U and retardation

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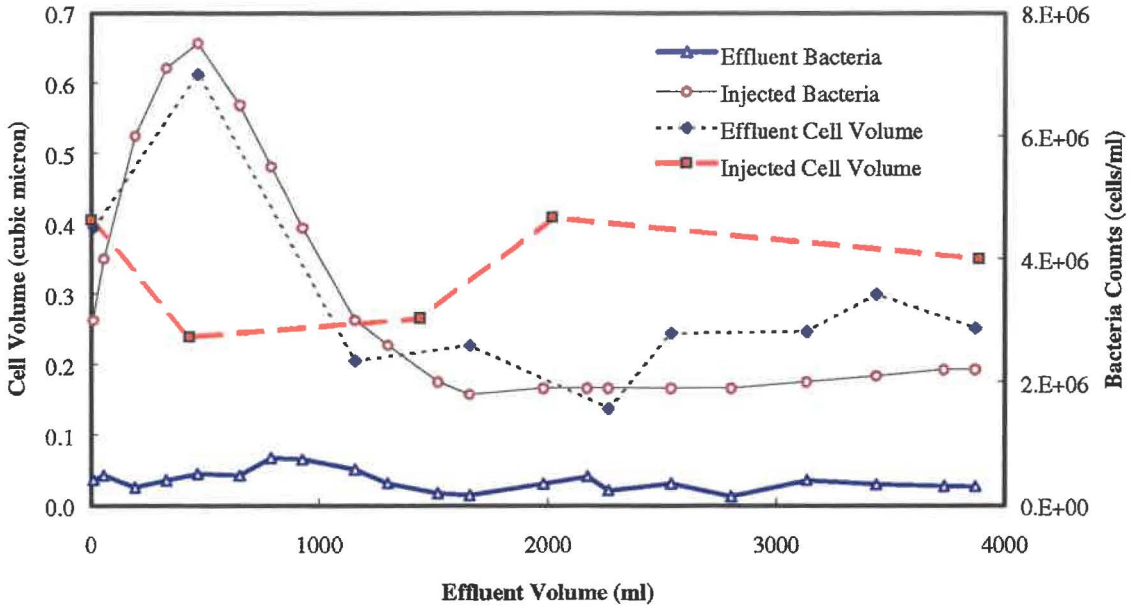

Figure 3. Live microbe cell numbers, and volume in effluent and mock injection.

fittings for the single porosity model of Parker and van Genuchten (1984). Fitted parameters are listed in Table 1. For the enriched bacteria test, the fitted uranium retardation was 2.7 compared to 2 for the same core without enrichment. For the humic test the fitted uranium retardation was 12.8 compared to 9.4 and 21 for normal conditions. For both tests the variation is within the normal range observed, indicating the colloids did not impact the uranium transport. Americium did not elute at detectable activities in either test, and plutonium did not elute in the humic test. Thus, the colloids did not effect the transport of those actinides at measurable levels.

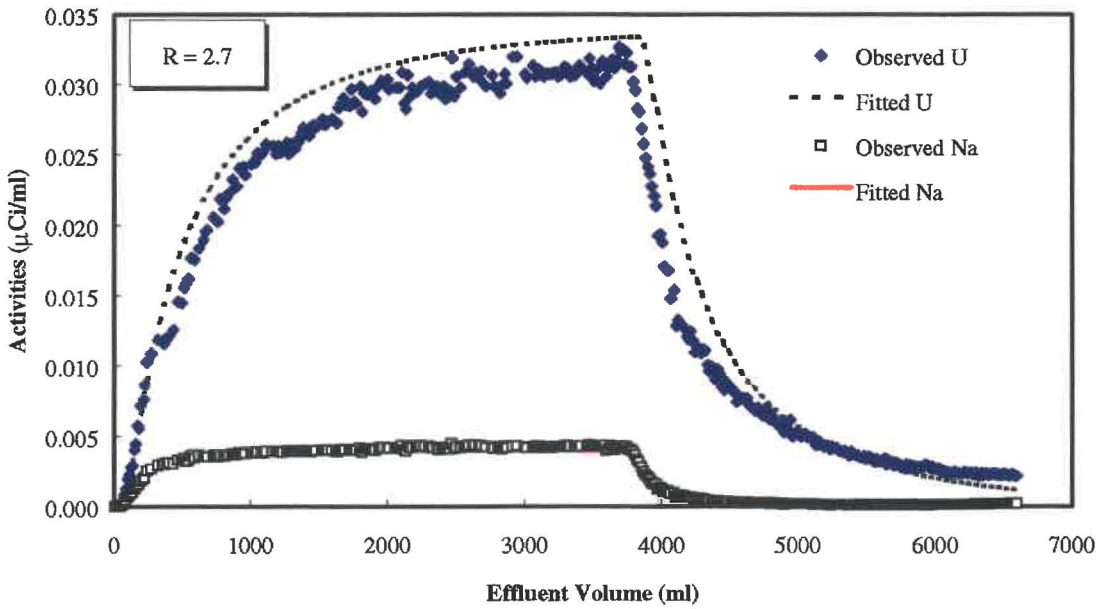

Figure 4. ^{22}Na and ^{232}U effluent activity for live microbe tests.

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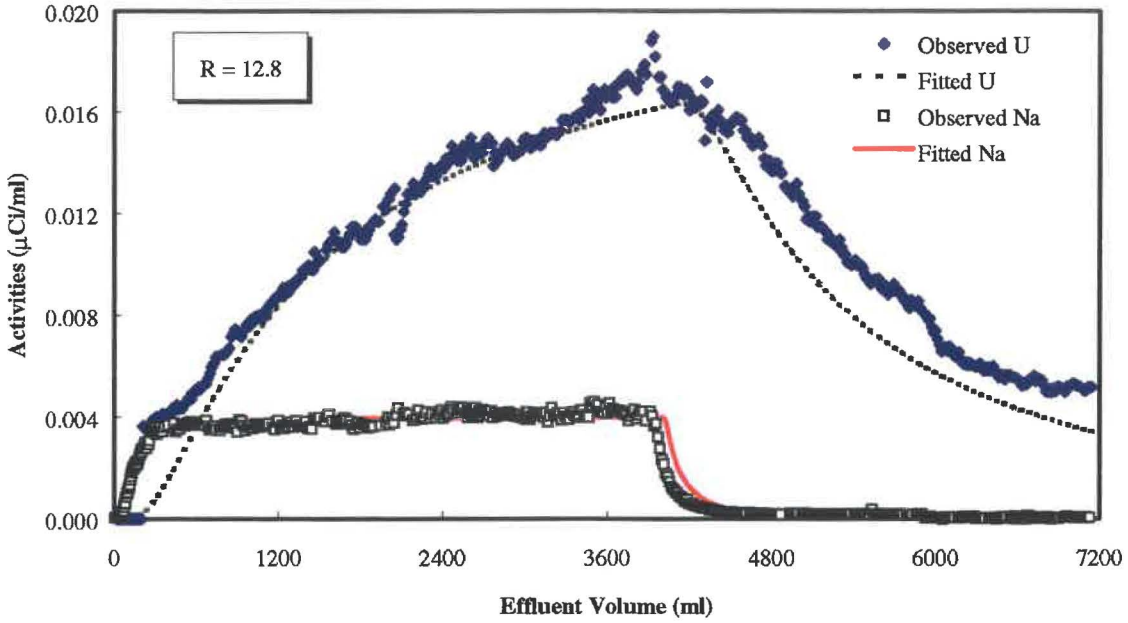

Figure 5. 22Na and 232U effluent activity for humics test.

CONCLUSIONS

The presence of an enriched microbe population had no significant impact on the retardation of uranium and americium in the intact Culebra cores. Even through the cores contained some relatively large pores, microbe cell counts were reduced an order of magnitude over the 10 cm column. This reduction is consistent with the filtration observed by Yelton et al., (1996). Likewise, the presence of humic compounds had no significant impact on the retardation of uranium, plutonium and americium in theCulebra cores.

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