

The Interaction of Groundwater and Fresh Basalt Fissure Surfaces and Its Effect on the Migration of Actinides¹

CONF-830375--1

DES3 014348

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Experiments are being performed at Argonne National Laboratory (ANL) (1) to identify interactions of radionuclides and repository components that effect nuclide migration and (2) to assess changes in nuclide migration caused by modifications expected upon aging of the waste, clay backfill, and rock. The experiments are conducted with radioactive borosilicate glass, bentonite and mechanically fissured basalt rock in flowing water analogous to their configuration in a breach of a nuclear waste repository. Changes undergone by the groundwater as it passed through the fissure include: (1) drop in pH from 10 to 8, (2) loss of suspended particulate, and (3) loss of dissolved/suspended U, Np, and Pu. These effects, also studied as functions of radiation dose and of laboratory "aging" of the repository components, are related to the predicted long-term performance of a nuclear waste repository.

This paper is a progress report of an experimental program underway at ANL that studies the interaction of flowing simulated groundwater and the components that may be used in a nuclear waste repository constructed in basalt. The components are placed in the water stream analogously to the configuration that could occur from a breach of the repository. The movement of radionuclides by the flowing water is determined by the chemical composition of the groundwater and interactions with components at various positions along the flow path.

¹Work supported by the Nuclear Regulatory Commission under Contract, FIN A-2230.

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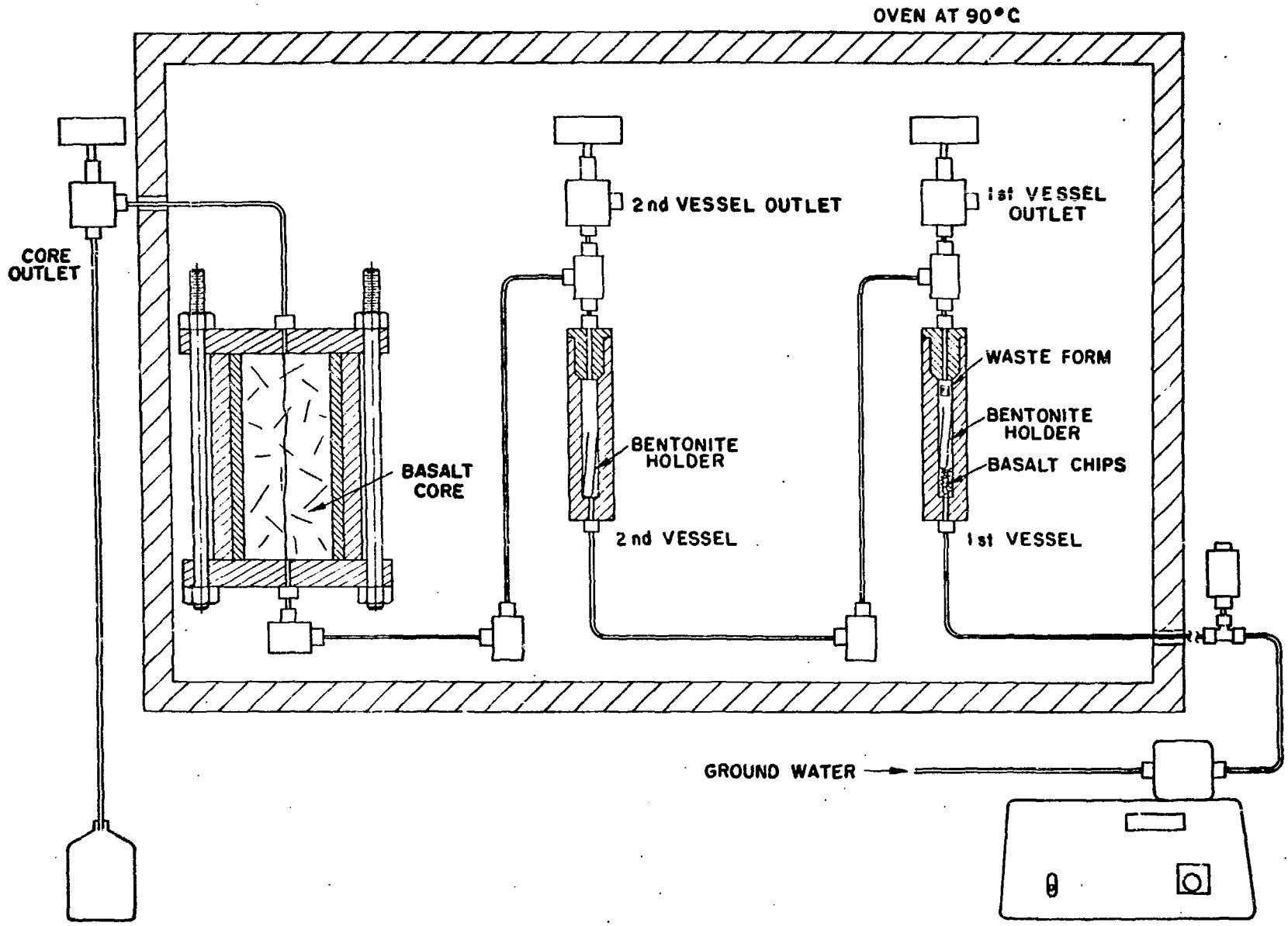
The potential for radionuclide migration by groundwater flow from a breached nuclear-waste repository depends on the leaching of radionuclides from solid waste and on the chemical reactions that occur as a radionuclide moves away from the repository. Therefore, migration involves the interactions of leached species and the groundwater components with (1) the waste and canister, (2) the engineered barrier, and (3) the geologic materials surrounding the repository. Some of these interactions would occur in the radiation and thermal fields centered on the solidified waste. Rather than trying to predict what the important interactions are and then to study them individually, we consider the combination of all potential interactions, using these experiments, termed analog experiments.

A schematic of the apparatus for an analog experiment is shown in Figure 1. In an experiment, groundwater is pumped through the system so that it passes through the first vessel, which contains basalt chips, bentonite, and the glass waste form; then through the second vessel, which contains more bentonite; and then through a narrow basalt fissure in the third vessel.

Five experiments have been initiated and a sixth experiment will begin soon; three of these experiments have been completed. Table I lists these experiments, shows how they have differed, and shows their current status. Experiments 4 and 5, labeled as "1000 yr aged" and "2000 yr aged", use radioactive solid, bentonite and basalt rock that have been hydrothermally modified in the laboratory to hydrate the surfaces in a similar manner and to a degree that would occur over extended periods of time in the repository. "Unaltered" refers to materials receiving no hydrothermal treatment before their use in the experiments.

Table I. Analog Experiments Completed or Currently Underway

Experiment Number	Condition of Waste Form Backfill and Rock	Apparatus and Experimental Condition	Status
1	Unaltered	Hastelloy, HC-276	Completed
2	Unaltered	Monel-400	Completed
3	Unaltered	Hastelloy, HC-276, Y field	Completed
4	"1000 yr aged"	Hastelloy, HC-276	Near completion
5	"2000 yr aged"	Hastelloy, HC-276	On line
6	"1000 yr aged" bentonite and rock/unaltered waste form	Hastelloy, HC-276	To begin soon



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Figure 1. Analog experimental apparatus

Experimental Materials

The simulated groundwater used in these experiments was prepared to represent the composition of groundwater from Well DC-6, Grande Ronde Formation, which samples water primarily from 200 m below the Umtanum unit (1). Its composition is shown in Table II. This groundwater composition is close to that suggested by Jones to be the reference groundwater formulation for the Basalt Waste Isolation Project (2). The water used in these experiments was continually sparged with N₂ gas before it entered the apparatus to reduce the dissolved oxygen level and to limit CO₂ pickup.

Basalt cores were cut from rocks supplied by BWIP personnel and ground on a lathe with distilled water coolant to size (6.83-cm diameter by 14.60-cm long). These rocks were Pomona-flow basalt from the Pasco basin in the vicinity of the Near Surface Test Facility.

Because these core were not permeable to water even at high pressure, they were mechanically split to provide a path for water flow. To split the core, a sharp hardened steel edge was pressed along the length of the core, using a hydraulic press, until the cores fractured (~28,500 kg). Details of the operation and the mounting of basalt cores in the core holder vessel can be found elsewhere (3).

Table II. Recipe for Simulated Groundwater
Prepared for Analog Experiments¹

Compound	Quantity Added to 20L of H ₂ O (mg)	Concentration in m Mol/L
NaCl	3512.	2.99
Na ₂ SO ₄	3194.	1.12
Na ₂ B ₄ O ₇ ·10 H ₂ O	246.	0.0331
NaF	1636.	1.95
Na ₂ CO ₃	1201.	0.57
NaHCO ₃	1841.	0.71
Na ₂ SiO ₃ ·9 H ₂ O	11,462	2.01
K ₂ SO ₄	84.9	0.032
MgSO ₄ ·7 H ₂ O ²	3.58	0.024
HCl (1M) ³	61 mL	3.05

¹ The concentration of major constituent ions in ppm are presented in Table III.

² Added from a concentrated solution--1 mL to 20 L.

³ Added to reduce the pH of the groundwater to 9.9.

The bentonite backfill material is sodium montmorillonite from Crook County, Wyoming. A 500 g sample of this material, SWy-1, was obtained from the Clay Mineral Society, Source Clay Minerals Repository, Department of Geology, University of Missouri, Columbus, MO.

The waste form was prepared by adding to 40 g of PNL 76-68 frit containing 3.4 wt % U_3O_8 : 0.65 μCi of ^{137}Cs , 100 μCi of ^{85}Sr , 45 μCi of ^{133}Ba , 1640 μCi of ^{141}Ce , 2000 μCi of ^{152}Eu , 500 μCi of ^{241}Am , 23 mg (1400 μCi) of $^{239}PuO_2$, and 115 mg (80 μCi) of $^{237}NpO_2$. This mixture was melted in a 95% Pt/5% Au crucible with a procedure detailed elsewhere (3), and cut into 24 wafers with an average weight of 0.4 g and average dimensions of 1.1-cm x 0.90-cm x 0.13-cm. These waste-form wafers were characterized in an earlier publication (4).

Materials of construction of vessels, tubing, and couplings for each experiment were either Hastelloy HC-276 or Monel-400.

Experimental Apparatus

Analog experiments are run in the apparatus shown in Figure 1 by flowing simulated groundwater through the apparatus at a rate of 0.5 mL/hr. This flow rate corresponds to a linear velocity of ~ 30 m/y past the waste form and ~ 500 m/y through the basalt fissure. The groundwater is initially at room temperature but is brought to the temperature of the apparatus, 90°C, in the length of tubing leading to the first vessel. There is an initial "equilibration" of the groundwater and the geologic components of the system before the waste form is loaded into the first vessel. As is evident from the data of the first three analog experiments in Figure 2 and an auxiliary experiment in Figure 3, an equilibration period of ~ 30 days is sufficient for the groundwater to reach a steady-state condition with the repository components. This steady state condition will be discussed at greater length in the result section.

During the first experiment it was found that to maintain flow through the basalt fissure it was necessary hold the fissure open by placing two strips of 0.14-mm thick gold ribbon along the length of the fissure. Before this was done, the pump pressure to maintain the 0.5 mL/h flow rose in several days from an initial 11 psig to one greater than 150 psig (the maximum transducer value). The core had become clogged by bentonite that had been transported by the flowing eluate from the first and second vessels. Even with the gold strips in place, the pump pressure rose to ~ 130 psig by the end of an experiment. Hydrothermally-altered bentonite was not transported by the flowing groundwater and, therefore, no increase in pressure with time has been noted for these experiments (Table I, 5 and 6).

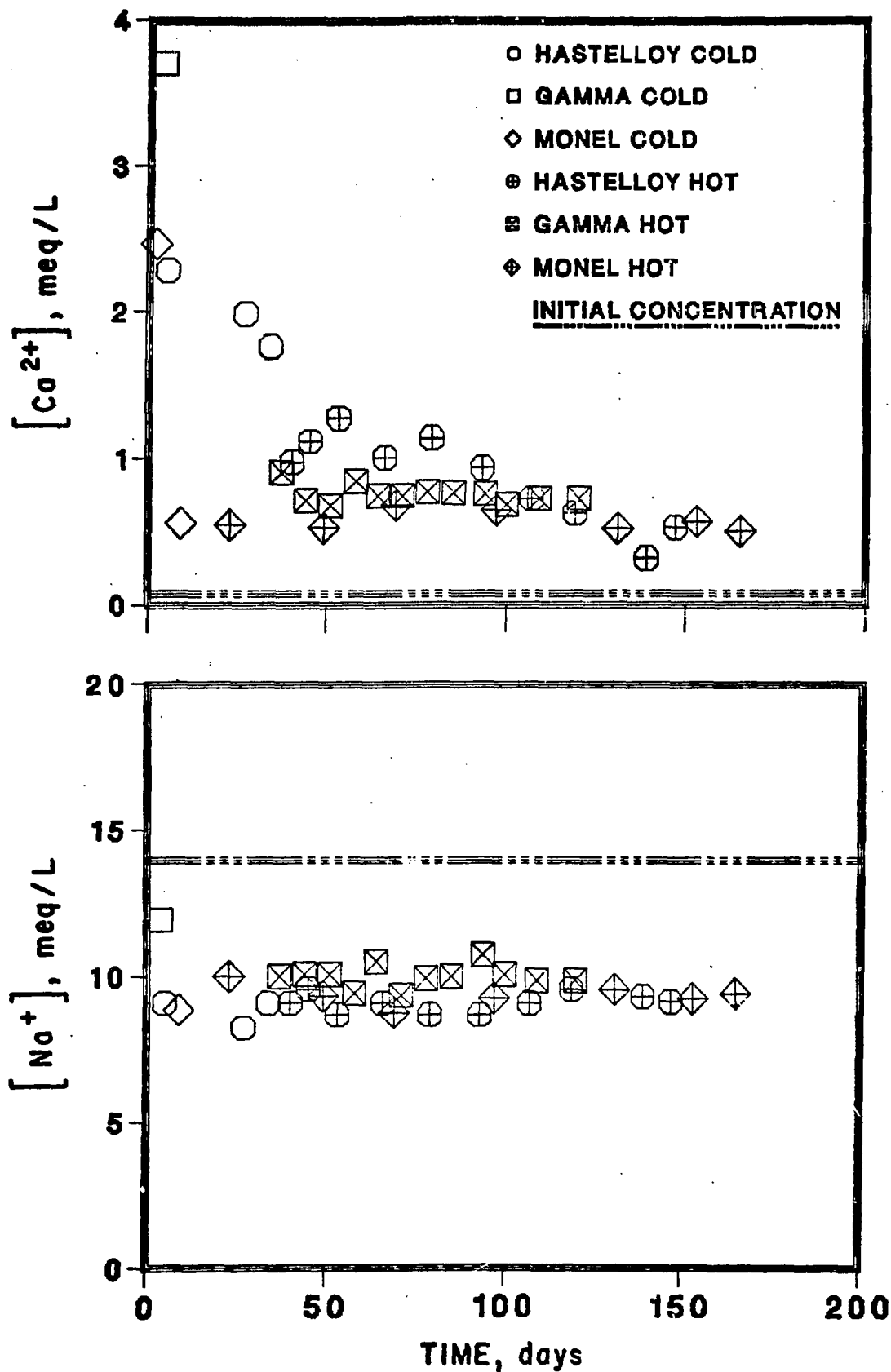


Figure 2. Attainment of steady-state concentration of sodium and calcium ions in the first three analog experiments.

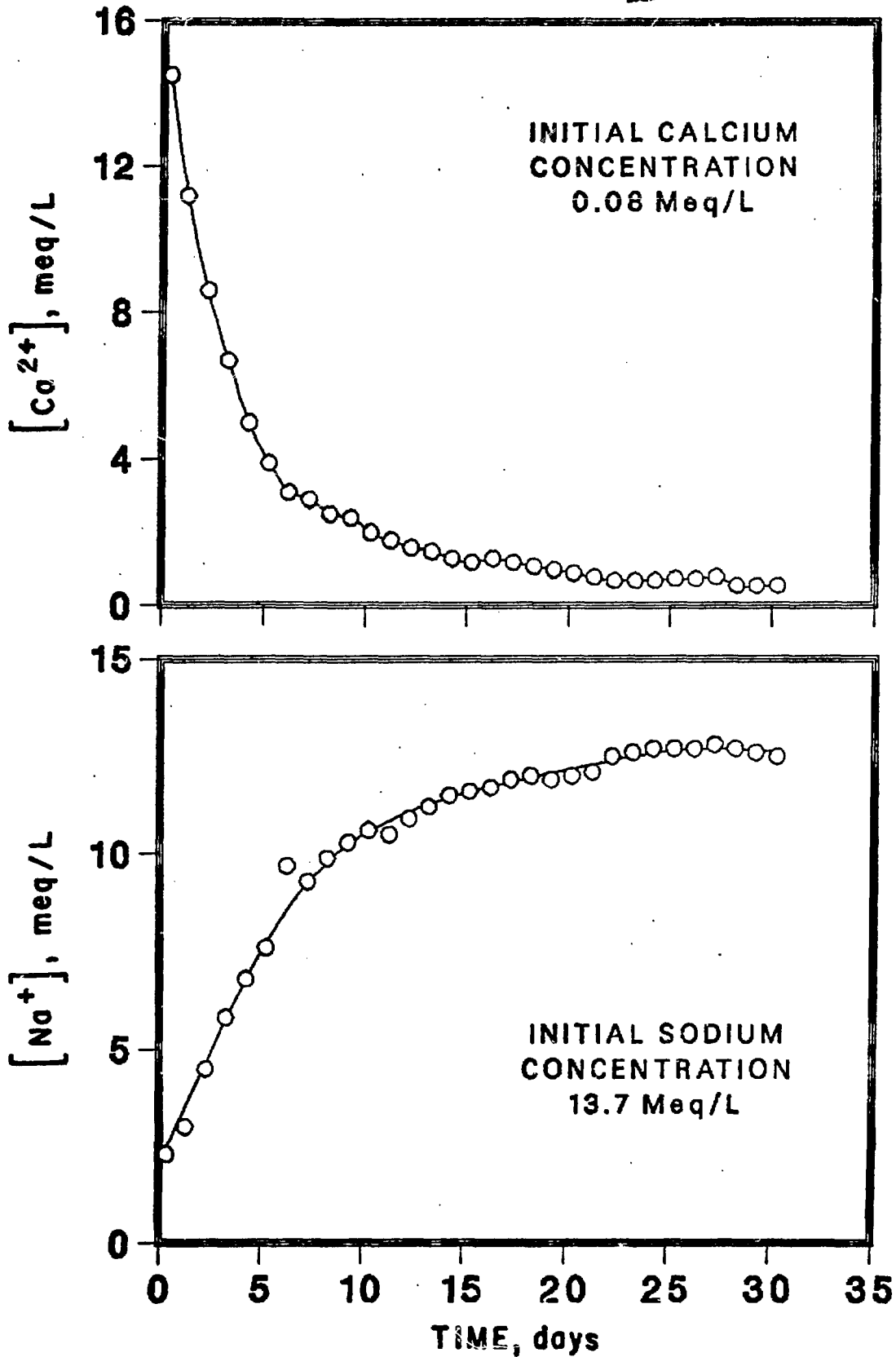


Figure 3. Attainment of steady-state concentration of sodium and calcium in auxiliary experiments.

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The special properties of bentonite clay (i.e., small particle size and swelling capability) made it impossible to localize the clay in the apparatus in a loose state. To maintain the bentonite in the system without obstructing the flowing water, a special dispersing system for bentonite was devised. This system comprised of a Hastelloy C-276 tube (2" long by 1/4" I.D.), crimped at one end, and 1 g bentonite packed into the tube to ~1-inch from the top. When the tubes were contacted with groundwater, the bentonite swelled out of the tube and bentonite particles constantly dropped into the flowing groundwater.

The third analog experiment was performed in the presence of a Cobalt-60 gamma ray source. The average dose rate to experimental components was 1×10^5 rad/h.

Experiment Shutdown

At the end of an experiment, the apparatus was disassembled from the downstream end. Flow of water was continued throughout this shutdown phase. As a part of the apparatus was removed, it was checked for bentonite and radionuclide content. Large samples (250 mL) of eluate were taken at the second vessel outlet and then the first vessel outlet as the shutdown continued.

Analytical Methods

Analog groundwater compositions were analyzed by emission spectroscopy (emission by inductively-coupled plasma, ICP) and by ion chromatography. Uranium concentrations were measured by laser fluorescent spectrometry and radiotopes by gamma- and alpha-counting analyses. Because of the high dissolved solid content (832 mg/L) of the groundwater, it was necessary to perform a chemical separation of ^{239}Pu and ^{237}Np by hexone extraction. These procedures are described elsewhere (5).

Hydro-, Thermo-alteration of Components

Basalt and bentonite were altered by placing them in an autoclave under simulated groundwater at 320°C for 30 and 60 days to simulate 1000 y and 2000 y aging, respectively. The waste-form wafers for both experiments were aged in the same manner by treating them for 17 days in saturated steam at 340°C. The details of these procedures and the rationale for their use have been published previously (6).

Results

As the simulated groundwater passes through the three vessels of the analog experiment (Figure 1), it undergoes compositional changes which appear to affect its ability to transport

actinide ions through the system. The following two sections describe separately (1) the changes in groundwater composition and (2) the actinide migration in the rock core fissure. Following these results is the discussion section, which briefly attempts to tie these two phenomena together.

Changes in Analog Groundwater Composition. The data in Figure 2 for the three analog experiments (which used unaltered waste form, bentonite, and basalt cores) show that the groundwater exiting the basalt fissure reaches a steady-state composition for $[Na^+]$ and $[Ca^{2+}]$ soon after the start of the experiment. (There are three symbols for the three separate experiments, experiments 1, 2, and 3 in Table I, that are either empty or with an X inside them. The term "cold" in the figure legend refers to the equilibration period of the experiments, before the waste form was put into vessel 1. "Hot" refers to that part of the experiment where the waste form was in place.) The steady-state concentration of sodium ion is substantially lower at the core's exit than that of the original groundwater solution. The calcium-ion concentration is much higher at the steady-state condition reached inside the core than at its original concentration.

An auxiliary column experiment, performed with groundwater and crushed basalt chips and with a basalt surface area to groundwater volume ratio equivalent to that of the analog basalt fissure ($2 \times 10^2 \text{ cm}^{-1}$), has elucidated this phenomenon. Data from this experiment, which are presented in Figure 3, show that the sodium-ion concentration of the groundwater does not decrease gradually from its initial value of 13.7 meq/L to its steady-state value of ~ 9.6 meq/L, but, rather, it initially falls to a much lower concentration, ~ 2 meq/L. Gradually, over an "equilibration" period, the sodium-ion concentration rises to its steady-state value. The calcium-ion concentration shows an opposite, yet similar, behavior. Calcium ion's initial groundwater concentration of 0.08 meq/L increases dramatically to 15 meq/L before gradually dropping to a steady-state concentration of 0.7 meq/L.

Table III shows ICP, ion chromatography, and uranium concentration data for outlet groundwaters from the first analog experiment. In general, alkaline-earth elements show the same behavior as does calcium; an initial very high concentration gradually falls to a steady-state concentration much higher than that of the initial groundwater. Some preliminary data for potassium ion show that it also follows the trend of calcium ion. The variation in anion concentrations is small in comparison to that of cationic species.

Table III. Major Constituent Concentrations of Groundwater
Samples which Exited the First Analog Experiment

Sampling Period	Collection Time, days	Chemical Constituent ¹									
		B ²	Ca ²	Mg ²	Na ²	Si ²	Sr ³	F ⁻²	Cl ⁻²	SO ₄ ²⁻⁻²	U ³
	Original Groundwater	1.30	1.5	<0.02	316	53.0	<2	35	165	110	0.04
Conditioning	1-10	1.37	45.9	14.4	210	63.5	220	15	175	140	0.16
Conditioning	24-31	1.43	39.9	12.3	190	51.2	170	20	165	125	0.12
Conditioning	31-38	1.42	35.6	7.2	210	49.3	130	25	165	125	0.13
Radioactive	1-3	1.50	19.7	5.0	210	47.4	110	ND	ND	ND	ND
Radioactive	6-9	1.58	22.4	3.1	220	51.7	100	ND	ND	ND	ND
Radioactive	13-17	1.53	25.7	6.5	200	55.7	120	20	165	115	2.7
Radioactive	27-29	1.46	20.3	2.8	210	47.8	90	ND	ND	ND	ND
Radioactive	37-45	1.51	22.8	5.6	200	51.2	110	ND	ND	ND	ND
Radioactive	52-59	1.46	18.8	5.4	200	54.8	100	25	165	110	6.1
Radioactive	66-73	1.46	14.7	4.7	210	49.2	80	ND	ND	ND	ND
Radioactive	78-85	1.38	12.7	2.4	220	49.6	60	30	160	105	6.7
Radioactive	98-105	1.38	6.4	0.5	215	50.9	44	30	160	115	ND
Radioactive	105-114	1.38	10.6	0.7	211	54.0	48	ND	ND	ND	ND
Radioactive	114-120	ND	ND	ND	ND	ND	ND	30	165	115	ND

¹ ND - no data collected.

² Reported in mg/L.

³ Reported in µg/L.

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Table IV shows the major cationic and anionic constituents in the seconds analog experiment groundwater in (1) its initial condition, (2) its condition at steady-state on exiting the second vessel, and (3) its condition at steady-state on exiting the unaltered basalt fissure. Quite apparent from these data is the small alteration in groundwater composition caused by interaction of the groundwater with the bentonite and waste form, when compared to that of the effect of groundwater interaction with unaltered basalt. The major difference in groundwater after it passed through the two bentonite-containing vessels was a slightly lower pH and sodium-ion concentration. (Changes in H_3SiO_4^- , H_2BO_3^- , and CO_3^{2-} are all attributable to lower pH values.) Passage of the groundwater through the basalt fissure greatly modified its cationic content and its pH. Again, there was no perceptible change in the total SO_4^{2-} , F^- , Cl^- , silicate, or borate concentration. The charge balance is accomplished by the decrease in pH (10 to 8) that occurs in the rock fissure. This decrease in pH decreased the negative ions in solution by neutralizing much of the H_3SiO_4^- , H_2BO_3^- , and CO_3^{2-} in the groundwater. The total (carbonate + bicarbonate) also decreased by ~40% from passage through the rock core.

Thus far we have discussed only the interaction of analog groundwater with unaltered bentonite and basalt. Table V, which contains data from the 1000 y analog experiment, shows the quite different, and quite limited, effect on groundwater composition of contact by the hydro-, thermo-altered basalt fissure. A comparison of the first and third data rows of Table V (original analog groundwater *vs.* that exiting the rock core ~90 days into the experiment) shows only limited compositional changes in the two solutions. The steady-state compositions of major groundwater constituents in the exiting groundwater are far closer to those of the original groundwater than those noted for fresh basalt experiments. The major differences that did occur as the groundwater passed through the fourth analog experiment were substantial increases in concentrations of calcium ion, potassium ion, and total (carbonate plus bicarbonate). The decreases seen for H_3SiO_4^- and H_2BO_3^- are due to their partial neutralization at the lower pH of the exiting groundwater.

The data for the second vessel outlet in Table V seem to show that groundwater composition is more affected by aged bentonite than by its unaltered form. The sodium-ion concentration dropped dramatically as calcium ion and potassium ion both rose. Anionic species concentrations also dropped. These data, however, were collected early in the experiment and are likely to be not representative of the steady-state condition of the system. The steady-state composition of the second vessel outlet solution will be measured soon, when the rock core is taken off-line at the end of the experiment.

Table IV. Major Constituents of the Second Analog Experiment
Groundwater Solution in meq/L (Unaltered Basalt)

Solution	pH	Na ⁺	Ca ²⁺	Mg ²⁺	Σ Cations
Initial Groundwater	9.9	13.7	0.075	nil	13.8
2nd Vessel Outlet ~150 Days	9.6	12.3	0.018	nil	12.3
Core Outlet ~100 Days	7.8	9.6	0.63	0.20	10.4

Cl ⁻	F ⁻	SO ₄ ²⁻	OH ⁻	H ₃ SiO ₄ ⁻	H ₂ BO ₃ ⁻	CO ₃ ²⁻	HCO ₃ ⁻	Σ Anions
4.65	1.84	2.29	0.08	1.10	0.10	0.96	1.33	12.4
4.84	1.82	2.43	0.04	0.67	0.08	0.82	1.05	11.8
4.51	1.58	2.19	nil	0.02	0.01	nil	1.33	9.6

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Table V. Major Constituents of the Fourth Analog Experiment
Groundwater Solutions in meq/L (Altered Basalt)

Solution	pH	Na ⁺	Ca ²⁺	K ⁺	Σ Cations
Groundwater	9.9	13.7	0.060	0.05	13.8
2nd Vessel Outlet ~10 Days	9.5	9.5	1.9	0.3	11.7
Core Outlet ~90 Days	9.5	13.6	0.61	1.4	15.6

Cl ⁻	F ⁻	SO ₄ ²⁻	OH ⁻	H ₃ SiO ₄ ⁻	H ₂ BO ₃ ⁻	CO ₃ ²⁻	HCO ₃ ⁻	Σ Anions
6.0	1.9	2.4	0.10	1.56	0.11	0.86	0.82	13.8
4.1	1.4	1.9	0.04	0.79	0.07	0.50	2.03	10.8
5.8	1.9	2.7	0.04	0.80	0.08	0.52	2.00	13.9

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The fact that the sums of anionic charges are less than the cationic charges in the data presented in Tables IV and V is of some concern. The differences, however, are within the experimental error limits of the collective analytical methods (ICP, ion chromatography, and acid/base titration and pH measurement) used to measure the concentrations of these species. This phenomenon will be studied further.

Actinide Migration in the Rock Core

Table VI shows the concentrations of plutonium, neptunium, and uranium measured at the inlet and outlet of the unaltered and altered basalt core fissures in the first five analog experiments (see Table I). Under conditions simulating a repository that was unaltered by earlier groundwater breach, both Np and Pu, in the concentrations developed in these analog experiments from the leaching of the waste form, were substantially retarded by the 14.6-cm basalt fissure. In fact, as can be seen from Figure 4, almost all of ²³⁷Np activity was sorbed on the first one-third of the rock fissure. Uranium retardation was determined to be not as complete.

Table VI. Sorption of Actinides by Basalt Fissures

Experiment	Pu ($\frac{\text{dpm}}{\text{mL}}$)	Np ($\frac{\text{dpm}}{\text{mL}}$)	U ($\frac{\text{ng}}{\text{mL}}$)
1 inlet	0.4	5.7	35.6
1 outlet	≤ 0.006	0.03	9.6
2 inlet	0.1	5.2	27.2
2 outlet	≤ 0.007	0.07	11.1
3 inlet	5.3	0.8	7.5
3 outlet	≤ 0.001	≤ 0.001	0.6
4 ¹ inlet	29.3	40.2	357
4 ¹ outlet	13.5	24.7	396
5 inlet	42.2	45.3	-
5 outlet	34.4	43.6	-

¹ U, and perhaps Pu and Np, are not at steady-state concentrations in the core inlet or outlet streams. The apparent increase of U as it passed through the core is due to the differing sampling times for the two streams.

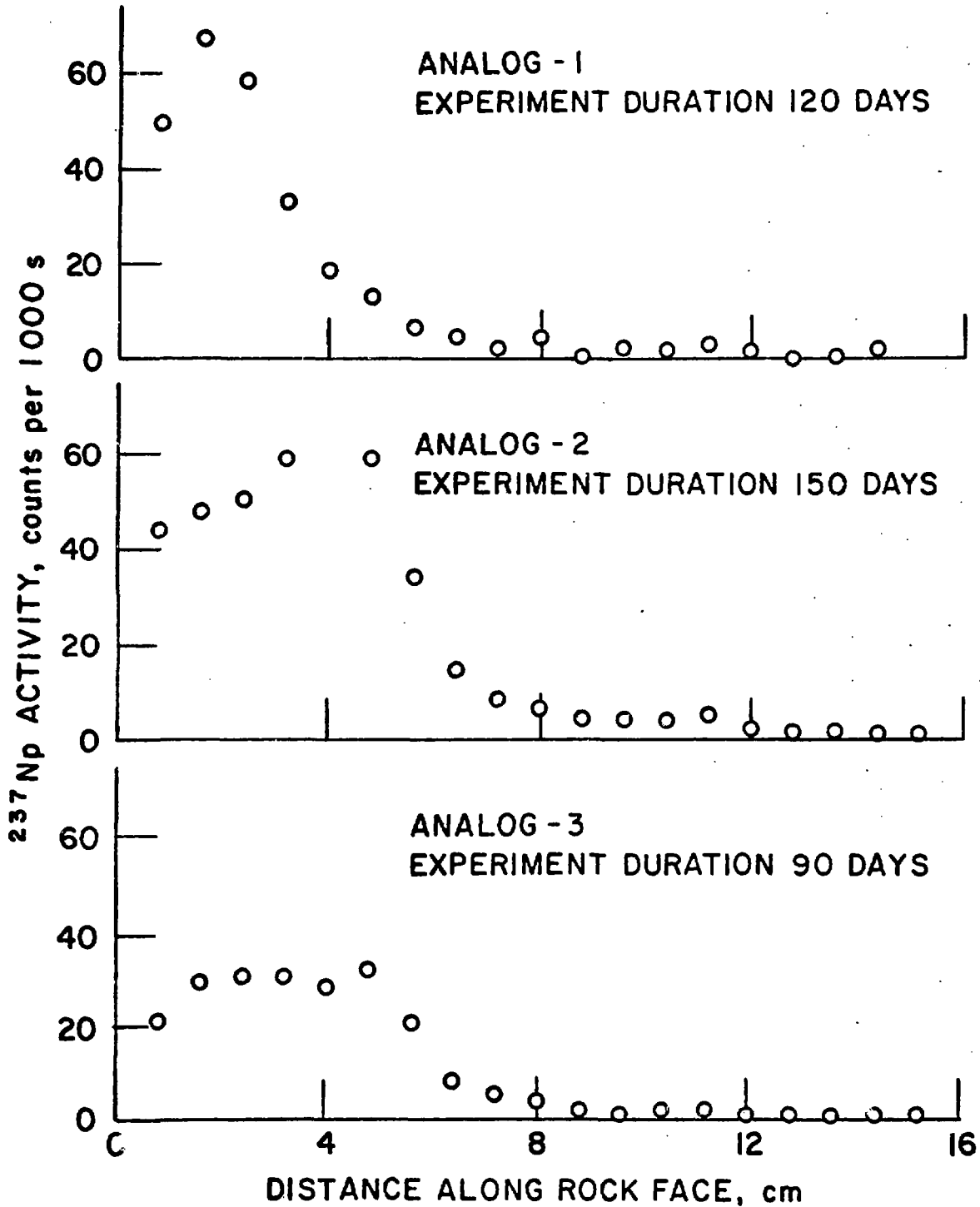


Figure 4. ^{237}Np sorption on unaltered basalt core fissures.

By filtering groundwater samples from the second vessel outlet through 0.1 μm filters, it was determined that the neptunium and uranium were both in soluble forms, and that most of the plutonium was associated with filterable bentonite particles suspended in the groundwater.

The oxidation states of the actinides are not known, but are controlled by their initial state in the simulated waste glass and by the mechanism of their leaching in the groundwater environment. Their oxidation states in the basalt fissure presumably would be controlled by the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox couple and the ferrous ion on the rock surface; at the pH range of 10-8, this couple would reduce higher oxidation states to Np(IV), Pu(IV), and U(IV) (7-10).

The differences in actinide element concentrations in the second vessel outlet samples for experiments with and without the presence of a gamma field (experiment 3 vs. experiments 1 and 2) is clearly evident from the data of Table VI. These differences are probably related to differing leaching characteristics of the waste with and without gamma radiation. Such effects have been reported by others (11, 12). We are planning future experiments to verify and to further study this result.

The actinide retardation data for the hydro-, thermo-altered repository component experiments (4 and 5) in Table VI show that only a small fraction of the actinides are retained by the rock core. Comparing these results to those of the unaltered fissure experiments, where Pu and Np were almost completely retarded by the rock core, one could conclude that altering the rock tends to pacify it. Another way to discuss these same data, and one that leads to a completely opposite conclusion, is in terms of the amount of activity retained by the rock core, or, the rate of actinide loss from the groundwater in terms of $\Delta \text{dpm}/\Delta \text{ml}$ of ^{239}Pu and ^{237}Np . These quantities are both higher for experiment 4 than for the three experiments with unaltered repository components. Experiment 6, yet to begin, should clarify this enigma. What is clear from comparing the data in Table VI is that actinide behavior in altered and unaltered repository situations will be quite different.

Discussion

The laboratory analog approach to identifying and measuring repository interactions has been demonstrated by a series of experiments. A strong correlation has been demonstrated between changes in groundwater composition and radionuclide migration. Conclusions that have been reached at this point in this continuing program are:

- Fresh, unaltered basalt fissures and surfaces will not be in equilibrium with groundwater that enters the nuclear waste repository area.

- A steady-state groundwater composition will be achieved in a fairly short time period (20-25 days for the laboratory analog experiments).

- While bentonite is transported by flowing, unaltered groundwater, interaction with unaltered basalt surfaces appears to agglomerate and to bind it.

- Unaltered basalt appears to effectively limit the migration of actinides dissolved or suspended in the groundwater.

- There was no perceptible difference in groundwater/basalt interactions or actinide migration due to laboratory analog apparatus construction materials (Hastelloy vs. Monel).

- Gamma radiation does not substantially effect groundwater compositional changes due to interactions with unaltered bentonite and basalt but appears to generate identifiable differences in actinide migration (e.g., uranium migration).

- Gamma radiation perceptibly modifies leaching characteristics of the waste form.

At this time, it is not possible to identify the mechanisms that are responsible for actinide retardation in unaltered basalt fissures. There are four possible mechanisms that may be responsible for the fissure actions:

1. Decomposition of Pu, Np, and U carbonate complexes due to drop in pH and CO_3^{2-} concentration.

2. Reduction of higher oxidation states of U, Np, and Pu by ferrous ion in basalt.

3. Changing of the surface charge on bentonite due to changing groundwater composition thus effecting its adsorption and agglomeration properties.

4. Coprecipitation of actinides with divalent-cation carbonates and sulfates.

Auxiliary experiments are underway and are being planned to elucidate the mechanisms of actinide retardation in altered and unaltered repository conditions.

Future work in this program will emphasize the modeling of laboratory analog experiments with computer codes designed to predict repository behavior. This relatively simple system is a good test for these computer codes' predictive reliability. Also, laboratory experiments will be run with rock cores containing specific naturally-aged fissures. This will be an important step in testing the appropriateness of laboratory aging of rock surfaces to simulate natural aging processes.

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