



PERMEABILITY AND PORE-FLUID CHEMISTRY OF THE BULLFROG TUFF IN A TEMPERATURE GRADIENT

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ABSTRACT

The permeability and fluid chemistry of the Bullfrog Tuff from the Nevada Test Site was studied under simulated nuclear waste repository conditions to determine the ease with which radio-nuclides could be carried in the environment. In our experimental system, pore fluids flowed radially through a cylindrical sample of the tuff from a high temperature outer edge of the rock, which ranged between 48° and 73°C. Confining and pore pressures simulated a depth of burial of approximately 1.2 km.

Permeabilities of the initial room temperature rock ranged from .4 to .85 microdarcies. Upon heating, these values increased several microdarcies due to thermal cracking. Exposure to the hot fluids over time reduced the permeability of the 250°C cylinders by 25-50% of the initial heated values, regardless of the chemistry of the pore fluids. The cylinder at 150°C showed no systematic change in permeability with time.

Chemical analysis of the discharged pore fluids indicate that the growth of minerals such as zeolites or smectite clays within cracks and pores of the tuff may have led to the observed permeability reductions. These reductions are less than those occurring in similar, previous studies of granites and quartzite, which showed permeability decreases of up to two orders of magnitude. The granite and quartzite samples had lower initial permeabilities, lower porosities and fluid flow predominantly through thin grain boundary cracks which gave rise to rapid crack clogging.

Although the Bullfrog Tuff does not show the dramatic permeability changes typical of other rock types, this study demonstrates that fluid flow can be slowed by up to 50% within a few weeks of exposure to higher temperatures.

INTRODUCTION

Investigations of the geochemical and physical changes of rocks reacting within a temperature gradient have applications in a number of fields. For example, hot rock/water interactions are important to studies of geothermal energy reservoirs (Charles and Bayhurst, 1983; Ellis and Mahon, 1964). Buried radioactive wastes will also produce a temperature gradient within the host rocks

that may cause mineral reactions to take place. Moore et al. (1983) and Morrow et al. (1981) found that significant permeability reductions occurred in heated granites under simulated waste repository conditions as a result of mineral growth within cracks and pores of the rocks. In this paper, we have studied the permeability and fluid chemistry of the Bullfrog Member of the Crater Flat Tuff, a proposed nuclear waste host rock from the Nevada Test Site, at elevated temperatures

and pressures. The purpose of this project is to investigate the changes that take place with time when groundwater comes in contact with heated rock, and to determine the ease with which potential radio-nuclide bearing groundwater could be carried into the environment.

SAMPLE DESCRIPTION

Samples of the Bullfrog Member of the Crater Flat Tuff were collected at the southwest end of Yucca Mountain, just outside the boundaries of the Nevada Test Site from the sides of an old stream cut. The Bullfrog at this location has four distinct layers. Going up-section they are: an air fall, a vitrophyre, a vapor-phase altered ash flow, and an unwelded ash flow. The samples were collected exclusively from the vapor-phase altered ash flow. They are devitrified, non-zeolitized and moderately welded. Deformed elongated pumice fragments are visible in the matrix, as well as large elongated voids.

PROCEDURE

The experimental technique is described briefly below; additional details are presented in Morrow et al. (1981) and Moore et al. (1983). The experimental assembly is shown in Figure 1. A cylindrical sample of the Bullfrog Tuff, 7.62 cm in diameter and 8.89 cm long, with a 1.27 cm diameter borehole, was used in the experiment. The borehole contained a coiled resistance heater to produce a temperature gradient between the center and outside of the rock. Water flowed radially through the tuff from the high temperature borehole to the lower temperature outer edge in response to a small differential pore pressure gradient between the center and outside of the rock. Cores were drilled such that flow would be along the bedding direction. Exposed metal in the high temperature area, such as the heating coil, was gold plated to reduce contamination of the fluids. Gold shims at the ends of the sample prevented water from leaking between the tuff and the impermeable fused silica cylinders that served as thermal insulators.

A thin, stainless steel mesh was wrapped around the outside of the sample to allow drainage of the pore fluids that have passed through the rock. In this way, the jacket for the sample assembly did not interfere with fluid flow, while still sealing the sample from the confining fluid. Pore pressure and hydrostatic confining pressure were held constant by a computer-controlled servo-mechanism. Samples of the pore fluids were

taken at intervals during the experiments to determine changes in water chemistry as a result of interaction with the heated tuffs. Fluid samples also were collected during initial, room-temperature permeability measurements.

Three experiments were conducted using the Bullfrog Tuff; the conditions are summarized in Table 1. Experiments 1 and 2 both had a borehole temperature of 250°C. Deionized water was used as pore fluid in the first experiment whereas a natural groundwater (J-13) from the Nevada Test Site was used in the second. The third experiment was conducted at a 150°C borehole temperature, again using the J-13 water. The composition of the J-13 groundwater was analyzed with the other pore fluids and is presented in Table 2. An attempt to maintain a pore pressure differential of 2.5 bars was made in all experiments. This caused an unreasonably high flow rate in experiment 2 and therefore a 1.4 bar differential was used in this experiment.

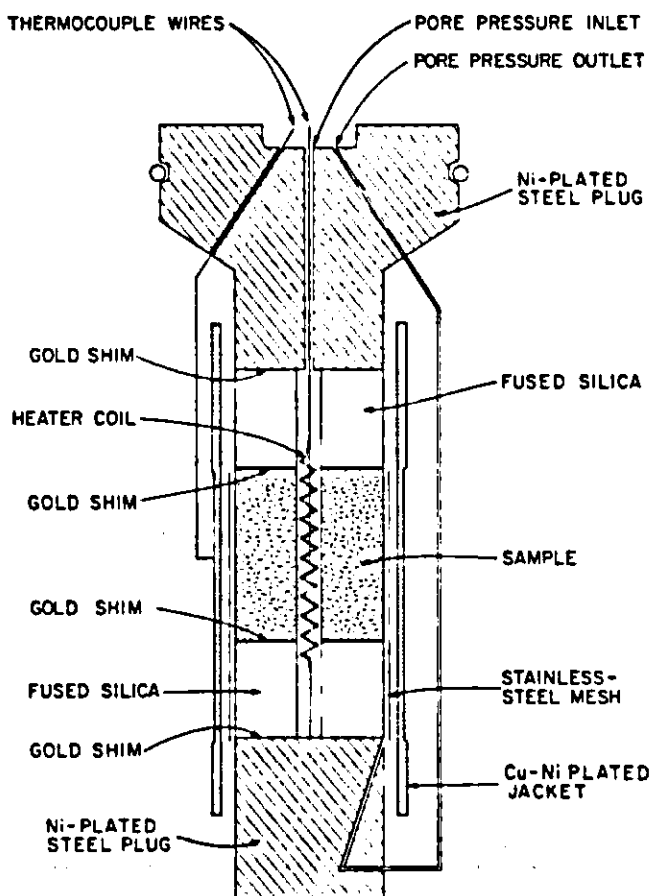


Figure 1. Schematic sample assembly.

TABLE 1
Experimental Conditions

Experiment #	1	2	3
Confining Pressure, bars	300	300	300
Pore Pressure, bars	100	100	100
Differential Pore Pressure, bars	2.4	1.4	2.5
Borehole Temp. °C	250	250	150
Jacket Temp. °C	68	73	48
Pore Fluid	Deion. water	J-13 water	J-13 water
Avg. Daily Flow Rate	65ml	100ml	14.5ml

PERMEABILITY

The variation in permeability with time was determined from measured changes in the mass flow rate over the constant pore pressure differential, using the radial flow form of Darcy's Law:

$$\frac{Q_m}{2\pi l} = -\frac{kr}{v} \frac{dP}{dr}$$

where Q_m is the mass flow rate, l the length of the sample, k permeability, r radius, and dP/dr the radial pore pressure gradient. v is the dynamic viscosity of water, which is a function of temperature and therefore varies with radius in these experiments.

Figure 2 is a plot of the changes in permeability of the tuff cylinders with time. The initial, room temperature permeabilities of the three samples varied between 0.5 and 8.5 μ da. The higher starting permeabilities of the tuffs in experiments 1 and 2 may be related to the presence of major fractures in both cylinders; the lower-permeability cylinder of experiment 3 showed no visible fractures. A time-stationary temperature gradient was established across the tuff cylinders approximately an hour after initial heating. Some transient permeability increases accompanied the temperature increase, probably related to thermal expansion in the tuffs. The cylinders in the 250°C experiments showed marked permeability

increases followed by equally rapid decreases in the first 6 hours of the heated experiment. Thereafter, the permeability of cylinder 2 continued to decrease slowly to approximately 70% of the pre-heated value. However, approximately 8 days elapsed before the permeability of cylinder 1 had completely returned to its initial value, and no further changes were measured. In the third experiment, at 150°C borehole temperature, only a slight permeability increase was observed upon heating, but the permeability remained at that higher value throughout the experiment.

FLUID CHEMISTRY

Sample Collection and Analytical Techniques

A 1.5 ml sample of the fluids discharged at the low-temperature, outer edges of the tuff cylinders was collected at intervals during the experiments. Immediately upon collection, each sample was passed through a 0.45 μ m filter and a 0.25 ml sample was separated for SiO₂ analysis. Room-temperature pH and conductivity measurements were immediately made on the remaining fluid.

Selected samples were analyzed for up to 12 dissolved species. The fluids reserved for silica analysis were digested overnight in a 1N NaOH solution and then analyzed by standard spectrophotometric techniques, using the molybdate blue method (ASTM, 1974, pp. 401-2). From the larger fluid sample, the cations Na, Ca, K, and Mg were determined using atomic absorption spectrophotometry, and the total dissolved inorganic carbon content was determined with a carbon analyzer. The anions Cl, F, SO₄, NO₃ and, where present, NO₂ and PO₄, were determined using ion chromatography. Selected results of these analyses are listed in Table 2. A more detailed list of the chemical analyses is included in Byerlee et al. (1983). The variations in pH and Na and SiO₂ concentrations with time during the heated experiments are plotted in Figures 3-5, using all available fluid analyses.

In near-neutral solutions, dissolved silica is essentially not ionized, and inorganic carbon in solution exists principally as bicarbonate ion. However, the solutions discharged from the tuffs have relatively high pH values, which lead to ionization of a large proportion of the silica content and formation of carbonate at the expense of bicarbonate ion. In order to estimate the extent of such ionization reactions, the fluid compositions were analyzed with the SOLMNEQ computer program (Kharaka and Barnes, 1973; modified by Mariner, personal communication, 1982), for temperatures of 25°C. These determinations are listed in Table 3.

TABLE 2
Chemical Analyses (mg/l) of J-13 Groundwater and Collected Fluids

Days	(@25°C) pH	+	+	++	++	-	-	-	-	-	-	-	Total C
		Na	K	Ca	Mg	SO ₄	NO ₂	NO ₃	F	Cl	PO ₄	SiO ₂	
J-13 Groundwater													
--	7.69	44	4.5	12	1.9	17	--	8.1	2.2	8.1	--	61	--
Experiment 1 Room Temperature													
0	9.49	480	12	7.5	0.9	200	--	390	6.9	280	--	22	44
2.0	10.18	860	16	7.6	0.8	300	--	530	3.4	390	Tr	NA	100
3.0	10.33	460	9	1.6	0.2	120	--	200	2.0	140	Tr	160	56
Experiment 1 Heated Run													
0.2	10.12	690	19	1.5	0.2	210	--	310	5.3	240	Tr	380	78
1.2	10.34	590	14	0.5	0.2	110	--	180	6.0	110	Tr	700	55
2.0	10.29	440	11	0.7	0.2	39	--	68	3.2	68	Tr	590	54
3.9	10.04	310	7.1	1	0.1	15	--	24	1.8	21	Tr	500	60
8.2	9.72	230	7.1	--	0.02	15	5.9	15	1.4	20	Tr	370	56
9.0	9.62	250	10	2	0.1	36	17	9.3	1.7	48	--	330	60
10.0	9.77	200	5.9	1.9	--	9	4.9	6.4	1.5	15	Tr	330	44
13.9	9.70	150	4.4	--	0.1	3.6	--	2.3	0.8	5.5	Tr	300	38
Experiment 2 Room Temperature													
4.0	9.46	160	5.9	22	0.22	97	2.7	5.7	3.3	13	Tr	71	60
Experiment 2 Heated Run													
0.3	8.87	400	22	34	0.17	250	--	160	8.8	110	Tr	160	59
0.9	9.07	310	16	18	0.06	190	0.62	81	9.6	59	Tr	200	55
2.3	9.14	220	14	11	0.06	120	11	21	7.6	26	Tr	230	65
4.0	9.07	170	11	9.1	0.09	68	14	2.4	6	17	Tr	240	62
10.0	8.76	110	9.8	6	0.05	35	4.2	1.0	4.0	9.6	Tr	240	41
17.0	8.69	100	9.6	9.2	0.04	31	5.4	--	3.8	8.8	Tr	230	43
21.2	8.53	90	9.7	9.4	0.06	27	3.3	--	3.3	8.7	Tr	230	43
Experiment 3 Room Temperature													
1.0	9.85	390	4.4	1.2	0.06	70	19	34	4.5	130	7.5	150	84
Experiment 3 Heated Run													
1.3	10.27	660	11	1.9	0.06	89	32	40	7.5	180	16	350	110
2.2	10.27	680	11	--	0.02	84	36	31	8.2	180	11	370	100
6.0	10.58	620	5.6	--	0.12	54	13	29	7.0	88	6.9	730	84
8.0	10.64	540	5.4	1.9	0.12	40	6.8	20	5.7	51	5.8	660	63
13.0	10.51	410	3.3	--	0.06	30	2.4	8.6	4.4	24	5.4	560	51
21.0	10.24	270	3.6	2.5	0.12	24	1.7	3.2	3.8	17	6.6	410	46
25.1	10.03	220	3.6	--	0.02	23	0.3	1.5	3.6	17	6.3	320	48
37.0	9.77	160	3.3	--	--	18	--	0.1	3.3	13	7.2	210	42

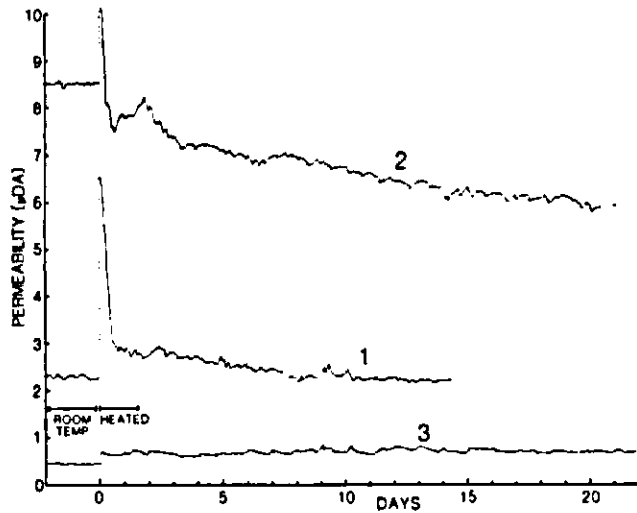


Figure 2. Changes in permeability of the Bullfrog Tuff as a function of time. Numbers refer to experiment number described in text. Experiment 1: 250°C and distilled H₂O; experiment 2: 250°C and J-13 groundwater; experiment 3: 150°C and J-13 groundwater.

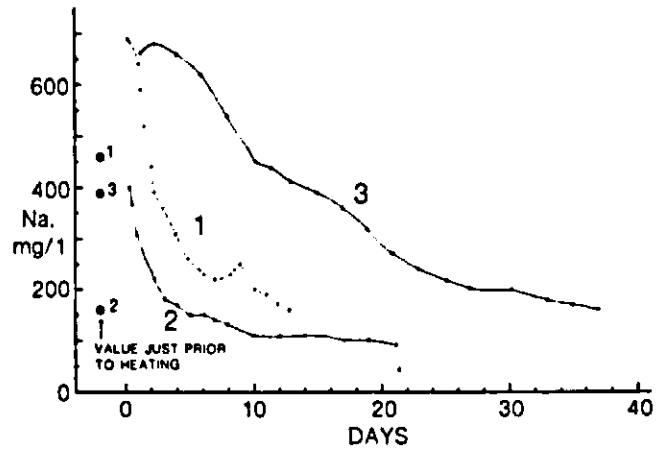


Figure 4. Changes in Na content (mg/l) with time of the discharged fluids from the Bullfrog Tuff cylinders.

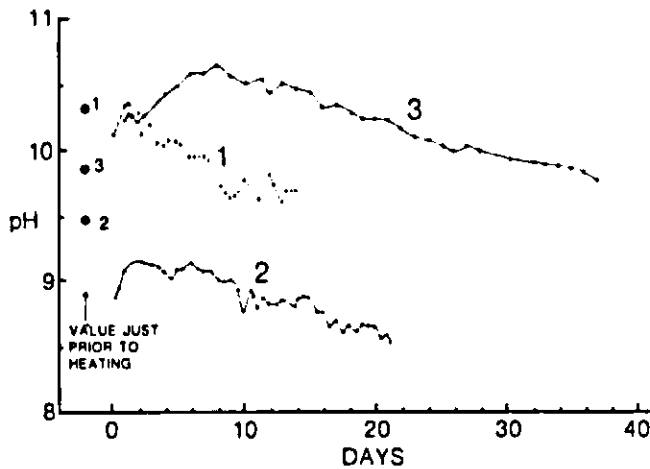


Figure 3. Changes in pH with time of the discharged fluids from the Bullfrog Tuff cylinders.

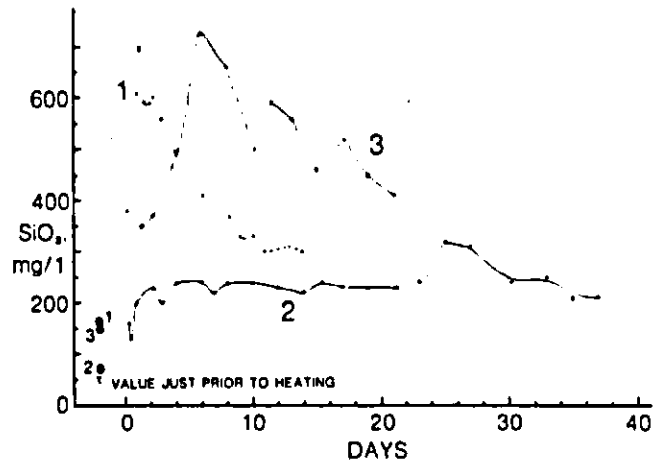


Figure 5. Changes in total dissolved silica (as mg/l SiO₂) with time of the discharged fluids from the Bullfrog Tuff cylinders.

TABLE 3
Dissolved Species (mg/l) in Collected Fluids,
Calculated From Measured Values With
SOLMNEQ Computer Program

Days	H ₄ SiO ₄	H ₃ SiO ₄	H ₂ SiO ₄	HCO ₃	CO ₃
Experiment 1 Room Temperature					
0	19	16	0.15	170	48
2.0	N.A.	N.A.	N.A.	190	310
3.0	35	200	13	98	180
Experiment 1 Heated Run					
0.2	130	460	19	170	210
1.2	150	900	61	92	180
2.0	140	750	43	100	160
3.9	200	580	18	170	140
8.2	250	340	4.8	200	75
9.0	250	270	3.1	230	72
10.0	210	310	4.9	150	63
13.9	210	260	3.4	140	46
Experiment 2 Room Temperature					
4.0	65	48	0.37	220	51
Experiment 2 Heated Run					
0.3	210	42	0.09	280	22
0.9	240	76	0.25	250	26
2.3	270	97	0.36	270	30
4.0	290	89	0.27	240	25
10.0	330	49	0.07	200	7.8
17.0	330	40	0.05	210	7.3
21.2	340	29	0.02	210	5.1
Experiment 3 Room Temperature					
1.0	82	160	3.2	270	150
Experiment 3 Heated Run					
1.3	85	440	26	200	360
2.2	90	470	27	190	330
6.0	90	950	110	81	290
8.0	72	860	120	64	250
13.0	84	730	70	70	190
21.0	110	510	25	110	130
25.1	130	370	11	140	100
37.0	130	200	3.1	150	60

Simplified cation-anion balances determined from the combined values of Tables 2 and 3, are less than 10% for some samples. Possible reasons for some of the larger discrepancies include: (1) The calculations were made at 25°C, but the solution concentrations may still reflect the higher-temperature conditions of the experiments. Unfortunately, the large temperature gradient in the experiments makes it difficult to choose a more appropriate temperature for the SOLMNEQ calculations. (2) For such high-pH solutions, even a slight error in pH can have a significant effect on the calculated ionic concentrations in Table 3. Because of this, the calculated values in Table 3 should be viewed only as approximations of the true values.

Room-Temperature Fluids

The fluids collected during the preliminary, room-temperature permeability measurements acquired considerable amounts of dissolved species (Tables 2 and 3). After 2 or 3 days of pumping, however, the ionic concentrations began to decrease fairly rapidly. The sequence of increasing and then decreasing ionic concentrations is shown for experiment 1 in Tables 2 and 3. Before heating the tuffs, we continued pumping until the conductivity of the discharged fluids was below 1000 μ mhos.

The room-temperature fluids were characterized by high pH, high Na contents relative to other cations, and relatively high SiO₂ contents (Tables 2 and 3). The principal anions in the early-collected, high-conductivity samples were HCO₃, Cl, NO₃, SO₄, and CO₃. Trace amounts of NO₂ and PO₄ were present in some samples. Some Mg and Ca concentrations in room-temperature fluids from experiments 2 and 3 were unusual in being lower than the J-13 water used as pore fluid in the experiments.

Mineral-fluid interactions seem insufficient to account for the high concentrations of dissolved material contained in the room-temperature fluids. The rapid decrease in solution concentrations with time also suggests the removal of a finite amount of readily leached material, rather than continuous mineral reaction. In addition, some species in the fluids, such as NO₃, PO₄, Cl, and SO₄ do not come from minerals in the tuff. Instead, they probably were derived principally from materials that were loosely adhered onto mineral surfaces or deposited on vug walls. Some of the species, such as Cl or S, could have had a magmatic origin, being concentrated in intercrystalline spaces because few of the minerals replacing glass in the tuffs could accommodate them (Ellis and

Mahon, 1964). Species such as NO_3 and PO_4 , may be unique to desert environments or may have had an organic source (West and Skujins, 1978), are found in the J-13 groundwater and may have been deposited along cavities in the tuff by evaporating groundwaters. In support of this possibility, the tuff samples were collected from the sides of an intermittent stream, where they would be continually subjected to alternating wet and dry conditions.

The loss of Mg and, in some cases, Ca from the J-13 waters does suggest that some kind of mineral-water interaction was taking place. However, the low temperatures and relatively rapid flow rates would seem to argue against mineral growth in the short time available. Instead, some simple ion-exchange process, perhaps in clay minerals, may be responsible for the observed decreases. Such exchange reactions would readily occur at low temperatures.

Heated Fluids

The concentrations of dissolved species in the discharged fluids increased rapidly upon initial heating and then gradually decreased again (Tables 2 and 3). The maximum concentrations reached in experiments 1 and 3 were similar to each other and somewhat higher than those of the second experiment. Total silica contents (Fig. 5) and pH values (Fig. 3) generally increased over the first few days, and then gradually dropped; the exception was in experiment 2, where silica increased to 220-240 ppm (as mg/l SiO_2) and then remained constant (Fig. 5). Because the ratio of bicarbonate to carbonate and the amount of dissociation of silica in the solutions (Table 3) vary directly with pH, the concentrations of CO_3 , H_3SiO_4 , and H_2SiO_4 rose and fell with the pH.

By the end of the experiments, concentrations of Na (Fig. 4), total SiO_2 (Fig. 5), and $\text{CO}_3 + \text{HCO}_3$ had leveled out to well above those of the starting fluids, and they dominated the final solutions. Concentrations of Cl, SO_4 , and F decreased rapidly to near zero in experiment 1 and to near the initial J-13 values in 2 and 3. Some NO_3 may have been converted to NO_2 during the experiments, and the final concentrations of both these ions together were lower than for the starting J-13 groundwater in experiments 2 and 3. In addition, all Mg concentrations and some Ca and K concentrations were below J-13 values.

Several competing factors may be important in controlling the heated fluid compositions. Among them are: (1) the removal of additional adhered materials from mineral surfaces, (2) the presence of a temperature gradient, (3) the effects of flow rate, and (4) the

complicated mineralogy of the tuffs. As a result, it is difficult to determine the principal controls of the final, relatively stable solution compositions in the three experiments. However, a few tentative conclusions can be drawn.

Ions such as Cl, SO_4 , and NO_3 , whose concentrations decreased rapidly from high initial values to 0 or J-13 levels, probably comprise those intercrystalline species that were not removed during the room-temperature permeability measurements. The initial concentrations of Na, HCO_3 and CO_3 , and SiO_2 also may have been augmented by materials leached from mineral surfaces; however, their final, relatively high concentrations probably are due to tuff-water interactions, as influenced by temperature and flow rate. As shown in Table 1, the third experiment had the lowest rates of fluid flow and the second experiment the highest. At the lower flow rates, more time was available for material to go into solution. As a result, solution concentrations throughout experiments 1 and 3 were similar, despite the much lower temperatures of the third experiment. Although the first two experiments had similar temperature ranges, the higher flow rates of the second experiment caused solution concentrations to be concomitantly lower.

The use of deionized water or J-13 groundwater in different experiments had little apparent effect on the permeability of the tuffs and no influence on the discharged solution compositions, except to raise the base level of some species. The major importance in using J-13 was in the observation that some elements were lost from the fluids as they flowed through the tuffs. The reduction of the NO_3 contents to values below those of J-13 in experiment 3 may be a function of the instability of nitrate rather than the removal of NO_3 from solution by reaction with the tuff. The apparent conversion of some NO_3 to NO_2 during parts of the experiments also suggests that NO_3 is relatively unstable; the loss of all NO_3 may be caused by vaporization of NO_3 to N_2 gas. In addition, ion exchanges similar to those proposed for the room-temperature fluids may explain part of the observed decreases in Mg, Ca, and K contents.

The growth of minerals such as smectite clays or zeolites in open spaces in the tuff is another possible cause for the reduced Mg, Ca, and K content. From the solution compositions, it is impossible to tell whether mineral growth actually occurred, and the reacted cylinders have not yet been examined to identify any such minerals. However, additional calculations using SOLMNEQ were made to determine the degree of saturation of the fluids analyzed with respect to different minerals at the temperature of the outer edge

of the cylinder rather than room temperature. These calculations, using the fluids of experiment 3, days 8 and 37, indicate that the discharged fluids had at least the potential to deposit some minerals near the low-temperature end of the cylinders.

Because Al determinations were not made, it was impossible to determine whether important aluminosilicate minerals such as clays or zeolites could have been deposited from the fluids. However, at the time of discharge, both the 8 and 37 day solutions of experiment 3 were supersaturated with respect to a number of silica phases, including quartz, chalcedony, and cristobalite. In addition, the 8-day sample was supersaturated with respect to a variety of Ca- and Mg-carbonate minerals.

SIGNIFICANCE OF RESULTS TO NUCLEAR WASTE DISPOSAL

Despite the possibility of mineral growth during heating runs, the permeability of the Bullfrog Tuff cylinders did not decrease dramatically in experiments of up to 5 weeks. Flow rate reductions ranged between 25 and 50%. The permeability change in these experiments contrasts with our previous studies on granite (Morrow et al., 1981; Moore et al., 1983) and on quartzite (unpublished data). The granites and quartzite lacked any large voids, so that fluid flow was concentrated along microfractures and grain boundaries. Material such as silica that dissolved at high temperatures was redeposited at low temperatures in the granite and quartzite samples. Because of the narrow aperture sizes and low flow rates more conducive to mineral reaction, permeability reductions of up to 2 orders of magnitude were measured in the granites, and over 3 orders of magnitude in the very tight quartzite. Similar mineral deposition or growth may have occurred in the Bullfrog Tuff samples, but because of the large initial pore and vug spaces, small decreases in their size caused by mineral growth would have little effect on permeability.

For proposed nuclear waste disposal in unsaturated zones of the Nevada Test Site, the lack of permeability decrease in the examined tuffs apparently is desirable, as it keeps downward-percolating groundwaters from accumulating around the canisters. However, it should be remembered that other tuff layers at the Nevada Test Site may not have such high porosities and permeabilities. As a result, their permeabilities potentially could be more drastically changed by mineral growth accompanying localized heating and fluid flow. In addition, the uptake of Mg, Ca, and some K from solution by the tuff may be important when the materials used in constructing the

nuclear waste repository are considered. The groundwaters are so dilute that any mineral growth resulting from such element uptake would have at best a minor influence on the permeability of tuff layers with large void spaces. However, some proposed backfills and cement grouts may provide a sufficient source of elements such as Ca and Mg to cause extensive mineral growth even in the tuffs considered here, with resulting crack-filling and permeability reductions. Such possibly deleterious interactions between site materials and the host tuff layers should be investigated.

The relatively high pH acquired by the waters flowing through the tuffs may make the solutions corrosive with respect to the canisters. The long-term importance of groundwater pH, if potentially corrosive, to a waste disposal site is unknown. As seen in Figure 3, the pH of the discharged fluids did become slightly less basic with time. In addition, the low rainfall at the Nevada Test Site would limit the amount of water reaching the level of the canisters, which also would limit the amount of possible corrosion.

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