PERMEABILITY AND PORE-FLUID CHEMISTRY OF THE TOPOPAH SPRING MEMBER OF THE
PAINTBRUSH TUFF, NEVADA TEST SITE, IN A TEMPERATURE GRADIENT -
APPLICATION TO NUCLEAR WASTE STORAGE

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ABSTRACT

The Topopah Spring Member of the Paintbrush Tuff from
the Nevada Test Site is being investigated by the Nevada
Nuclear Waste Storage Investigations project (NNWSI) as a
possible nuclear waste repository host rock.  Changes with
time of the permeability and fluid chemistry of the Topopah
Spring Member have been measured in samples subjected to a
temperature gradient.  Maximum temperatures of the imposed
gradients ranged from 90° to 250°C; minimum temperatures
were 36° to 83°C.  Confining and pore pressures simulated
a depth of about 1.2 km, which is greater than the proposed
repository depth, but chosen for comparison with previous
studies at these pressures.  Pore fluid used in the experi-
ments was groundwater from the Nevada Test Site; the
direction of pore-fluid flow was from the high- to the
low-temperature side of the tuffs.

Initial permeabilities of the tuff samples ranged
from 3 to 65 μdarcys, the wide range in values resulting
from differences in the void and fracture geometries of
the samples.  Heating the tuffs produced no change in
permeability in the lowest temperature experiment and only
small changes at higher temperatures.  The fluids
discharged from the tuffs were dilute waters of near-
neutral pH that differed only slightly from the original
groundwater composition.

Since proposed burial in the Topopah Spring Member
would be in the unsaturated zone, the high initial
permeabilities and the absence of permeability change with
heating may be desirable, because downward-percolating
waters would be able to drain into deeper formations and
not collect at the repository level.  In addition, any
fluids that may come in contact with waste canisters will
not have acquired any potentially corrosive characteristics
through interaction with the tuff.

INTRODUCTION

Investigations of the physical and chemical changes that take place
in rocks exposed to high temperatures are important for understanding the
processes that occur around radioactive waste disposal sites.  Morrow
et al. [1] and Moore et al. [2] have measured the changes in permeability
and water chemistry that take place with time in granitic rocks subjected
to a temperature gradient.  The purpose of those reports was to determine
the effect of localized heating on the repository rock around the buried
radioactive waste canisters.  Similar studies are being conducted on two
tuffaceous rock units from the Nevada Test Site that have been proposed
as nuclear waste repository horizons.  Results for the Bullfrog Member of
the Crater Flat Tuff have been described by Morrow et al. [3].  This paper
reports the results for the Topopah Spring Member, lowermost tuff member

of the Paintbrush Tuff, which at the level of the proposed repository is
densely welded, devitrified, and nonzeolitized. X-ray diffraction
analysis of the groundmass of the tuff showed the presence of potassium
feldspar, along with both cristobalite and lesser amounts of quartz. The
samples used in this study also contain elongated lithophysae up to a few
centimeters in length which are not necessarily present at the repository
level. Quartz and trace amounts of calcite were identified by x-ray
diffraction analysis as minerals lining the walls of the lithophysae. A
more thorough petrographic description of this tuff has been made by
Waters and Carroll [4].

EXPERIMENTAL PROCEDURE

The experimental assembly is shown in Figure 1. A cylindrical
sample of the Topopah Spring Member, 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 outer edge 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 pressure gradient between
the center and outside of the rock cylinder. The rock was oriented so
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 reaction with 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 separated the rock cylinder from the outer
jacket; this arrangement allowed removal of fluids that passed through
the rock. Additional details about the experimental technique are
presented in Moore et al. [2].
Samples of the pore fluids were taken at intervals during the
experiments to monitor changes in water chemistry resulting from inter-
action with the heated tuffs. Fluid samples were also collected during
initial, room-temperature permeability measurements. Fluid collection
procedures and analytical techniques are described in Morrow et al. [3].
Three experiments at different temperatures were conducted using the
Topopah Spring Member. The conditions are summarized in Table 1. Ground-
water collected from the Nevada Test Site (J-13) was used as the fluid in
all experiments; the chemical composition of this water is presented in
Table 2. Confining pressures of 30 MPa and pore pressures of 10 MPa were
chosen in order to duplicate the conditions of earlier studies on granites
and the Bullfrog Member of the Crater Flat Tuff. These pressures,
simulating a depth of burial of about 1.2 km, are greater than those
encountered at the depth of the Topopah Spring Member (300-350 m).
Borehole temperatures of 90 and 150°C were chosen for these experiments
to simulate the possible host-rock temperatures in the proposed
repository. The 250°C experiment was conducted in order to compare the
results with the granite studies at this temperature [1, 2].
An attempt was made to maintain a pore pressure differential of
0.15 MPa in each experiment. However, the sample in experiment 3
contained a large void that was continuous from the borehole to the outer
edge of the cylinder, and it was not possible to establish a measurable
pore pressure gradient across the rock without an unreasonably high flow
rate. Therefore, permeability measurements were not made on this sample.
Instead, in the first part of experiment 3, a constant flow rate
$(2 \times 10^{-4}$ cm$^3$/sec) was maintained and changes in water composition
monitored. After about 17 days, the effects of flow rate on the pore
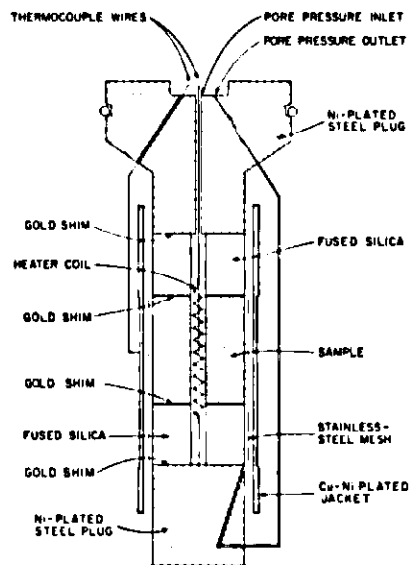fluid composition were investigated by varying the flow rate over four

FIG. 1. Experimental sample assembly.

TABLE I.

| Experiment No. | 1 | 2 | 3 |
|---|---|---|---|
| Borehole temperature (°C) | 150 | 90 | 250 |
| Jacket temperature (°C) | 50 | 36 | 83 |
| Confining pressure (MPa) | 30 | 30 | 30 |
| Pore pressure (Mpa) | 10 | 10 | 10 |
| Differential pore pressure (MPa) | 0.15 | 0.15 | Not Measured |
| Pore fluid | J-13 | J-13 | J-13 |
| Duration of experiment (days) | 20 | 17 | 21 |
| Average flow rate (cm$^3$/sec) | $1.15 \times 10^{-3}$ | $1.45 \times 10^{-3}$ | $2 \times 10^{-4}$ |
| Periods of no flow through the rock during the heated run (days) | | 3.8 - 5.9 | |
| | | 6.8 - 7.8 | |
| | | 9.7 - 13.9 | |

orders of magnitude from $2 \times 10^{-5}$ to $2 \times 10^{-2}$ cm$^3$/sec (Table 2).
Fluid samples were collected at the end of each flow period of a given
rate.

The effect of contact time between the pore fluids and heated rock
cylinder was also investigated in experiment 2, but in a slightly
different way. Flow was halted for intervals of 25, 50, and 100 hours
during the course of the experiment. Fluids were collected immediately
following each period of no flow (Table 2) in order to sample the pore
fluids that had been in contact with the tuff during that time.

## PERMEABILITY

The variations in permeability with time for experiments 1 and 2
were determined from measured changes in the mass flow rate over the
constant pore pressure differential, using the radial flow form of
Darcy's Law:

$$\frac{Q_m}{2\pi\ell} = -\frac{kr}{\nu}\frac{dP}{dr}$$

where $Q_m$ is the mass flow rate, $\ell$ the length of the sample, k perme-
ability, r radius, and dP/dr the radial pore pressure gradient. The
dynamic viscosity of water, $\nu$, is a function of temperature and therefore
varies with radius in these experiments. The variable viscosity is
incorporated into a computer program which calculates permeability from
flow rate and pressure data.

The changes in permeability with time of the tuffs from the first
two experiments are plotted in Figure 2. The initial, room-temperature
permeabilities of experiments 1 and 2 were 3 and 65 µdarcys, respectively.
The large difference is probably due to variations in their internal void
and fracture geometries. A steady state temperature gradient was estab-
lished across the tuff cylinders within an hour of initial heating. In
response to the temperature increase, the permeability of tuff cylinder 1
(150°C borehole temperature) increased immediately to 8 µdarcys and
subsequently rose more slowly to a stable value of 10 µdarcys. During
experiment 2, the permeability of the tuff increased only slightly after
heating to a borehole temperature of 90°C; permeability changed slightly
following the periods without fluid flow, but the values stayed within
the range 60-70 µdarcys.

## FLUID CHEMISTRY

The compositions of the fluids discharged from the Topopah Spring
samples showed only modest changes from the starting J-13 composition
(Table 2). The pH of most of the discharged room-temperature and heated
fluids was reasonably close to the J-13 pH of 7.6. Most of the fluids
collected at room temperature had compositions nearly identical to J-13.
After initial heating, solution concentrations increased slightly, the
amount of increase being directly related to temperature; with time, most
of the ions in solution except K, Ca, and $HCO_3$ returned to J-13 levels.

In experiment 2, the amounts of K, Ca, and $HCO_3$ were higher in the
fluids discharged from the tuff immediately after the nonflow periods,
due to the longer time available for water-rock reaction. The increases
in Ca and $HCO_3$ were proportional to the length of the intervals without
flow. The other dissolved species were not appreciably affected by the
longer reaction times. In experiment 3, the concentrations of most ions
in the discharged fluids decreased progressively toward J-13 levels as
flow rates were increased (Table 2).

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

| Sample | Days | (@ 25°C) pH | SiO$_2$ | Na | K | Mg | Ca | HCO$_3$ | SO$_4$ | NO$_2$ | NO$_3$ | PO$_4$ | F | Cl |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| J-13 Groundwater | | 7.69 | 61 | 44 | 4.5 | 1.9 | 12 | 120 | 17 | -- | 8.1 | -- | 2.2 | 8.1 |
| EXP. 1 | | | | | | | | | | | | | | |
| Room-Temperature | 0.97 | 7.60 | 42 | 53 | 13 | 2.2 | 22 | 170 | 27 | 4.5 | 6.2 | 1.7 | 2.8 | 19 |
| | 13.80 | 7.55 | 42 | 42 | 27 | 1.8 | 27 | 180 | 25 | -- | -- | 4.7 | 2.1 | 13 |
| Heated Run | 0.15 | 7.52 | 86 | 46 | 31 | 1.1 | 17 | 170 | 22 | -- | 0.64 | 6.5 | 2.0 | 13 |
| | 0.40 | 8.09 | 90 | 46 | 26 | 1.1 | 17 | 160 | 20 | -- | -- | 4.7 | 1.9 | 10 |
| | 2.13 | 7.90 | 90 | 43 | 18 | 0.84 | 19 | 140 | 21 | .28 | 1.2 | 2.6 | 2.2 | 9.7 |
| | 7.94 | 6.90 | 87 | 44 | 14 | 0.70 | 19 | 140 | 20 | 2.2 | .71 | 2.5 | 2.2 | 8.4 |
| | 12.85 | 7.38 | 100 | 44 | 13 | 0.59 | 17 | 140 | 19 | 2.5 | -- | 1.7 | 2.2 | 8.8 |
| | 19.85 | 7.38 | 110 | 44 | 14 | 0.57 | 17 | 140 | 19 | .10 | -- | 2.6 | 2.2 | 8.9 |
| EXP. 2 | | | | | | | | | | | | | | |
| Room-Temperature | 0.02 | 6.94 | 54 | 50 | 21 | 1.8 | 12 | 140 | 23 | 1.7 | 6.6 | 1.2 | 2.8 | 14 |
| | 1.71 | 8.00 | 44 | 41 | 14 | 1.6 | 18 | 140 | 18 | 1.1 | 4.2 | 2.1 | 2.2 | 8.9 |
| Heated Run | .88 | 7.62 | 56 | 44 | 12 | 1.6 | 20 | 150 | 20 | -- | -- | 1.4 | 2.2 | 8.6 |
| | 1.88 | 7.63 | 56 | 46 | 17 | 1.5 | 20 | 150 | 19 | -- | -- | 3.2 | 2.1 | 9.3 |
| after: | 3.11 | 7.61 | 56 | 48 | 10 | 1.6 | 19 | 150 | 19 | -- | -- | .96 | 2.1 | 7.7 |
| 50 hrs no-flow | 6.01 | 7.53 | 54 | NA | 16 | 1.9 | 25 | 170 | 21 | -- | -- | .66 | 2.2 | 9.9 |
| 25 hrs no-flow | 7.97 | 7.65 | 55 | 43 | 14 | 1.6 | 22 | 160 | 20 | -- | -- | 1.7 | 2.1 | 9.7 |
| 100 hrs no-flow | 13.96 | 7.32 | 54 | 45 | 15 | 1.8 | 26 | 180 | 21 | -- | -- | .38 | 2.0 | 9.7 |
| | 16.91 | 7.71 | 57 | 45 | 9.6 | 1.5 | 20 | 160 | 19 | -- | -- | .68 | 2.2 | 8.3 |
| EXP. 3 | | | | | | | | | | | | | | |
| Room-Temperature | -- | 7.51 | 110 | 57 | 25 | 2.6 | 24 | 170 | 28 | -- | 6.4 | 2.9 | 3.0 | 22 |
| Heated (2x10$^{-4}$cm$^3$/sec) | .89 | 7.19 | 120 | 68 | 54 | 3.2 | 44 | 240 | 35 | -- | 9.3 | 6.0 | 2.2 | 40 |
| | 2.81 | 7.25 | 150 | 60 | 53 | 2.9 | 56 | 310 | 26 | -- | -- | 3.6 | 1.3 | 21 |
| | 6.75 | 7.28 | 220 | 50 | 43 | 2.4 | 52 | 290 | 21 | -- | -- | 2.2 | 1.5 | 13 |
| | 10.75 | 7.19 | 270 | 50 | 36 | 1.9 | 51 | 260 | 20 | -- | -- | 1.4 | 1.5 | 10 |
| | 16.74 | 7.00 | 270 | 49 | 30 | 1.9 | 43 | 250 | 21 | -- | -- | 1.7 | 1.5 | 8.8 |
| flow rate: | | | | | | | | | | | | | | |
| 2 x 10$^{-3}$cm$^3$/sec | 17.00 | 7.46 | 390 | 43 | 17 | .93 | 32 | 180 | 19 | -- | -- | 1.1 | 2.1 | 8.1 |
| 2 x 10$^{-2}$cm$^3$/sec | 17.06 | 6.92 | 410 | 43 | 9.2 | .34 | 17 | 110 | 19 | -- | 1.3 | .66 | 2.3 | 7.6 |
| 2 x 10$^{-4}$cm$^3$/sec | 17.73 | 7.27 | 280 | 47 | 29 | 1.1 | 31 | 190 | 20 | -- | .18 | 1.0 | 2.0 | 8.2 |
| 2 x 10$^{-5}$cm$^3$/sec | 20.73 | 6.97 | 250 | 55 | 33 | 1.8 | 40 | 270 | 21 | -- | .62 | 1.3 | 1.4 | 8.9 |
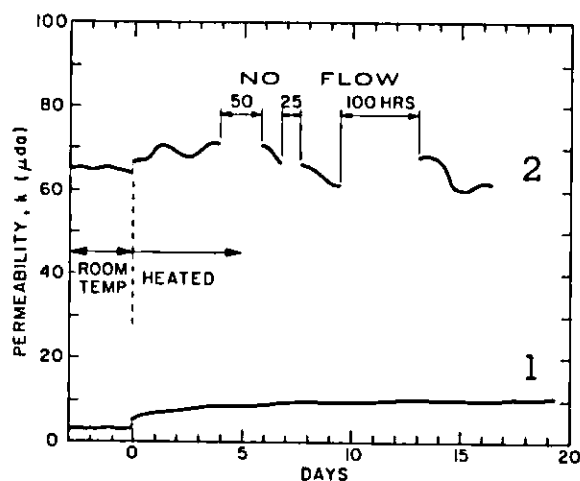
FIG. 2. Changes in permeability of the Topopah
Spring Member as a function of time. Numbers
refer to experiment number described in Table 1.
Experiment 1: 150°C borehole temperature;
experiment 2: 90°C.

The behavior of Mg, F, and $SiO_2$ differed somewhat from the other
constituents in solution. Magnesium was removed from the pore fluids
after heating of the samples in experiments 1 and 2; for example, the
final Mg concentration measured in experiment 1 was only about 30% that
of J-13 (Table 2). The loss of Mg from the fluids suggests either that
this ion was taken up by the tuff in some mineral reaction or that the
pore fluids became supersaturated with respect to some Mg-bearing mineral
which was precipitated. During the first part of experiment 3, when the
flow rate was held constant, Mg concentrations were higher than J-13;
however, at the faster flow rates, Mg was again removed from the J-13
pore fluid rather than added to it. The F contents of the fluids
collected during experiment 3 were also below J-13 levels.

In the room-temperature parts of experiments 1 and 2, some dissolved
silica was removed from the J-13 groundwater as it passed through the
tuffs (Table 2), again suggesting some water-mineral reaction, in this
case involving silica rather than Mg. In the heated runs, $SiO_2$
concentrations were at J-13 levels in the 90°C experiment and well above
J-13 levels in fluids collected during the higher temperature experiments.
In contrast to the ionic species in solution, $SiO_2$ concentrations in
experiments 1 and 3 increased with time over the first several days until
stable values were reached. $SiO_2$ concentrations increased with
increasing flow rate in experiment 3; this may be due to more rapid rates
of silica dissolution than of silica precipitation reactions.

COMPARISON WITH PREVIOUS STUDIES

## Permeability

In our previous studies on granite [1, 2] and quartzite (unpublished data), permeability decreases of up to three orders of magnitude were measured as fluids flowed down temperature gradients through the samples. In contrast to these other results, the permeability of the Topopah Spring Member did not decrease with time during the heated experiments. Instead, the tuff samples showed slight initial rises in permeability, probably related to thermal cracking, and few changes thereafter. In experiments conducted at 150°C, the permeability results for the Topopah Spring were generally similar to those described for the Bullfrog Member [3]. At 250°C, however, the Bullfrog showed an approximately 30% decrease in permeability over a 20-day period.

The differences in permeability behavior between the tuffs and the granites and quartzite may be related to the size of the flow apertures in the different rocks. In the granites and quartzite, fluid flow was confined to grain boundaries and microfractures; the permeability reductions apparently were achieved as minerals were dissolved at high temperatures and then redeposited along the narrow openings at low temperatures. However, the Bullfrog and Topopah Spring specimens were more porous. As a result, the initial permeabilities of the two tuffs were higher than for the granites and quartzite. During the heated runs for both the Bullfrog and Topopah Tuffs, the combination of larger openings and higher rates of fluid flow, with correspondingly reduced times for water-rock interaction, minimized the permeability reductions.

## Fluid Chemistry

The nearly neutral fluids collected from the Topopah Spring have much lower values of pH than the fluids from the Bullfrog, many samples of which had a pH greater than 10 [3]. In addition, the very low initial concentrations of dissolved material in fluids discharged from the Topopah Spring Member are in marked contrast to the concentrations of the first fluids collected from the granite samples [2] and the Bullfrog Member [3]. In the previously studied rocks [2, 3], large amounts of material went into solution both at room and elevated temperatures. The materials in solution at room temperature may have been acquired through mixing with pore fluids [2] or through the leaching of material adhered to grain surfaces and pore walls [2, 3]. The Topopah Spring samples apparently contained very little of such pore fluids or adhered species. With continued fluid flow at room- and elevated- temperatures, much of this readily available material was removed from these other rock types, making smaller the differences in solution concentrations between those experiments and the ones on the Topopah Spring. In the final solutions collected during the heated runs, dissolved species, particularly the cations, varied with the mineralogy of the different rock types.

Some of the other chemical trends observed in fluids from the Topopah Spring experiments are similar to trends observed in fluids from the granites [2], quartzite, and the Bullfrog [3]. For example, following initial heating in these earlier experiments, the total concentrations of ionic species in solution were initially high and then decreased slowly with time, whereas dissolved silica concentrations generally increased for the first several days. In the Bullfrog experiments, which also used J-13 groundwater as the pore fluid, the concentrations of Ca, Mg, and occasionally K in the discharged room-temperature and heated fluids were below the J-13 values. The removal of dissolved material from the pore fluids of the Bullfrog and Topopah Spring Members is evidence that some poorly understood mineral precipitation or exchange reactions took place during these experiments.

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## DISCUSSION OF RESULTS

The significance of initial permeability and permeability changes due to a temperature gradient for rocks composing a potential nuclear waste repository varies with different types of burial sites. In granitic terranes, the buried canisters would probably lie below the water table. In this situation, low permeability is more desirable. In addition, the permeability should remain low even when the rock is heated by radioactive decay in the canisters [1, 2]. Thus if leakage occurs for any reason, the wastes would be prevented from reaching the Earth's surface by pore-fluid migration.

At the Nevada Test Site, the proposed burial site in the Topopah Spring Member is above a deep-lying water table. In this case, buried canisters would not be in continuous contact with heated groundwater, thus reducing the possibility of leakage due to corrosion. In addition, any groundwater percolating down to the burial horizon should be able to flow past the canisters unimpeded, rather than collecting around them.

The initial permeability of the Topopah Spring Member is relatively high compared to granite and also to the Bullfrog Tuff at the Nevada Test Site [3] due to the fracture and void geometry of this rock. Heating of the Topopah Spring Member to temperatures below $100°C$ does not affect permeability, and heating to temperatures of $150°C$ has only a slight effect on permeability. Thus the drainage of fluids through this tuff will probably not be impeded by permeability changes related to heating. In addition, the interaction of heated dilute groundwater with the Topopah Spring Member leads to only minor changes in the fluid composition, and the resulting near-neutral, still-dilute fluids should not adversely affect any canisters with which they might come into contact.

## CONCLUSIONS

1. Permeability measurements made on samples of the Topopah Spring Member at room temperature and in a temperature gradient show that the initially high (3-65 µdarcys) permeabilities are little affected by heating to at least $150°C$. These permeability relationships are favorable for the burial of nuclear waste in an unsaturated zone.

2. The fluids discharged from the samples of tuff during the experiments are dilute, nearly neutral solutions which would be unlikely to corrode canisters containing radioactive waste.

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