PERMEABILITY CHANGES IN CRYSTALLINE ROCKS DUE TO TEMPERATURE: EFFECTS OF MINERAL ASSEMBLAGE

C. A. MORROW, D. E. MOORE, AND J. D. BYERLEE
U.S. Geological Survey, 345 Middlefield Road, MS/977, Menlo Park, CA 94025

ABSTRACT

The change in permeability with time of granite, quartzite, anorthosite and gabbro was measured while these rocks were subjected to a temperature gradient. The highest temperature (at the heat source) was fixed at 250°C, while low temperatures ranged from 60 to 111°C, depending on rock type. Permeability reductions of up to two orders of magnitude were observed, with the greatest reactions occurring in the quartzite. These changes are thought to be caused by dissolution of minerals at high temperatures, and redeposition of the dissolved material at lower temperatures. Quartz appears to be an important mineral in this self-sealing process. If very low permeability is desired around a nuclear waste repository in crystalline rocks, then a quartz-rich rock may be the most appropriate host.

INTRODUCTION

In order to determine the suitability of a potential nuclear waste repository site, the fluid flow characteristics of the host rock must be fully understood. The increase in temperature of the surroundings due to the radioactive decay of the waste material may cause permanent changes in the host rock which could affect the flow regime. In earlier studies, [1, 2], we found that the permeability of intact samples of Westerly granite was considerably reduced after prolonged exposure of the rock to heated fluids in a temperature gradient. These experiments simulated the conditions around a filled repository site. The permeability reductions were thought to be caused by the dissolution of minerals at high temperatures, and redeposition of some of this material within cracks and grain boundaries at lower temperatures, thereby inhibiting fluid flow. Similar experiments on a fractured sample of Westerly Granite [1], where the flow surface was studied under the scanning electron microscope, revealed that fibrous deposits had formed along the fracture at the lower temperature outer edge of the sample. These fibers were preferentially deposited; silica strands formed on quartz grains, whereas zeolitic deposits were found on feldspar grains. The permeability reductions were in good agreement with a model of permeability [3], based on dissolution and precipitation processes at the fluid/rock interface. Kranz and Blacic [4] also observed silica deposits and solution pits in silicate rocks which had undergone permeability reductions. The dissolution of minerals and redeposition of material within cracks and grain boundaries is apparently very effective in reducing the permeability of rocks. Such a "self-sealing" mechanism is considered desirable in saturated, low-permeability repository rocks, since the migration of radionuclide-bearing water into the surrounding environment may be suppressed.

It was suggested [5] that the presence of quartz in the granite was a major factor in the rapid permeability decreases measured during the experiments. In order to test this possibility, we have conducted additional experiments, using a nearly pure quartzite (Death Valley, CA) and two rocks that contain no free quartz, an anorthosite (Split Rock, MN) and a gabbro (Duluth, MN). These three rock types are described in the Appendix; the Westerly Granite sample is described in [2]. The results

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of these new experiments on crystalline rock types are the subject of this paper.

PROCEDURE

Cylindrical rock samples, 7.6 cm in diameter and 8.9 cm long, contained a heater within a central borehole (Figure 1). The heater produced a constant temperature of 250°C at the center of the rock. Temperatures decreased outwards from this heat source, so that along the cylindrical outer surface of the sample, they ranged from 60 to 110°C, depending on the rock type (Table I). Confining and pore pressures were maintained at 30 and 10 MPa, respectively, in all samples. Deionized water flowed radially from the heated borehole to the outer edge of the rock in response to a small pore pressure gradient. A thin stainless steel mesh surrounded the rock, allowing fluids to drain out of the sample, while still enabling the jacket to seal tightly against the rock. The discharged fluids were sampled regularly for chemical analysis.

The variations in permeability with time were determined from measured changes in the flow rate, using the radial flow form of Darcy's Law:

$$\frac{Q}{2\pi\ell} = - \frac{kr}{\nu}\left(\frac{dP}{dr}\right) \tag{1}$$

where Q is the mass flow rate, ℓ the length of the sample, k permeability, r radius, and dP/dr the pore pressure gradient across the radius of the sample. The dynamic viscosity of water, ν, is a function of temperature and therefore varies with radius in these experiments. The variable viscosity is accounted for in a computer program which calculates permeability from flow rate and pressure data.

RESULTS

Permeability

Permeability is shown in Figure 2 for the granite and the three new rock types. The reduction in permeability for the granite was around one and a half orders of magnitude (7.4×10^{-7} to 4.7 to 10^{-8} da). For the anorthosite, the drop in permeability was slightly less (2.7×10^{-7} to 1.6×10^{-8} da). The gabbro behaved in a similar manner to the anorthosite (2.9×10^{-7} to 2.5×10^{-8} da). The quartzite, on the other hand, sealed up almost completely; starting and ending values were 1.0×10^{-7} and 4.5×10^{-10} da, respectively.

In order to compare the relative changes in permeability of the four rock types, normalized permeabilities ($k/k_{initial}$) are plotted in Figure 3. This is a dimensionless plot, with the initial permeability values fixed at 1. As seen in the figure, the quartzite permeability showed the most rapid decrease, with the majority of the reduction occurring in the first 2 days. This sample had the greatest proportional change in permeability. The gabbro permeability also exhibited a rapid initial reduction. After about 4 days, however, the permeability showed little further change; as a result, the gabbro had overall the smallest proportional permeability reduction. The granite and anorthosite permeabilities decreased more gradually during the experiments. It appears that the granite permeability was still decreasing when the experiment was stopped; given more time, the proportional permeability reduction in the granite sample may have been closer to that of the quartzite.

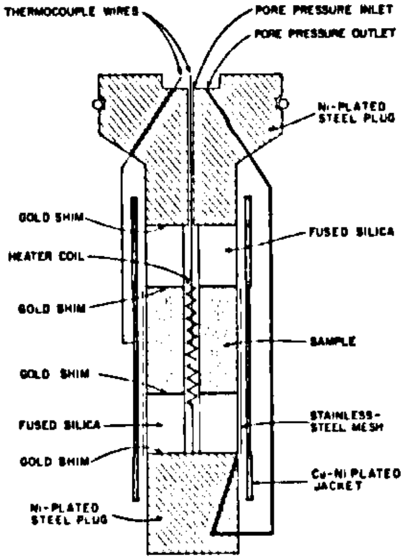

Figure 1. Sample assembly. Water flows from the pore pressure inlet through the central borehole to the sample, then radially out towards the cylindrical face of the rock. It is then collected in the stainless-steel mesh, and drains out through the bottom end plug.

Table I.

Concentration (mg/L) of Dissolved Species in Selected Discharged Fluids*

Rock Type	Jacket temp (°C)	Days	pH	SiO_2	Na	K	Mg	Ca	Al	HCO_3	SO_4	F	Cl	PO_4
Granite	84	1.0	6.8	210	34	13	3.9	53	NA	110	18	3.5	82	-
		5.0	7.6	250	27	9.0	2.1	33	NA	110	9.0	2.6	36	-
		8.9	7.2	180	28	7.6	1.9	31	NA	130	7.0	3.2	21	-
Quartzite	111	2.0	7.0	240	14	12	1.7	29	NA	69	30	.45	9.0	-
		14.9	6.8	170	16	12	2.4	37	NA	94	45	.85	12	-
Gabbro	62	2.0	7.0	50	43	2.4	0.3	1.5	.73	74	3.3	.55	5.1	1.4
		6.1	7.1	55	36	2.0	0.4	0.8	.06	76	2.5	.32	4.8	5.4
		17.0	7.2	80	33	1.1	0.2	0.4	.18	67	1.2	.40	3.2	3.2
Anorthosite	61	1.0	6.8	35	13	7.3	0.6	11	.43	61	2.1	.54	8.2	-
		4.0	6.7	74	12	3.7	0.4	4.3	.11	40	1.8	.18	2.8	0.6
		20.9	6.6	69	12	3.8	0.4	3.2	.05	46	.8	.21	2.	-

NA = not analyzed

*Data accurate to the significant figures shown.

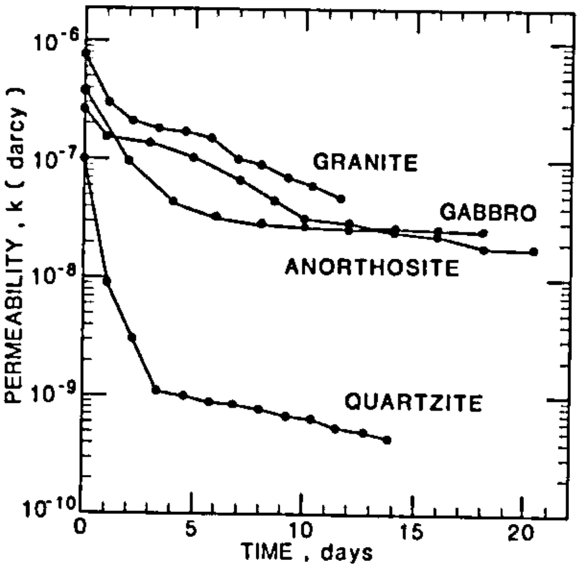

Figure 2. The change in permeability with time of four rock types. All samples were at confining and pore pressures of 30 and 10 MPa respectively, with a borehole temperature of 250°C. Jacket temperatures ranged between 60 and 110°C. The starting time refers to the time of the initial heating of the sample.

Fluid Chemistry

The compositions of the discharged fluids collected at the beginning, middle and end of the experiments are presented in Table 1. Only beginning and ending samples were possible during the quartzite experiment because of the low flow rate. The fluids obtained during the quartzite and granite experiments show several similarities, as do those from the gabbro and anorthosite. The granite and quartzite fluids have significantly higher dissolved silica contents and higher total ionic concentrations than the fluids from the other two rocks. The anorthosite fluids are the most dilute and the granite fluids the most concentrated overall. Sodium and calcium are the predominant cations and bicarbonate is the principal anion in all the analyzed fluids. The fluids from all four rock types are nearly neutral at room temperature; on the average, the anorthosite fluids have the lowest pH and the granite fluids the highest pH.

The higher concentrations of dissolved species in the granite and quartzite fluids may in part be a function of the higher jacket temperatures in those experiments. However, the higher dissolved silica contents of the fluids discharged from the granite and quartzite must also be related to the presence of quartz in these two rocks.

The fluid compositions listed in Table 1 were analyzed with the SOLMNEQ computer program [6], which computes the degree of saturation of the fluids with respect to various minerals as a function of temperature. Since the compositions reflect the state of the fluids as they leave the rock, the jacket temperatures were used in the computations.

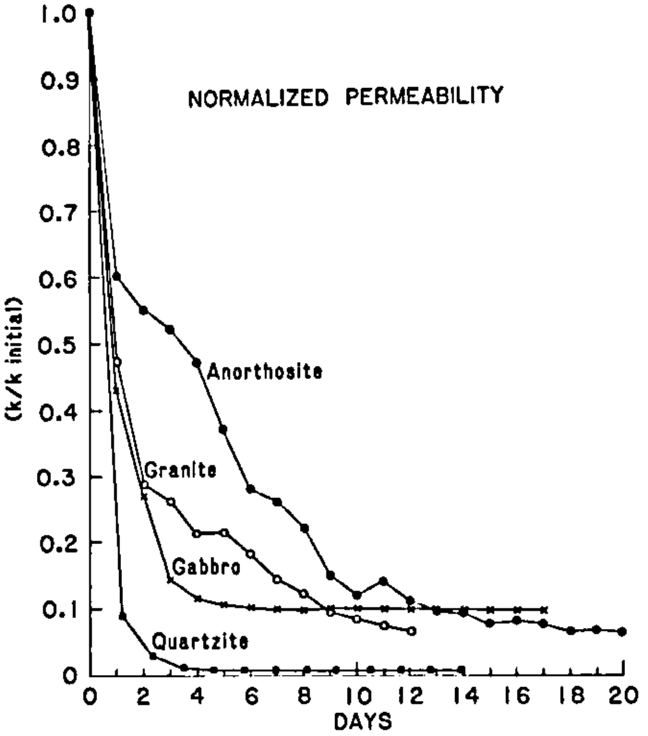

Figure 3. Normalized permeability of the four rock samples. All permeability values are divided by the initial permeability so that starting values are all fixed to 1.

TABLE II.

Reaction States (ΔG_R, in Kcal) of the Fluid Samples Collected at the Discharge Temperature

	Granite (84°C)			Quartzite (111°C)		Gabbro (62°C)			Anorthosite (61°C)		
Days / Mineral	1.0	5.0	8.9	2.0	14.9	2.0	6.1	17.0	1.0	4.0	20.9
Albite (low)						0.89		0.75			
Calcite		0.72	0.11								
Chalcedony	0.85	0.95	0.74	0.60	0.34	0.15	0.21	0.46		0.42	0.38
Illite						7.88	4.00	6.22	7.14	6.56	5.22
Kaolinite						8.47	4.83	6.70	7.79	7.21	6.28
Laumontite						7.99	4.51	6.53	7.70	7.19	5.55
Montmorillonite (Ca)						10.06	5.90	8.36	9.09	8.93	7.69
Quartz	1.27	1.37	1.16	0.96	0.69	0.63	0.69	0.94	0.41	0.91	0.86

Positive values of ΔG_R indicate that the solution is supersaturated with respect to a particular phase. Positive ΔG_R data for the minerals most likely to be associated with these rocks are listed in Table II, including clays, zeolites, calcite, albite and selected silica phases. Because Al determinations were not made during the granite and quartzite experiments, no data is available for aluminosilicate minerals for these two experiments. Therefore, these results are not directly comparable with the gabbro and anorthosite data. The SOLMNEQ program considers only reactions within the fluids. There may be mineral/fluid reactions occurring as well.

DISCUSSION

The permeability reductions and fluid chemistry data of the four rock samples fall into two groups that appear to be related to mineral assemblage. In the first group, the quartzite and granite rocks exhibited large permeability reductions and high concentrations of species in solution. In the second group, consisting of the quartz-free gabbro and anorthosite samples, the permeability reductions were not as pronounced, and the discharged fluids were more dilute.

The permeability changes may be related to the amount of dissolved species in solution, because more material is available for deposition when fluid concentrations are greater. This in turn results in larger permeability decreases as cracks and pores become increasingly clogged with secondary mineral phases. In this way, mineral assemblage plays an important role in the permeability reduction process because of the great variation in the solubilities of each mineral. Rocks containing highly soluble minerals (such as quartz) should show high concentrations of species in solution and possibly large permeability reductions as a portion of this material is redeposited at lower temperatures.

As an example of these differences in solubility, Charles and Bayhurst [7] found in a granodiorite altered by hydrothermal fluids, that the quartz grains were either severely etched or had disappeared entirely at higher temperatures, whereas feldspar grains were only partially dissolved and biotite appeared inert. Secondary phases of montmorillonite and various zeolites were abundant. These results are consistent with the comparative solubilities of each mineral, and also with our observations of permeability reduction in quartz-rich rocks.

A second important factor which influences permeability reductions in these four samples is the geometry and abundance of the cracks and pores where deposition is taking place. In a previous study on tuffaceous rocks from the Nevada Test Site [8], it was found that permeability was not greatly affected by the presence of hydrothermal fluids because of the high porosity (12-36%) and large pore size (mostly equant shaped voids) of the rock. Material deposited in these large apertures had a minimal influence on the permeabilities because the size of the flow path was not significantly reduced.

In contrast, the fluid path in these crystalline rocks (porosity around 1%) is largely through narrow grain boundary and intragranular cracks that are easily clogged by the deposition of clays and zeolites. The quartzite had the lowest initial permeability of the four samples. This low permeability (which indicates very low interconnected porosity), was also a contributing factor in the large permeability reduction, because the flow paths were minimal to begin with.

In conclusion, we have observed that quartz-rich, low porosity rocks showed the largest permeability reductions when subjected to hydrothermal fluids, of the rocks tested. If very low permeability is desired around a nuclear waste repository in crystalline rocks, then a quartz-rich rock may be the most appropriate host.

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APPENDIX. Rock Descriptions

Quartzite. The quartzite consists almost exclusively of equigranular quartz, that averages 0.3 mm in diameter. The quartz crystals characteristically show wavy extinction and sutured grain boundaries. Trace amounts of a fine-grained white mica fill some interstitial areas.

Anorthosite. This sample comes from an anorthosite inclusion within a diabase body at Split Rock, near Duluth, MN. The inclusion is almost wholly composed of plagioclase of composition $An_{70}Ab_{30}$ [9]. The plagioclase is largely fresh and unaltered, tabular in shape and relatively coarse-grained, with crystals ranging from 0.5 to 10 cm in length. The crystals contain many small fractures which are very narrow and apparently unfilled. Some larger fractures and veins contain a fine-grained alteration mineral, possibly kaolinite.

Gabbro. Gabbroic rocks of the Duluth Gabbro Complex can be divided into two groups that were intruded at different times [10]. The samples used in this study most closely resemble the spotted gabbro described by Taylor ([5], pp. 19-20) from the younger and volumetrically more important group of intrusives. Tabular plagioclase crystals, up to 15 mm in length, comprise 50-60 volume percent of the sample. Much of the plagioclase is twinned and compositionally zoned; the crystals show only a minor amount of alteration, that is concentrated along fractures. The plagioclase is enclosed in poikilitic crystals of clinopyroxene and magnetite. The gabbro also contains many rounded crystals of olivine. The olivine and some of the pyroxene crystals show complex alteration.