

Permeability of Granite in a Temperature Gradient

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Changes in permeability of granite were measured as water flowed through samples in a temperature gradient. The experimental conditions simulated those around the 'very deep hole concept' nuclear waste repository. Temperature was maintained between 200° and 310°C, in a borehole of a cylindrical sample. Confining pressures of 30 and 60 MPa, with corresponding pore pressures of 10 and 20 MPa, simulated depths of burial of approximately 1.2 and 2.4 km, respectively. A small pore pressure gradient enabled distilled water to flow from the borehole (high temperature) to the outside of the sample (low temperature). Tests were run for intact samples with initial permeabilities of several hundred nanodarcies and for samples containing throughgoing fractures, with initial permeabilities of about 1 millidarcy. In all cases, permeability decreased between 1 and 2 orders of magnitude at a rate that increased with higher temperatures. At 200°C, permeability dropped by an order of magnitude over a 1-month period, whereas at 310°C, permeability dropped sharply within a few days to 5% of the initial value. The dissolution of quartz and feldspar and redeposition of these minerals within cracks at lower temperature was found to be the major cause of reduction of permeability. If similar processes occurred near a deep hole nuclear waste repository site in granite, then migration of radionuclides away from the site might be suppressed, even if the rock surrounding the waste contained fractures.

INTRODUCTION

Nuclear waste repository schemes, as well as geothermal energy research, have prompted an increasing need to understand the long-term response of geologic environments to fluid flow at elevated temperatures. Fracture systems, as well as thermal and hydraulic gradients, play an important role in the transport of solutions through rock.

The fluid flow properties of crystalline rocks are of particular interest because of their potential use as a nuclear waste repository medium. Their low porosity, low permeability, and high strength are favorable characteristics for the safe containment of nuclear waste.

To date, few studies have investigated flow properties at elevated temperatures. Such information is vital to waste disposal and geothermal studies in assessing the long-term response of these systems. Because of the limited level of understanding of this complex phenomenon we undertook a series of experiments designed to study permeability changes in granite due to the flow of water in the presence of a temperature and pressure gradient. The focus of the study is to observe changes in a setting that simulated a nuclear waste repository; however, the results are equally applicable to problems involved with the extraction of geothermal energy.

Several parameters can affect permeability, including thermal cracking, chemical alterations due to high temperatures, pressure solution, and variable water chemistry. Past permeability studies have touched on these and other topics, and a few studies are mentioned here for their relevance to this investigation.

The effect of stress on granitic rock permeability has been extensively studied by *Brace et al.* [1968], and the transport properties of jointed granite are discussed by *Kranz et al.* [1979], *Pratt et al.* [1977], *Gale* [1975], and *Iwai* [1976]. It is known that the permeability of jointed rock is several orders of magnitude larger than that of intact rock. Also, permeability is greatly decreased by the closure of joints and cracks from applied stress.

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The permeability of crystalline rocks with hydrothermal solutions has been shown both to increase and to decrease with time [*Potter*, 1978; *Tester and Albright*, 1979; *Summers et al.*, 1978]. These results depend largely on the experimental conditions and equilibrium of the system, and clearly different results are desirable depending on the intended application.

Potter [1978] observed both increases and decreases in the permeability of granite as a result of differential thermal expansion and cracking of grains. Permeability minima were dependent on the temperature and pressure environments in which the rocks equilibrated. Cooler temperatures caused contraction and opening of intergranular channelways. Heating above equilibrium caused expansion and crushing of grain boundaries, also producing intergranular channelways and thus higher permeability.

Summers et al. [1978] measured permeability in Westerly granite over several days at temperatures up to 400°C and axial stresses from 0 to 3500 bars. Flow decreased owing to the dissolution of mineral grains and subsequent deposition within grain boundary cracks. The reduction in flow rate did not systematically depend on axial stress.

This progress report is concerned mainly with the effects of temperature-induced geochemical changes on fluid flow through granite under simulated in situ conditions around a borehole in a nuclear waste repository. Future studies will consider other aspects of the problem in more detail.

SAMPLE PREPARATION AND PROCEDURE

Figure 1 shows a schematic view of the sample assembly. Cylindrical samples of Westerly granite, 7.62 cm in diameter and 8.89 cm long with a 1.27-cm borehole, were used in the study. The borehole contained a coiled resistance heater, creating a temperature gradient between the inside and the outside of the rock. Water flowed from the borehole radially out to the edge of the sample. This configuration resembles that of the proposed 'very deep hole' method of nuclear waste isolation, in which canisters would be dropped down deep wells and then plugged with an impermeable material that is compatible with the particular wall rock. Flow away from the borehole in these experiments is of particular interest, as in

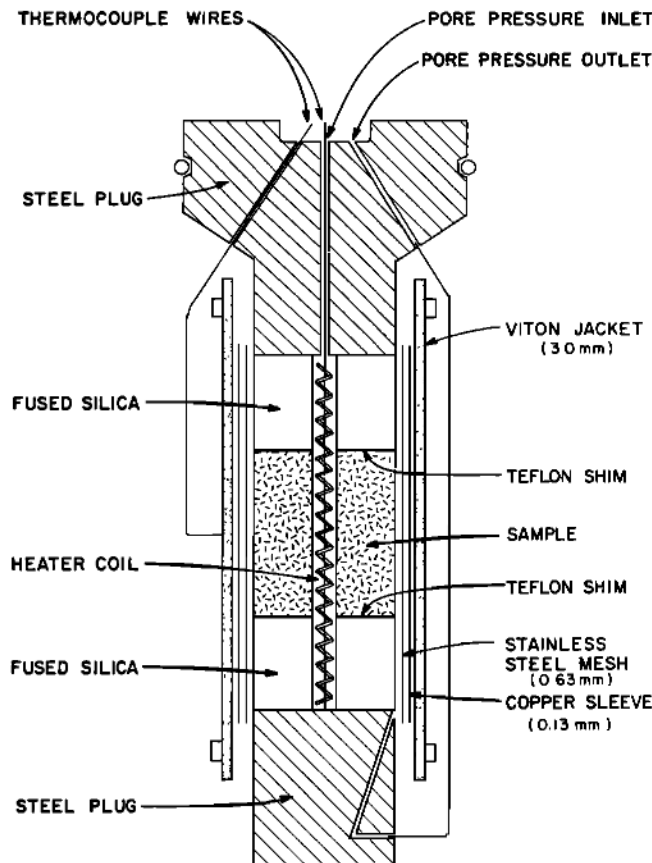


Fig. 1. Schematic sample assembly.

the natural case, radionuclides could migrate away from the waste via groundwater flow.

All samples were wrapped with a thin sheet of stainless steel mesh to allow drainage of pore fluid from the surface of the sample. The mesh was covered with a 0.013-cm sheet of copper. The entire assembly was jacketed in a 0.30-cm viton sleeve to seal out the confining fluid. The copper jacket ensured that the viton did not press through the steel mesh onto the rock surface and inhibit fluid flow. Teflon shims (Figure 1) prevented water from leaking at the interfaces between the sample, silica cylinders, and steel plugs. The fused silica cylinders at the ends of the sample acted as thermal insulators to help maintain a uniform temperature along the length of the sample. Temperature varied by 2°C between the middle and the ends of the sample. Temperatures in the range of 200°–

310°C were chosen for these experiments on the basis of expected temperatures around a borehole [Apps *et al.*, 1978].

Hydrostatic confining pressure and pore pressure were held constant by a microprocessor control system. Distilled water was used as the pore fluid medium to allow chemical reactions to occur between the solution and the rock and at the same time to introduce no new chemical constituents into the system.

In the natural condition, permeability around a borehole would be strongly dependent on structures developed as a result of tectonic activity. For this reason it is important to study the flow properties of both intact and fractured samples. Thus of the three experiments described in Table 1 the sample for experiment 3 was an intact specimen, whereas the samples for experiments 1 and 2 both contained a single throughgoing fracture. These were made by mechanically expanding the borehole, creating two tension fractures 180° apart along the length of the rock.

The confining and pore pressures of experiments 1 and 2 ($P_{\text{conf}} = 600$ bars, $P_{\text{pore}} = 200$ bars) represent a depth of burial of approximately 2.4 km, where confining pressure is due to the weight of the overlying rock and pore pressure results from the hydrostatic head. Experiment 3 simulates a depth of about 1.2 km in the earth. The lower confining and pore pressures ($P_{\text{conf}} = 300$ bars, $P_{\text{pore}} = 100$ bars) were chosen because of the much lower permeability expected from an intact sample. At these pressures, flow rate and differential pore pressure remained in an accurately measurable range throughout the experiment. Current feasibility studies of the very deep hole disposal concept extend to a depth of 10 km [Roy and White, 1975; U.S. Department of Energy, 1979].

Permeability was determined by either (1) monitoring the differential pore pressure between the center and the outside of the sample while maintaining a constant flow rate (experiment 1 and the first half of experiment 2) or (2) measuring a changing mass flow rate over a constant pore pressure drop (experiment 3 and the last half of experiment 2). Differential pore pressure could be resolved to 0.01 bar and was kept low (5 bars or less) to ensure that flashing to steam was not caused by a sharp pressure gradient; thus pore fluids remained in liquid at all times. The flow condition in experiment 2 was changed midway through the experiment to prevent a large buildup of differential pore pressure.

Permeability is not uniform throughout the sample, owing to the imposed pressure and temperature gradients and non-uniform mineral alteration. Thus reported permeability is an average value over the radius of the sample. However, we are

TABLE 1. Experimental Conditions

	Experiment		
	1	2	3
Sample configuration	split	split	intact
Confining pressure, bars	600	600	300
Pore pressure, bars	200	200	100
Flow condition	constant flow rate	constant flow rate and constant head	constant head
Mass flow rate, g/s			
initial	1.5×10^{-4}	1.5×10^{-4}	2.0×10^{-4}
final	1.5×10^{-4}	2.4×10^{-5}	1.6×10^{-6}
Temperature, °C			
borehole	200	280	310
jacket	95	125	115
Duration of experiment, days	32	6	12

TABLE 2. Flow Rate and Resulting Pressure at Room Temperature

	Mass Flow Rate Q_m , g/s	Differential Pressure, bars
	2.88×10^{-3}	5.8
Flow rate decreased	3.02×10^{-4}	0.6
Flow rate increased	2.88×10^{-3}	6.0

not so much interested in the exact value as in the significant changes in permeability that occur over time.

Permeability for the intact sample was determined assuming that Darcy's law holds. For radial flow,

$$\frac{Q_m}{2\pi l} = \frac{kr}{\nu} \frac{dP}{dr} \quad (1)$$

or

$$k = \frac{Q_m}{2\pi l \Delta P} \int \nu \frac{dr}{r} \quad (2)$$

where Q_m is the mass flow rate, l is the length of the sample, k is permeability, r is the radius of the sample, dP/dr is the pressure gradient between the center and the outside of the sample, and ν is the dynamic viscosity of water; ν is a function of temperature and therefore varies with radius in these experiments. This equation is not appropriate for describing flow in the two cracked samples, because flow is not uniformly radial but concentrated along the crack. The parallel plate model analogy to Darcy's law [Gale, 1975] is often used to describe flow in this case. Flow per unit crack length is given by

$$Q = (d^3/12\nu)(dP/dx) \quad (3)$$

where d is the separation between two parallel plates. To test if the parallel plate model was valid for describing flow through the irregular hydrofracture surface, we tested the proportionality of flow rate to differential pressure at room temperature. When flow rate was increased or decreased by a factor of 10, the pore pressure also changed by the same amount. The results are shown in Table 2. The absolute values of flow rate and pressure are not significant here, only their ratios. Since the flow rate was indeed proportional to differential

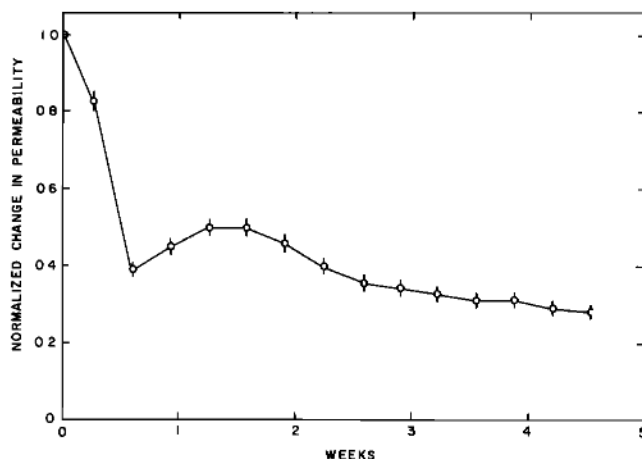


Fig. 2. Normalized change in permeability of experiment 1 sample over a period of 32 days. Sample contained a throughgoing fracture and had a borehole temperature of 200°C.

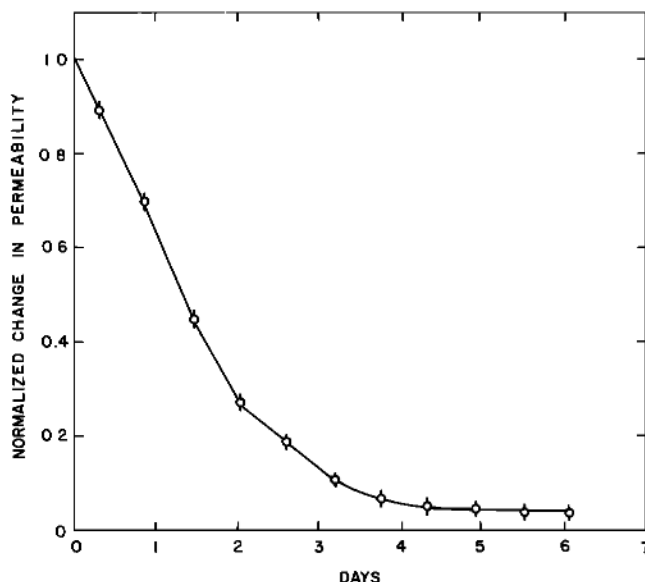


Fig. 3. Normalized change in permeability of experiment 2 sample over a period of 6 days. Sample contained a throughgoing fracture and had a borehole temperature of 280°C.

pressure, then the parallel plate model was applicable to the hydrofractured surface.

Because the separation between plates (crack width d) was not known in these experiments, (3) could not be used directly. Then instead of crack width we substitute a constant λ such that the flow per unit crack length is given by

$$\frac{Q}{l} = \frac{\lambda}{\nu} \frac{dP}{dx} \quad (4)$$

The constant λ is similar to permeability in that it relates flow in a crack to the pressure gradient. However, λ has units of distance cubed rather than distance squared and therefore is not strictly a crack 'permeability.'

OBSERVATIONS

Permeability was found to decrease with time for all samples. The effect of temperature is pronounced; Figures 2-4 show the change in permeability with time normalized to the initial permeability. Temperatures refer to the temperatures measured at the borehole as given in Table 1.

The cracked specimen at 200°C showed unexplained irregular behavior in the first 1½ weeks (Figure 2) before a smooth trend in permeability was apparent. After 32 days, permeability had decreased to 30% of the initial value and was continuing to decline at a slow rate when the experiment was ended. At 280°C, again with flow through a crack, permeability decreased rapidly within the first 4 days and then continued more slowly thereafter (Figure 3). The intact specimen at 310°C showed similar behavior, although the steep decline in permeability was more rapid, extending over only a 1-day period (Figure 4).

Initial and final values of permeability for the three experiments are given in Table 3. The permeability of the cracked samples in experiments 1 and 2 is shown as λ , as derived above. Based on flow rates of the experiments prior to rising the temperature, the introduction of a crack into an intact sample raised the flow rate by an order of magnitude.

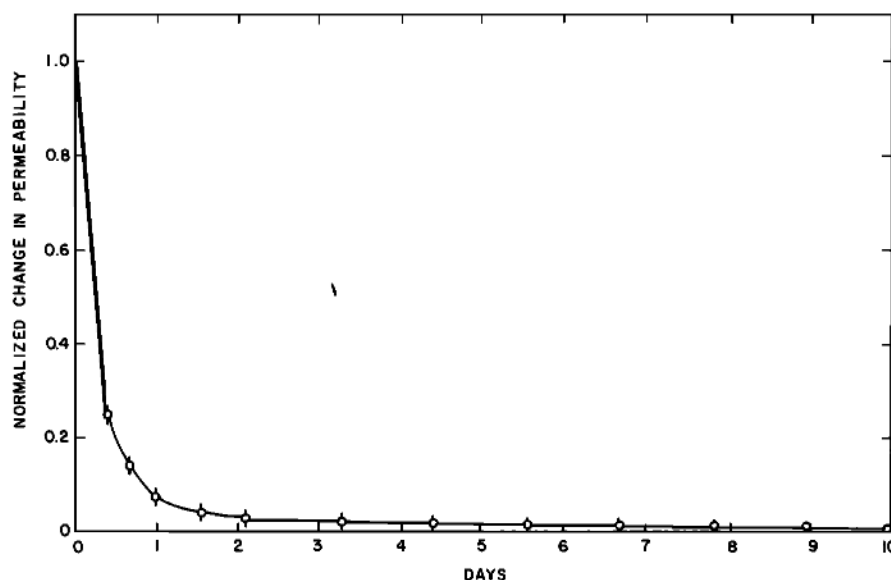


Fig. 4. Normalized change in permeability of experiment 3 sample over a period of 10 days. Sample was intact, with a borehole temperature of 310°C.

The split faces of the samples in experiments 1 and 2 and a fracture surface of the sample in experiment 3 were examined using the scanning electron microscope (SEM) and attached energy dispersive X ray analyzer. The observations were compared with those of a fresh piece of Westerly granite to determine changes as a result of exposure to temperature and flow. Thin sections were also analyzed to find possible differences in mineralogy between the borehole and the outer side of the rock. These two techniques revealed somewhat different information, because deposition along grain boundaries exposed from fracturing was more easily detected with the SEM and thin sections were more useful in observing alteration within grains.

At a borehole temperature of 200°C, dissolved and redeposited silica was found near the edge of the sample with the SEM. The silica occurred as patchy masses or long, thin fibers on top of quartz grains. Silica fibers were not present in the fresh samples of Westerly granite. Figure 5 shows a typical example of deposition features. Here silica is preferentially precipitated on a quartz grain.

At higher temperatures, similar features were observed near the edge of the sample. In addition to silica, feldspar constituents were present in the deposits. Figure 6 shows calcium-rich fibers on top of a calcium-rich plagioclase. These are probably fibrous zeolites, favored over clay precipitation because of the neutral pH of the pore fluids. Alteration products tended to favor precipitation on grains of similar composition. Note how the fibers have grown down the fresh faces of the albite twins exposed from the artificially produced fracture. Such structures were not present in the fresh samples of Westerly granite.

It was difficult to identify unequivocally dissolution features near the borehole by using the SEM because of the irregular fracture surface and misleading surface structures (lines and pits) present in the fresh samples. In future experiments this difficulty can be overcome by observing etching along saw-split polished faces.

Preliminary thin-section analysis yielded evidence of mineral reaction in one of the samples, the intact granite block of experiment 3. Fresh Westerly granite shows some evidence of natural, probably deuteric alteration that in part consists of the partial alteration of plagioclase by sericite, saussurite, and calcite. Generally, the sericitic alteration is a very fine grained, pale green material that is disseminated in the plagioclase crystals. Saussuritic alteration is also fine grained and poorly crystallized, but some good crystals of epidote/clinozoisite are observed.

Reaction features produced during experiment 3 consisted of modifications to these natural, preexisting alteration minerals in plagioclase, in an 8- to 10-mm-thick concentric zone extending into the block from the borehole edge. Within this zone the sericitic material was altered to a distinctive bright yellowish-brown, somewhat coarser grained chloritic phyllosilicate that showed anomalous dark yellowish-green interference colors under the crossed nicols. These yellowish-brown minerals were especially well developed along cleavages and internal fractures of the plagioclase, although small amounts of the phyllosilicate occurred at some grain boundaries. In addition, saussuritic alteration minerals intergrown with the yellowish-brown phyllosilicates may be slightly better crystallized after the experiment on the average than those occurring elsewhere in the sample. With increasing distance

TABLE 3. Initial and Final Permeabilities

	Experiment	Initial λ , cm ³	Final λ , cm ³	Initial k , darcies	Final k , darcies
Calculated from parallel plate model	1	2.94×10^{-4}	8.18×10^{-5}		
	2	1.72×10^{-5}	6.29×10^{-7}		
Calculated from radial flow model	3			3.00×10^{-7}	2.20×10^{-9}

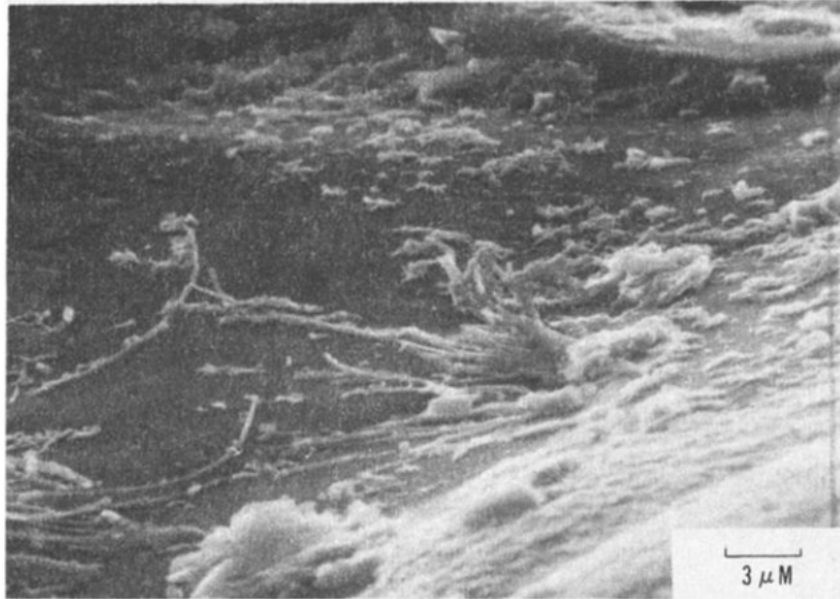


Fig. 5. SEM photomicrograph of deposition features in experiment 1 sample. Silica from solution has redeposited as fibers on a quartz grain. Magnification is 30,000 \times .

from the borehole, alteration features became progressively less pronounced and disappeared entirely 10 mm from the borehole edge.

Similar alteration features were not observed in granite in the other two experiments. In these, flow was concentrated within the fracture, and solution/redeposition features at the surface predominated over chemical reactions within the granitic block. In the intact block all flow must occur along grain boundaries and cracks of the rock itself, which may promote reaction of the water with the minerals in the rock. In addition, the experiment with the intact block was conducted using the highest borehole temperature, 310°C, which may have facilitated reaction. The concentration of reaction products near the heat source in the borehole supports this idea. Cal-

cite and the fine-grained and poorly crystallized sericitic and saussuritic minerals are the minerals most likely to show the most alteration given the short duration of these experiments. At the lower maximum temperatures of the other two experiments, mineral reaction rates may have been slowed sufficiently so as not to be detected.

The observed geochemical changes resulting from flow at high temperatures are consistent with other studies of systems at elevated temperatures. It is worthwhile to mention some of these results because they explain in part the observations of this study as well as reactions that may have occurred but were not evident from the analysis techniques used here.

Quartz solubility has been extensively studied; much of this work is summarized by *Holland* [1967]. For temperatures less

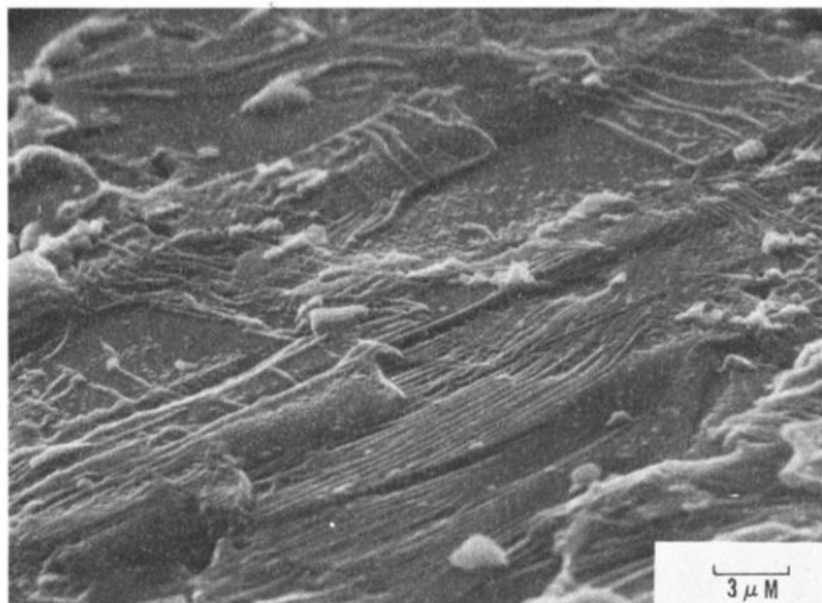


Fig. 6. SEM photomicrograph of fibrous Ca-rich zeolitic deposits on a plagioclase grain of experiment 2 sample. Magnification is 30,000 \times .

than about 325°C, quartz solubility decreases with decreasing temperature and is extremely low at temperatures less than 1600°C. Thus simple cooling is an important mechanism in precipitating quartz [Holland, 1967]. Decreasing pressure at constant temperature also results in a slight decrease in quartz solubility. Dissolution of quartz is essentially independent of pH of the aqueous solution and is also independent of the concentration of dissolved salts in the geologically important range of *P* and *T*. In the pressure range of approximately 250–400 bars, quartz solubility in water for the three temperatures used in these experiments is about 800 ppm at 310°C, about 700 ppm at 280°C, and nearly 300 ppm at 200°C [Kennedy, 1950].

Thus it is not surprising that quartz was observed near the edge of the sample (Figure 5). On the basis of the discussion above we infer that a large amount of quartz dissolved near the heater (borehole). As temperature dropped in the direction of flow, the pore fluid became supersaturated with respect to quartz, and quartz was deposited along cracks and channelways, the amount of deposition varying with *T*, *P*, and the flow rate. Cracks that are clogged by deposition very effectively inhibit fluid flow and thus reduce permeability. Minerals other than quartz behave in a similar manner. For instance, experimental results of a number of workers (see, for example, Gruner [1944], O'Neill [1948], Hemley [1959], Orville [1962, 1963], and Hemley and Jones [1964]) have shown that variations in *T*, *P*, pH, and salt concentration of the coexisting fluid phase influence the stability of feldspar minerals. Feldspars remain stable in near-neutral alkaline solutions in the temperature range 250°–400°C and pressures to 100 bars; at least weakly acidic environments are required to permit hydrolytic base leaching of feldspars to produce muscovite or clays [Gruner, 1944; O'Neill, 1948; Meyer and Hemley, 1967].

Morey and Chen [1955] and Morey and Fournier [1961] passed a continuous stream of distilled water over powdered samples of albite and microcline in closed vessels. Liquids were collected from the vessels and analyzed, and samples and vessel were examined for the presence of alteration minerals. With increasing temperature and pressure, progressively larger amounts of albite and microcline are dissolved into solution. At the same conditions of reaction, albite has a higher rate of dissolution than does microcline. In addition, Morey and Fournier [1961] observed that Na-rich layers of perthitic microcline were preferentially dissolved, leaving behind skeletal crystals of potash feldspar.

Charles [1978] passed distilled water over polished disks of granodiorite at 295°C and 330 bars total pressure for periods of 1–8 months. The disk subjected to 8 months flow of hydrothermal fluid showed 14% weight loss. The reactivity of minerals in the granodiorite was quartz \gg microcline > plagioclase > mafic and accessory minerals. The presence of quartz contributing SiO₂ to solution apparently suppressed the reactivity of the feldspars. Throughout most of the experiment the total concentration of dissolved material in solution was 720 ppm, and the SiO₂ concentration was 680 ppm. The amount of Na in solution was 24 ppm, the K concentration was near 7 ppm, and the Ca concentration was approximately 0.25 ppm. Secondary minerals on the disks included the zeolites philipsite and thomsonite that formed coatings on plagioclase and vermiculite that occurred as prominent black crystals adjacent to mafic minerals. Quench fibers of silica formed on residual quartz grains and on other minerals. Wollastonite and silica precipitated on the experimental apparatus.

It is clear from these examples of feldspar studies that the dissolution and precipitation of feldspars and their breakdown products are significantly more complex than the quartz system. The results of Charles [1978] on the reactivity of minerals support our observation that silica precipitation in cracks was dominant over that of feldspar constituents, particularly at lower temperatures. Also, plagioclase feldspars were preferentially altered over orthoclase, as reported by Morey and Chen [1955] and Morey and Fournier [1961].

DISCUSSION

It has been shown that permeability decreases with time, at a rate that is strongly dependent on temperature. At a borehole temperature of 200°C the permeability gradually decreased to one third of the original value over a period of several weeks. With increased temperature the permeability drop was much sharper, occurring within 4 days at 280°C. For the intact sample at 310°C the major permeability drop occurred within the first day and a half. Final permeability values of the two higher-temperature experiments were about 3 to 5% of the initial values, regardless of the stress applied to the rock mass.

These results agree with those of Summers *et al.* [1978], who found a rapid decrease in permeability with time owing to decreasing solubility of dissolved minerals, and hence deposition and clogging of pores. However, pore water flashing to steam caused by the sharp pore pressure gradient was probably responsible for the marked solubility decrease. The resulting deposition and drop in permeability occurred within the first half day of the experiments, independent of temperatures up to 400°C.

In the experiments reported here, mineral deposition was due mainly to decreasing solubility along the temperature gradient and not to the production of steam through a pressure drop. Differential pore pressure was small, and the fluids remained in the liquid state throughout. Thus the variable rates of decrease of permeability are consistent with the model of a highly temperature dependent system.

Variations of physical factors as well as temperature will affect the time required to achieve a permeability drop. For instance, increased effective stress will tend to close cracks and inhibit flow but at the same time enhance the solubility of minerals in the rock by pressure solution. A decrease in the flow rate will increase the amount of time available for reaction within the rock.

Clearly, several chemical and mechanical parameters can affect permeability. Temperature gradients, as well as groundwater chemistry, in situ stresses, fracture density, and rock mineralogy, must be understood in order to make well-founded decisions on nuclear waste disposal sites.

Our studies demonstrate that the permeability of granite in a temperature gradient decreases with time. This decrease occurs even in samples containing a throughgoing fracture. If the same process occurs in the natural situation, then migration of radionuclides away from a repository in granite may be suppressed, even when the rock surrounding the repository contains preexisting fractures or they are developed at a later time by tectonic activity.

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