Permeability reduction of a natural fracture under net dissolution by hydrothermal fluids

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[1] Flow-through tests are completed on a natural fracture in novaculite at temperatures of 20°C, 80°C, 120°C, and 150°C. Measurements of fluid and dissolved mass fluxes, and concurrent X-ray CT imaging, are used to constrain the progress of mineral dissolution and its effect on transport properties. Under constant effective stress, fracture permeability decreases monotonically with an increase in temperature. Increases in temperature cause closure of the fracture, although each increment in temperature causes a successively smaller effect. The initial differential fluid pressure-drop across the fracture increases by two orders of magnitude through the 900 h duration of the test, consistent with a reduction of an equivalent hydraulic aperture by a factor of five. Both the magnitude and rate of aperture reduction is consistent with the dissolution of stressed asperities in contact, as confirmed by the hydraulic and mass efflux data. These observations are confirmed by CT imaging, resolved to 35 microns, and define the potentially substantial influence that benign changes in environmental conditions of stress, temperature, and chemistry may exert on transport properties. *INDEX TERMS:* 5104 Physical Properties of Rocks: Fracture and flow; 5114 Physical Properties of Rocks: Permeability and porosity; 5194 Physical Properties of Rocks: Instruments and techniques; 5134 Physical Properties of Rocks: Thermal properties; 8135 Tectonophysics: Hydrothermal systems (8424). **Citation:** Polak, A., D. Elsworth, H. Yasuhara, A. S. Grader, and P. M. Halleck, Permeability reduction of a natural fracture under net dissolution by hydrothermal fluids, *Geophys. Res. Lett.*, *30*(20), 2020, doi:10.1029/2003GL017575, 2003.

1. Introduction

[2] Data constraining the role of pressure solution in fractures are sparse, but are available at elevated temperatures (>300°C) in granite [*Moore et al.*, 1994], and at lower temperatures (50°C–150°C) in tuff [*Lin et al.*, 1997]. These are augmented by results for available composite aggregates of quartz [*Elias and Hajash*, 1992], halite [*Gratier*, 1993], calcite [*Zhang et al.*, 1994] and albite [*Hajash et al.*, 1998], at moderate temperatures (23°C–150°C), and the same material suites at elevated temperatures and pressures [e.g., *Zoback and Byerlee*, 1975; *Siddiqi et al.*, 1997]. The limited studies on fractures [*Moore et al.*, 1994; *Lin et al.*, 1997; *Durham et al.*, 2001] suggest an increased sensitivity of their transport properties to thermal, hydraulic, mechanical, and chemical processes, over porous medium flows. This is apparent even at temperatures as low as 100°C, where the mobile dissolved species is silica, the test duration is of the order of a month [*Elias and Hajash*, 1992; *Lin et al.*, 1997], and where permeability may be reduced by a factor of 10^4 [*Lin et al.*, 1997].

[3] We present results from water flow-through experiments in a natural fracture in Arkansas novaculite at an effective stress of 3.5 MPa and at temperatures of 20°C–150°C. The experiment follows progress of dissolution and precipitation, and constrains behavior with the continuous measurement of evolving flow impedence, effluent mass balance of silicon, and fracture porosity recovered from periodic scanning by X-ray CT.

2. Method

[4] The flow experiment was conducted on a cylindrical core (25 mm diameter × 90 mm length) of low porosity (<1%) Arkansas novaculite containing a single diametral natural fracture. Arkansas novaculite has a uniform grain size of the order of 1–6 μm, and quartz content of >99.5% [*Lee et al.*, 1991]. The core-halves are mated, constrained within a Viton® sleeve, and placed inside the low X-ray-attenuation aluminum core holder. End-to-end pressure drop is recorded along the sample with prescribed flow rates in the range 0.225–0.9 ml/m applied at 3.5 MPa confining stress. The sample is held at uniform temperatures in the range 20°C–150°C (Figure 1).

[5] The sample was X-ray imaged concurrent with the flow test. An effective stress of 3.5 MPa was applied to the sample for the duration of the test. Sample temperatures were stepped and held at 20°C (for 25 h), 80°C (50 h), 120°C (400 h), and 150°C (425 h).

[6] Differential pressure between the sample inlet and outlet were recorded to a resolution of ±40 Pa throughout the duration of the flow-through tests. The negligible porosity of the novaculite matrix [*Lee et al.*, 1991] enabled flow rates to be converted directly to equivalent hydraulic aperture via the parallel plate approximation, [e.g., *Witherspoon et al.*, 1980; *Silliman*, 1989] as

$$Q = \frac{\Delta p}{12\mu l} b^3 w \tag{1}$$

where μ is temperature-dependent dynamic viscosity of the fluid [$ML^{-1}T^{-1}$], l is length of the sample [L], w is width of the sample [L], Q is flow rate [L^3T^{-1}], Δp is differential pressure [$MT^{-2}L^{-1}$], and b is the hydraulic aperture of the fracture [L].

3. Fluid and Mass Transport Behavior

[7] Changes in fracture transport characteristics that result from the removal or redistribution of mineral mass

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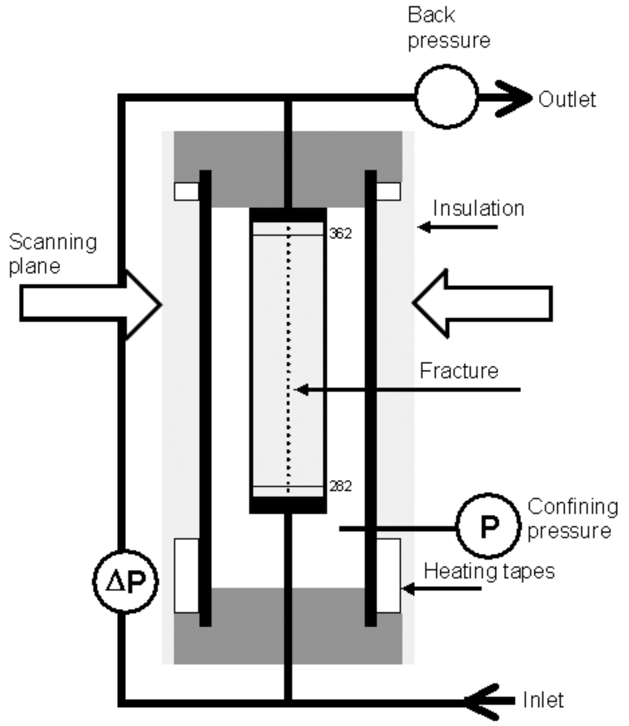

Figure 1. Schematic of flow-through experiment. Preheated water is circulated through the uniformly heated interior core sample and effluent pipe.

within a fracture may be constrained by measuring the evolution of permeability with the concurrent loss or redistribution of mineral mass. Mass loss from the sample is unambiguously recorded by the effluent mineral flux, but redistribution may only be discerned by imaging. Independent measurements of fluid and mineral flux, concurrent with non-invasive imaging, are used to constrain the processes controlling mass redistribution within the fracture. Measured fluid fluxes constrain hydraulic apertures; mineral fluxes and imaging constrain changes in mechanical apertures [*Piggott and Elsworth*, 1993].

3.1. Observations

[8] The evolving change in fracture aperture, under a constant effective stress of 3.5 MPa, is evaluated from the recorded flow-rates and differential pressures (equation (1)), as shown in Figure 2. The initial hydraulic aperture of 12.3 μm at 20°C, falls to 11.4 μm with the initiation of the test. This closure is interpreted as the minor crushing of asperities and interstitial propping grains, as the fracture seats. Heating the sample to 80°C results in a sharp decline in effective hydraulic aperture that ultimately asymptotes to 8.3 μm at 75 hours. Rapid heating of the fracture to 120°C, under conditions of constant effective stress, sharply reduces the fracture aperture to 4.4 μm within 60 hours followed by a slow decrease of a few tenths of a micron spread over the following 100 hours (at constant temperature). The slow monotonic reduction in the fracture aperture is attributed to the redistribution of quartz within the fracture. This could result from the combined effects of dissolution of contacting fracture asperities or propping grains, and precipitation in flow-throats. Two sharp decreases in fracture aperture are apparent at 103 h and 115 h and are correlated with shutdown of the water injection pump for 2 min and 1.5 hours, respectively. A third resulted from pump failure. In all instances, changes are sufficiently rapid (visible instantly after the flow reinitiated) that asperity breakage under elevated effective stress is the likely cause. These stress-related reductions in aperture are irreversible. The core was finally heated to 150°C (430 h after the beginning of the experiment), resulting in a slow steady decrease in aperture over the following 500 hours at constant temperature (from 3.8 to 2.7 μm). The overall change in aperture over the 900 h test is of the order of 9.6 μm, representing an 80% reduction in the initial aperture, and slowing significantly with test progress. These changes are large in comparison with the potential contribution from thermal expansion that would counter the closure due to asperity dissolution. For a coefficient of free thermal expansion of 10^{-5}°C^{-1}, and a mean asperity height of 12 μm, thermal dilation over the 130°C range in test temperature is of the order of 25 nm, and negligible in comparison with the observed dissolution response.

[9] Changes in Si concentration measured during the first one hundred hours of the test are shown in Figure 2. The elevated concentration at 20°C, relative to the concentration at 80°C (0.85 vs. 0.58 ppm respectively), likely represents the rapid removal of crushed particles surrounding the contacting asperities. Concentrations measured at temperatures of 80°C to 120°C are in the range 0.75 to 0.88 ppm, with a mean of 0.77 ppm. A mean concentration of 0.77 ppm, distributed through the 0.0196 m^3 of water injected during the test, corresponds to 0.015 g of Si removed, or 0.03 g of quartz.

3.2. Mechanistic Behavior

[10] Apparent from the continuous increase in impedence throughout the duration of the test, mean hydraulic aperture monotonically decreases. The hydraulic measurements yield rates of aperture closure, *db*/*dt*, of the order of 10^{-11} to

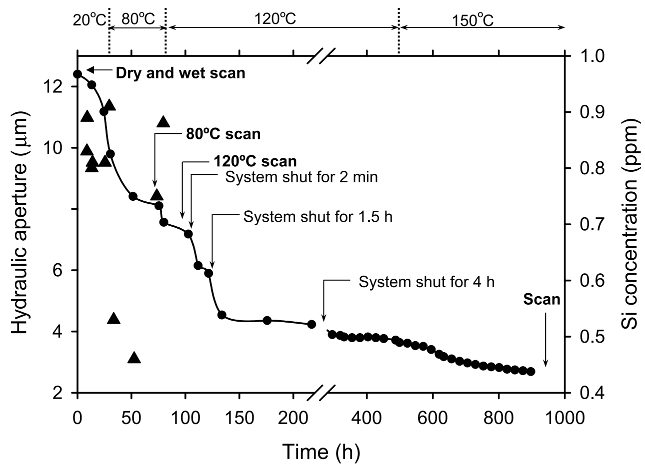

Figure 2. Change in fracture aperture with test duration. Hydraulic aperture evaluated from measured steady flux and axial pressure differential (equation (1)). Scan sequences, temperature transitions, brief test interruptions and aqueous concentrations of silicon measured from the effluent fluid (triangles) are noted.

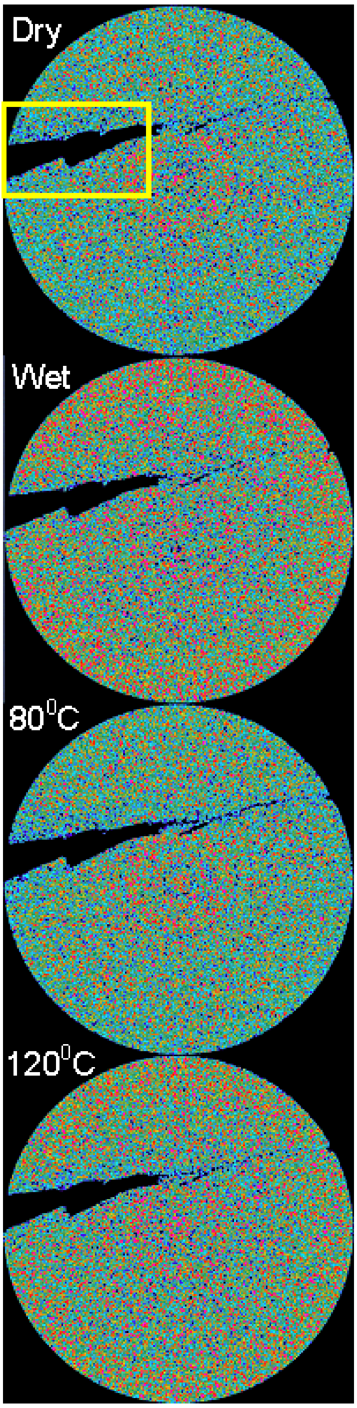

Figure 3. Scanning sequences for the core 57 mm downstream of the inlet of the 90 mm long core. Scans are for the initial dry core, and then wet at 20°C, 80°C and 120°C. The boxed region is processed in Figure 4 to define dissolved mass and relative displacement of fracture faces.

10^{-13} m/s, as recovered from the data of Figure 2. This closure rate is directly measured, does not require a priori knowledge of asperity contact area, but is unable to distinguish between likely mechanisms of fracture closure - dissolution of asperities, or precipitation over the fracture walls will result in indistinguishable signatures. The net removal of quartz throughout the test suggests that dissolution at asperity contacts is the dominant mechanism effecting the reduction in hydraulic aperture. If the mass flux were supplied predominantly by free face dissolution, the fracture would gape with net effluent mass flux.

[11] Rates of aperture reduction, db/dt, recovered from recorded effluent concentrations, may be compared with those independently recovered from the hydraulic test. For dissolution from beneath the contact area between two fracture surfaces, of area A_c, the rate of fracture closure, db/dt, may be defined as,

$$\frac{db}{dt} = \frac{QC_p}{A_c\rho} \tag{2}$$

where C_p is the concentration of quartz in the effluent [ML^{-3}], and ρ is the density of quartz. This enables the change in aperture to be determined from effluent concentrations and measured flow rates, if fracture contact area is constrained. For flow rates in the range 0.9 (80°C) to 0.225 ml/m (150°C), and with the concentration of Si of the order of 0.77 ppm (C_{pSiO2} = 1.65 × 10^{-3} kg/m^3), unmeasured fractional contact areas in the range 20% to 50% yield $db/dt \approx 10^{-11}$ to 10^{-13} m/s. These rates are congruent with those determined independently from the hydraulic measurements and corroborate the important role of dissolution at contacting asperities in mediating the transport properties of natural fractures. Additionally, for measured flow rates, concentrations, and hydraulically determined rates of aperture change, db/dt, the change in contact area with test progress may be determined directly (equation (2)). Fractional contact area transits from c. 25% (20°–80°C), to 50% (120°C), and is congruent with the range 5–30% reported elsewhere where hydro-chemical effects are not apparent [e.g., *Pyrak-Nolte et al.*, 1987; *Zimmerman and Bodvarsson*, 1996].

3.3. X-ray CT Imaging

[12] Non-invasive imaging may be used to further constrain the role of asperity dissolution in mediating changes in fracture apertures. Imaging interprets mechanical apertures, and at sufficiently high resolution, may discriminate between net dissolution and the combined effects of dissolution and precipitation that merely redistribute mineral mass.

[13] A montage of scans is shown in Figure 3 for a section 57 mm downstream of the inlet. The panels are for the core confined to 3.5 MPa and scanned first dry, and subsequently wet at 20°C, 80°C, and 120°C. Scan occurrences are shown in time in Figure 2, and in space in Figure 1. The panel illustrates the evolution of the sub-horizontal aperture with the progress of the test, where a rock chip is absent. Fracture aperture qualitatively reduces with both progress of the test and increase in temperature. Quantitative changes in aperture may be recovered from the scan area where the rock wedge is absent.

[14] Figure 4 shows a 14 mm wide section of the fracture, 57 mm from the inlet as identified in the inset of Figure 3. Figure 4a identifies displacement of the fracture walls, and Figure 4b the overall change in aperture for a threshold CT-number of 1600. Apparent is a general reduction in aperture with progress of the experiment of the order of 0–50 μm. Net fracture closure is registered, but the magnitude

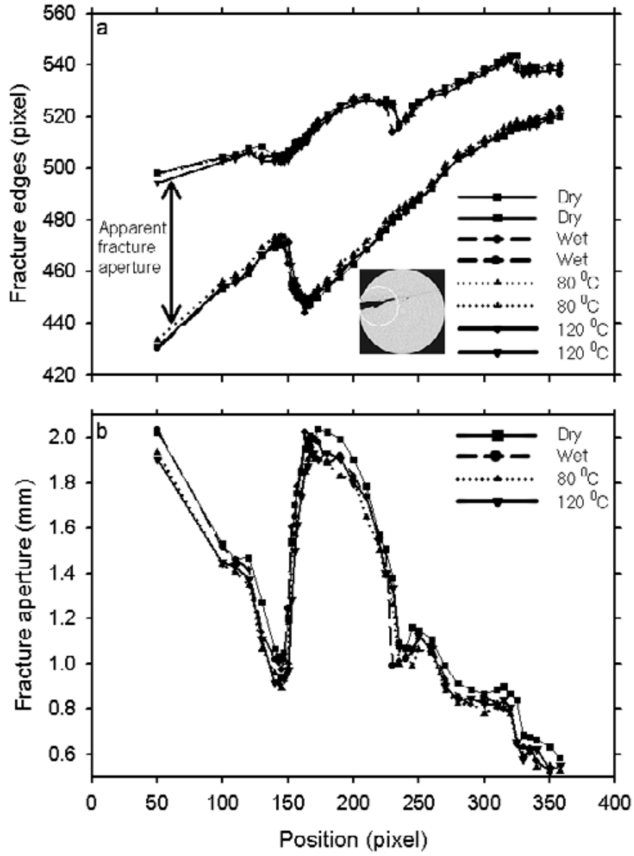

Figure 4. Distribution of fracture aperture, evaluated across the observed fracture 57 mm downstream of the inlet. Profiles are shown for (a) positions of the fracture walls, and for (b) fracture aperture. Data reduced for scans completed dry, and wet at 20°C, 80°C and 120°C.

is much larger than the hydraulically measured c. 10 μm aperture reduction. This is likely due to the rotation of the core halves, around a fulcrum to the right side (Figure 3) of the fracture, with attendant amplification of in fracture displacements. With a pixel size of 37 μm, the resolution of the imaging technique is insufficient to confirm the magnitudes of fracture closure apparent from the hydraulic and mineral mass removal data. Despite this, net closure of the fracture is confirmed from the imaging data, congruent with the hypothesis that mineral mass is predominantly removed from contacting asperities.

4. Conclusions

[15] The results indicate a general correspondence between the mass of quartz removed from the contacting asperities of a conductive fracture, the measured change in hydraulic aperture, and the non-destructively observed removal of mass from the fracture contact area. Measured changes in hydraulic aperture are high, amounting to an 80% reduction in fracture aperture over the 900 h duration test, and a corresponding hundredfold decrease in fracture permeability. The majority of this change in aperture occurs below a temperature of only 120°C. Both the measured absolute magnitude of aperture change, and its rate of reduction, db/dt, are consistent between hydraulic measurements and recorded mineral efflux, and agree nominally with imaged observations of fracture aperture. A consistent model links the change in aperture with the rate of removal of propping asperities in contact. Hydraulic measurements of aperture change are absolute, and require no assumptions other than the applicability of the parallel plate approximation to flow. Constraint with the mineral efflux data requires an assumption of contact area. For contact areas in the range 25–50%, hydraulically observed rates of aperture change in the range 10^{-11} to 10^{-13} m/s are reproduced for the measured efflux concentrations. Consistent with the data, the rate db/dt reduces from 10^{-11} to 10^{-13} with time, corresponding to the general increase in fracture contact area with the progress of the test. Changes in aperture are surprisingly large and rapid, even for modest test temperatures and flow rates, suggesting that benign changes in the thermal, stress, or chemical environment may exert profound changes in transport characteristics.

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