

MODELING THE COUPLED HYDROGEOCHEMICAL AND MECHANICAL BEHAVIOR OF A SINGLE ARTIFICIAL FRACTURE IN NOVACULITE ROCK

J. Gwo, T. Cao, R. Fedors, and J. Rubenstone

U.S. Nuclear Regulatory Commission, Office of Nuclear Material Safety and Safeguards, Washington, D.C. 20555-0001,
U.S.A. Email: Jin-Ping.Gwo@nrc.gov, Tianqing.Cao@nrc.gov, Randall.Fedors@nrc.gov, James.Rubenstone@nrc.gov

The hydrological, geochemical and mechanical properties of the host rock immediately surrounding a high-level nuclear waste repository may undergo physical and chemical changes over the lifetime of the facility. To adequately assess safety performance of a proposed facility, it is important to be able to reproduce and predict the outcome of key experiments targeting the physical and chemical processes. The international DECOVALEX project (DEvelopment of COupled models and their VALidation against EXperiments) was established by a consortium of waste management organizations, regulators and research organizations to help improve capabilities in the interpretation of complex experiments, numerical modeling, and prediction of the behavior of coupled systems. One of the modeling tasks in the current project phase, DECOVALEX-2015, focuses on the laboratory experiments on a novaculite rock column with a single artificial fracture. Novaculite is a microcrystalline quartz rock. The rock column was subject to a mechanical confining pressure and varying applied temperatures. Water flow rates and pressure gradients were measured and hydraulic apertures were estimated, while the chemical composition of the outflow was monitored. The objective of this research is to develop a conceptual framework and physicochemical understanding of the processes that may affect the evolution of the fracture aperture. Multiple and multi-scale coupled hydrological, mechanical and geochemical modeling exercises were performed to predict the evolution of hydraulic apertures and outflow chemical signatures. The comparison of the laboratory and modeling results has led to key understandings in the relative importance of thermal, hydrological, mechanical, and geochemical processes and to the identification of the strength of coupling among these processes in a crystalline hard rock.

I. INTRODUCTION

The host rock immediately surrounding a high-level nuclear waste repository undergoes physical and chemical processes that may result in changes to its hydrological, geochemical and mechanical properties over the lifetime of the facility. Coupled thermal (T), hydrological (H), mechanical (M) and chemical (C) processes in the near field environment surrounding a high-level radioactive waste (HLW, including spent nuclear fuel) repository may

affect the waste isolation capability of the engineered as well as the natural barriers. Geological processes such as pressure dissolution of the rock matrix, thermal stress caused by fission products in the disposed HLW, and the degradation of HLW waste package materials could result in a relatively complex THMC environment for safety performance assessment. In particular, these processes may alter the flow and transport processes in the near field within fractured hard rocks such as granites that are being considered as potential HLW repository host rocks. Fractures are preferred pathways for flow and, if well connected, may lead to transport of radionuclide to potential human exposure pathways in a relatively short period of time.

To adequately assess safety performance of a proposed facility, it is important to be able to reproduce and predict the outcome of key experiments targeting the physical and chemical processes at a range of spatiotemporal scales. The DECOVALEX project (DEvelopment of COupled models and their VALidation against EXperiments) was established and maintained by a range of waste management organizations, regulators and research organizations to help improve capabilities in the interpretation of complex experiments, numerical modeling, and prediction of the behavior of coupled systems. One of the modeling tasks in the current project phase, DECOVALEX-2015, focuses on the laboratory experimental results obtained from a novaculite rock column with a single artificial fracture^{1,2}, with model development supplemented by laboratory experiments on a similar column with a single natural fracture^{3,4}. Novaculite is a microcrystalline quartz rock. Both rock columns were subject to mechanical confining pressures and varying applied temperatures. Fluid (water) flow rates and pressure gradients were measured and hydraulic apertures were estimated, while at the same time the chemical composition of the outflow was monitored.

The objective of this study is to develop a conceptual framework and physicochemical understanding of the processes that may affect the evolution of the fracture apertures which may in turn change the apparent permeability of the rock sample. The laboratory experiments by Yasuhara et al.¹ and Polak et al.³ and multiple, multi-scale coupled hydrological, mechanical and geochemical modeling exercises to predict the evolution of hydraulic apertures and outflow chemical signatures are described in this paper.

II. BACKGROUND AND EXPERIMENTAL SETUP

Disposal of heat-generating SNF and HLW inevitably creates thermal stress on the host rock in the near field of the repository. In response to the thermal stress, flow paths such as interconnected fractures in hard rock (e.g. granite) repositories may be modified by thermal and the accompanying mechanical strains. Strains from external loads may also result in changes to aperture size and distribution in addition to general dissolution and re-precipitation of minerals on open fracture surface. At the pore scale, these strains exert a concentrated stress field on the asperity contact areas between fracture walls. Under the stress field, the rate of mineral dissolution at the asperity contact is accelerated, resulting in a diagenetic compaction and deformation process or pressure dissolution. Under this process, the size of the contact areas may widen but the fracture opening may decrease until a critical stress on the contact areas brings the process to a rest. Stress corrosion cracking, a process where inflowing water induce failure of the mineral grains through pore-scale, chemically enhanced mechanical effects⁵, may also alter fracture surface topography. It usually results in the dislodging of small, crushed mineral fragments around the perimeter of the contact area disc. It may initiate at the onset of the thermal and mechanical strain and potentially accompany the pressure dissolution process until the fracture opening is mechanically or hydraulically closed. As a result of pressure dissolution, the minerals at the contact areas are dissolved and transported by diffusion and advection downstream along the groundwater gradient, resulting in mineralogical signature in the groundwater and on open fracture walls when these minerals re-precipitate. Mineral fragments from stress corrosion cracking may dissolve, deposit in place within the pore space, or be mobilized or remobilized along the flow path.

The two processes, pressure dissolution and stress corrosion cracking, have been rigorously investigated in the past. In particular, laboratory column experiments have been conducted and results analyzed to a great extent by the original investigators as well as the researchers that participated in the DECOVALEX-2015 exercises. The experiments of interest to this study are two Arkansas novaculite cylindrical cores of about 90 mm long, containing a single natural and artificial diametrical fracture, as described in Polak et al.³, and Yasuhara et al.¹, respectively. Arkansas novaculite has a uniform grain size of the order of 1–6 μm , and quartz content greater than 99.5%⁶. The rock samples were confined laterally by tri-axial pressure vessel and the two ends of the core were plugged by steel caps that were pierced by flow tubes. The pressure vessel was heated to various temperatures to control the thermal effect on the rock sample. De-ionized water was injected into the fracture at various prescribed flow rates. Chemical composition and

pH of the water were measured as well as the pressure drops across the samples. For the Yasuhara experiment, the fracture surfaces were profiled by profilometry to measure the roughness of the fracture wall and the fracture opening was scanned by X-ray CT and casted by Woods metal to measure the distribution of the apertures, before and after the experiments, respectively. For the Polak experiment, concurrent X-ray imaging was conducted during the flow test to measure the distribution of fracture apertures. The hydraulic apertures of the entire fracture were calculated by the cubic law on the basis of the measured pressure drops and the flow rates.

III. CONCEPTUAL FRAMEWORK AND GOVERNING EQUATIONS OF A COUPLED THMC SYSTEM

An open, plane-like but rough fracture under thermal and mechanical stresses can be conceptualized as a tortuous two-dimensional network of channels in which the height and width of the channels may change due to pressure dissolution and stress corrosion cracking at asperity contact areas and may affect the hydraulic properties of the channels. The minerals dissolved and mineral fragments dislodged from the asperity contact areas may re-precipitate and deposit within open areas of the channel network as well as be transported outside the channel network. These coupled HMC processes may potentially alter the channel network geometry and influence the mechanical properties of the rock specimen. Some of the participants of the DECOVALEX-2015 project have considered the possibility of this approach². Our approach here is similar, but we assume that stress corrosion cracking does not affect aperture reduction and does not impact the coupled hydrogeochemical processes. These assumptions will be examined later in this paper.

A conceptualization at the entire novaculite rock column scale may be formulated, in which the decrease of fracture aperture may be represented by an empirical formula derived from the surface roughness profile of the fracture openings (e.g., see discussion in Reference 4). Mass removed from the truncated asperity contacts can thus be used to determine the amount of mineral dissolved as well as the reduction of the aperture for hydrogeochemistry calculations. Flow and transport processes are represented by first order mass balance equations accounting for minerals diffusion out of the contact area, their precipitation at open pore space, and their removal from a stir-mixing batch-like system.

Mechanistic understanding of the pressure dissolution process suggests that mass dissolution rate at the asperity contact is a function of mineral properties, e.g., the molar mass of solid grains, usually expressed as the product of molar volume V_m and grain density ρ_g ; the disjoining and critical stresses at the contact of the asperity σ_a and σ_c , respectively; the area of the contact $A_c = \pi d_c^2/4$ where d_c

is the diameter of the contact area; system temperature T ; and the dissolution rate constant of the solid grain k_+ :

$$dM_{diss}^{PS} / dt = \frac{3V_m^2(\sigma_a - \sigma_c)k_+\rho_g A_c}{RT} \quad (1)$$

where R is the gas constant and t is time and the superscript PS indicates pressure dissolution at the asperity contact area. Rate of mass diffusing out the contact area can be represented by a first order differential equation⁴:

$$dM_{diff} / dt = 8\pi D_b \omega (C_{int} - C_{pore}) \quad (2)$$

where D_b is the diffusion coefficient, ω is the thickness of the water film within the asperity contact (assumed to be 4 nm wide), and C_{int} and C_{pore} are the mineral concentrations within the water film and open fracture space, respectively. Dissolved minerals may also come from free-face dissolution within open fracture space:

$$dM_{diss}^{FF} / dt = k_+ A_{pore} \rho_g V_m \quad (3)$$

where A_{pore} is the nominal geometric surface area of the open fracture space and the superscript FF indicates dissolution on free fracture surface. The dissolution rate constant is temperature dependent and can be represented by an Arrhenius-type relation as follows:

$$k_+ = k_+^0 \exp(-E_{k_+} / RT) \quad (4)$$

where k_+^0 is the pre-exponential constant and E_{k_+} is the activation energy. Because the solution in the water is considered far from saturation due to the relatively large hydraulic gradients, precipitation of minerals onto open fracture surface removes negligible amount of minerals from the aqueous phase before the water is discharged from the fracture. Accounting for the aforementioned processes results in the following two mass balance equations for the contact area and the open fracture space, respectively:

$$\frac{dM_{diss}^{PS}}{dt} = \frac{dM_{diff}}{dt} + V_{int}(t) \frac{dC_{int}(t)}{dt} + C_{int}(t) \frac{dV_{int}(t)}{dt} \quad (5a)$$

$$\frac{dM_{diss}^{FF}}{dt} - C_{pore}(t)Q + \frac{dM_{diff}}{dt} = V_{pore}(t) \frac{dC_{pore}(t)}{dt} + C_{pore}(t) \frac{dV_{pore}(t)}{dt} \quad (5b)$$

where V_{int} and V_{pore} are the volume of the contact area and open fracture space, and Q is the flow rate through the open fracture space. Change of aperture, b , is related to the dissolved mineral mass at the asperity contact:

$$\frac{db}{dt} = \frac{dM_{diss}^{PS}}{dt} \frac{4}{\pi d_c^2 \rho_g} \quad (6)$$

Using parameter values reported in Yasuhara et al.⁴ at $T = 150^\circ\text{C}$, the authors compared the calculated apertures from solving equations (1) through (6) with those

obtained in Yasuhara et al.⁴, as shown in Figure 1. The authors note that the mass balance equations in Yasuhara et al.⁴ did not include the last term in equation (5a), which accounts for the storage of dissolved minerals within the contact area as a result of volume change. As the contact area increases over time, more dissolved minerals are retained within the area, resulting in less minerals diffusing out of the contact area. In effect, the reduction of aperture is smaller according to our formulation here relative to that reported in Yasuhara et al.⁴. For the multiscale conceptual framework described in the next section, this increasing volume of the film at the asperity contact area is accounted for by a film disc with an advancing perimeter at the edge of the asperity contact area, with the diffusion of dissolved minerals within the disc film modeled by a second order mass balance equation at the pore scale in a stepwise manner.

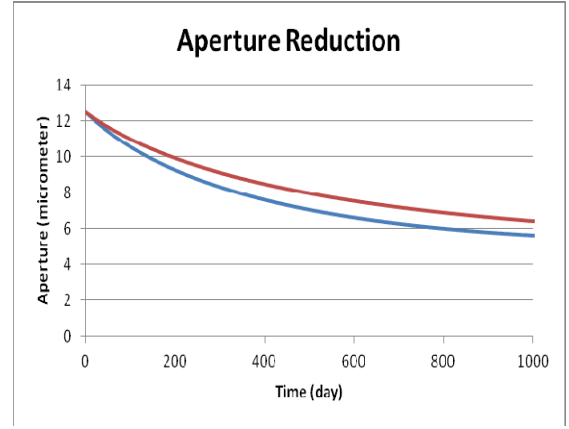


Fig. 1. Decrease of aperture using Yasuhara's equations (blue) and equations 1 to 6 (red) respectively.

Besides pressure dissolution, stress corrosion cracking around the edge of the contacting asperity is another pore scale process that may affect fracture aperture reduction.⁵ Tensile stress concentrations induced near the edge of contacting asperities may mediate fracture growth perpendicular to the open fracture wall according to a chemical kinetic rate law when mechanical effects are expected to dominate and before local stresses drop below an activation threshold (see Figure 2). Rates of aperture reduction due to stress corrosion cracking were shown to be always higher than for pressure dissolution, especially during the initial stage of stress loading and at low temperatures⁵. Stress may also propagate to the free-face area of a fracture and result in free-face corrosion cracking. Although the stress corrosion cracking process is not included in the study and calculations reported here, we note that it may increase the total surface areas around the asperity, thereby increasing the apparent rate of novaculite dissolution. We also note that scanning electron

microscope (SEM) images taken from similar experiments on a granitic rock column with a single artificial fracture⁷ suggest small mineral fragments from crushed contacting asperity as a result of the stress corrosion process deposit near or within asperity contact areas. Sizes of these small mineral fragments are in the sub-micron to a few microns range. We note that the SEM images do not indicate that the mineral fragments have or have not been mobilized or remobilized as a result of change of fluid velocity in the fracture.

According to the stress corrosion cracking conceptual model shown in Figure 2, if and as the mineral fragments dislodge from the asperity cone, the asperity contact area may be reduced. The local disjoining stress may increase; stress corrosion cracking and pressure dissolution processes may continue into a later stage of the aperture closure process. The exact nature and effects of this interaction between the two processes, however, have not been investigated by the previous investigators.⁷

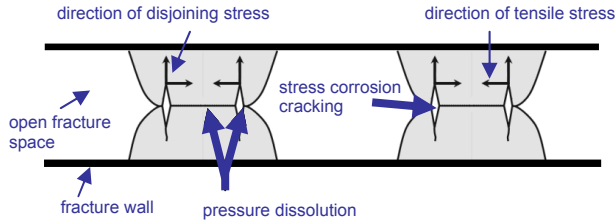


Fig. 2. Pore scale processes that may contribute to reduction of fracture aperture. Pressure dissolution at asperity contacts and stress corrosion cracking around the edge of the asperity contact area are induced by external stress loading and act on different locations around the asperity.

IV. A MULTISCALE MODELING APPROACH

A mechanistic approach may be necessary to account for at least three scales of the THMC coupled processes. At the pore scale, the asperity contact area will widen as more mineral is dissolved over time. Accounting for the widened contact area is necessary as demonstrated above. At the column scale, transport of dissolved minerals, including those from open fracture surface, needs to be accounted for at a length scale orders of magnitude larger than the pore scale. And, for the model to be applicable to realistic field or even repository scale, numerical models such as those based on finite element or finite difference solutions of coupled hydrogeochemical mass and mole balance equations may be necessary. For this research, we consider only the first two scales.

A multiscale modeling approach is thus devised in anticipation of the needs to derive sufficient insights for potential hard rock repository systems. At the contact area, the amount of minerals resulting from pressure dissolution is taken as distributed source into a stagnant

film that is widening. The diffusion of mass out of the contact area can be represented by the following equation, with the asperity conceptualized as one single truncated cone (see Figure 3):

$$\frac{\partial C_{\text{int}}}{\partial t} = D_b \left(\frac{\partial^2 C_{\text{int}}}{\partial r^2} + \frac{1}{r} \frac{\partial C_{\text{int}}}{\partial r} \right) + \frac{1}{V_{\text{int}}(t)} \frac{dM_{\text{diss}}^{\text{PS}}}{dt} \quad (7)$$

where r is the radial coordinate as shown in Figure 3. In reality, there may be numerous contacting asperities, collectively having the same contact area as represented in Figure 3. For hypothesis testing and a simple, first-order analysis, the collection of contacting asperities is represented as one single truncated asperity cone within a unit fracture opening area. The effect of this hypothesis and conceptual representation on aperture reduction and on dissolution within and diffusion of dissolved mineral out of the contact area is discussed later in Section V. For the approach described in this study, equation (2) is substituted by equation (7) with a moving boundary at the perimeter of the contact area film disc. The dissolved mineral discharged from the outer boundary of the film disc is integrated to obtain the amount of minerals entering the open fracture space.

The flow and transport processes of dissolved minerals and their geochemical interactions with the solid phase and other chemical species in the aqueous phase at the column scale are modeled by numerically solving a suite of flow and reactive transport equations of coupled hydrology and geochemistry via the numerical simulator HBGC123D⁸. HBGC123D solves simultaneously the equations for kinetic and equilibrium biogeochemical reactions in saturated and unsaturated porous media. The groundwater and heat transport equations and the ordinary differential and algebraic equations of mixed biogeochemical reactions are solved iteratively. Geochemical processes considered in HBGC123D include aqueous complexation, precipitation and dissolution, adsorption, ion-exchange, redox, and acid-base reactions. Details of the coupled mass transport and biogeochemical system solved in HBGC123D can be found elsewhere.⁹

The modeling approach above was applied to the two rock columns described in Polak et al.³, and Yasuhara et al.¹. The former rock column contains a natural fracture and a modeling exercise of the aperture evolution and outflow chemistry of silicate dissolution were described in Yasuhara et al.⁴, using an approach similar to that described in Section III above. The multiscale approach described in this section is also applied to the experimental results (see next section), with values of model parameters taken directly from Yasuhara et al.⁴. Values of two parameters, the radii of the contact area (r in Figure 3) and the base of the asperity cone, were varied to fit the hydraulic aperture curves from the experiment and to obtain the discharge rate of dissolved minerals

around the perimeter of the contact disc film. The second rock column contains an artificial fracture and the experimental results reported in Yasuhara et al.¹ were also analyzed by the authors. We used the same set of parameter values derived from the modeling exercise for the natural rock column and the same curve fitting approach because both of the rock columns are composed of Arkansas novaculite and the experimental setups and procedures and the THMC processes are nearly identical. The heterogeneous geochemical reactions considered in the modeling exercise include aqueous acid-base reactions of siliceous species (e.g., H_4SiO_4 , H_3SiO_4^- , and $\text{H}_2\text{SiO}_4^{2-}$) and kinetic dissolution and precipitation of dissolved silica (Si). Model parameters of the geochemical reactions were taken from Yasuhara et al.⁴ and standard thermodynamic databases¹⁰.

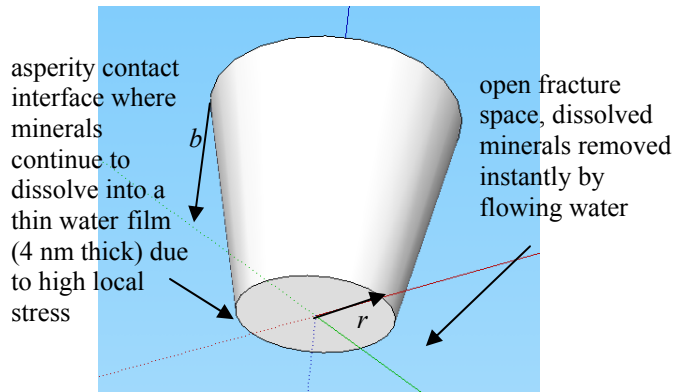


Fig. 3. Conceptual representation of an asperity undergoing localized pressure dissolution in the open space of a fracture.

V. RESULTS AND DISCUSSION

For the Polak column, the flow experiments were conducted at four temperatures (20, 80, 120, and 150 °C, see Figure 4). For our modeling exercise here, the only mechanism that is assumed to be responsible for aperture reduction is pressure dissolution, though stress corrosion cracking could potentially result in additional reduction of fracture aperture. Our approach here is to use the parameters value reported in Yasuhara et al.⁴ to fit the hydraulic aperture evolution curve, by solving the pore scale mass balance equation (7) and then by solving the HC coupled equations at column scale, using HBGC123D to determine the concentration of dissolved mineral species in the discharge fluid.

Figure 4 presents the experimental results and model fit to the hydraulic aperture from experimental observations. The model parameters adjusted to obtain the fit to experimental results are the radii of the base and tip of the truncated asperity cone (Figure 3) for each of the time periods during which the temperature was held steady. As a result, the truncated asperity cone is

essentially representative of the contacting asperities over the entire fracture surface, instead of an individual contacting asperity. The results of the model fitting exercise suggest that the conceptual framework, including a dissolving asperity cone and the mass diffusion process, as represented by equation (7), may be able to describe the aperture reduction process at pore scale.

The calculated contact ratios as a result of this study are in the range of 0.79% to 4.59%, comparing to an estimate of 0.43% to 3.39% and another of 0.43% to 19.09% from Figure 8 of Yasuhara et al.⁴ We note that an additional fitting parameter, or the roughness factor in the ranges of 40 to 300 and 300 to 3000 for the two estimates above, was used in Yasuhara et al.⁴ to improve model fitting to hydraulic apertures. In Yasuhara et al.⁴, the roughness factor was applied to the dissolution rates at the contact area as well as the open fracture surface. The roughness factor essentially multiplies the removal of mass from the asperity contact area predicted by equation (1), but its physical meaning would need to be more clearly defined, especially within the contact area disc film. It was further suggested in Yasuhara et al.⁴ that the latter estimate may produce more reasonable prediction of mineral outflow concentrations. However, laboratory data available for this comparison spans only a fraction of the entire experiment, and the predicted silicate outflow concentrations are at most 50% of the measured.

The conceptual model and governing equations outlined in Section III were also applied to the Yasuhara experiment on a novaculite rock column with an artificial open fracture. More geochemical data are available from the experiment to elucidate the controlling processes for aperture reduction. For our calculations applied to this experiment, only the descending limb of the aperture evolution curve is fitted by the pore scale mass balance equations (1), (6) and (7) because the proposed conceptual framework cannot account for increase in fracture aperture in the later stage of the laboratory experiment. The model fits to hydraulic aperture calculated from laboratory results are shown in Figure 5. The curve fitting approach is the same as that used for the Polak rock column and the outcome suggested that the pore scale conceptual model proposed in this study can be validated.

The calculated, dissolved silica diffused out of the asperity contact area, as calculated at the pore scale, is used as an internal source into a one-dimensional flow tube representing the whole length of the open fracture. The flow tube consists of a finite element mesh with 90 one-dimensional cells, each 1 mm long. Silica dissolution from the solid grains of the open fracture wall also provides additional source for the transport of aqueous mineral species toward the outflow end of the novaculite rock column. The total silicon (Si) concentrations, as measured in the laboratory and calculated in this study, are shown in Figure 6.

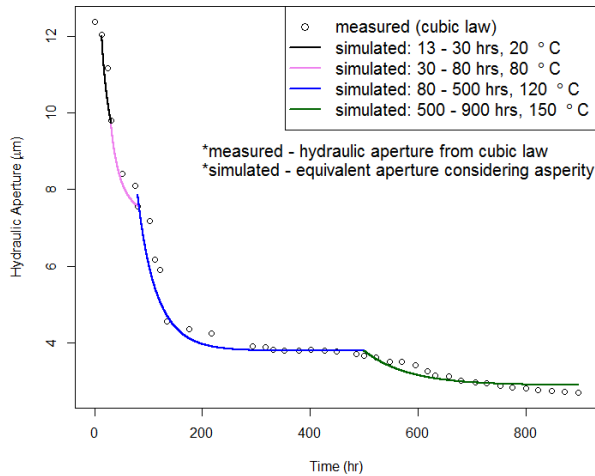


Fig. 4. Fracture apertures measured by Polak et al.³ (circles) and fitted in this study (lines).

The model calculated total Si concentrations in Figure 6 suggest that at 20 °C the primary source of aqueous mineral species is open fracture wall silica dissolution. Contribution from pressure dissolution is only a very small fraction. Furthermore, the model calculated outflow concentrations are about four orders of magnitude smaller than the laboratory measured, suggesting that the conceptual framework as proposed in this study needs to be revised to account for additional silica dissolution and/or transport processes. In particular, we recognize that the internal source term for geochemical reaction and hydrological transport may have contributions from both pressure dissolution and stress corrosion cracking. However, no internal source term contribution from stress corrosion cracking has been characterized by previous investigators.

Other uncertainties regarding modeling an artificial single fracture may include the rough surface of open fracture as evidenced from fracture surface profilometry¹. Roughness on the wall of open fracture surface may increase the surface area of the wall and enhance free-face dissolution by orders of magnitude. Mineral fragments from stress corrosion cracking also provide additional surface area for free-face dissolution. The incoming fluid, assumed as de-ionized water with a pH of 7, may also contain dissolved CO₂, making the geochemical system more complicated than a simple silica dissolution process within a closed system. The high local stress at asperity contacts may also result in highly concentrated solution of dissolved minerals for which the assumption of diluted solution may be questionable for equation (7). Within the asperity contact area, the diffusion of dissolved silica out of the area also slows as the contact area increases and the local stress decreases. This decrease in contribution of internal source term to the column scale transport process is evident in Figure 6 (red line), where the relative

contribution of pressure dissolution to outflow concentration decreases over time after each perturbation of fluid velocity as the asperity contact area and the diffusion path length increase.

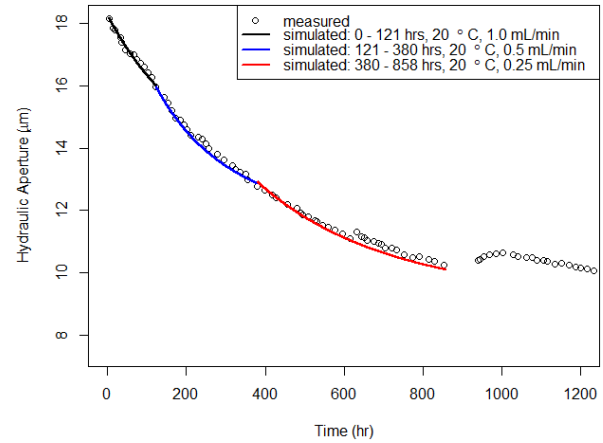


Fig. 5. Fracture aperture calculated on the basis of laboratory experimental data (open circles, Reference 1) and model fitted aperture reduction curves (solid lines).

In the proposed conceptual framework of this study, we collectively represent the contact asperities within a unit fracture opening area as one single truncated asperity cone. This representation has the effect of artificially increasing the diffusion length of dissolved minerals out of the disc film at the contact area. As a result, the internal source term from pressure dissolution by multiple but smaller contacting asperities may be under-estimated. This, in turn, suggests that the proposed conceptual framework needs to be refined and the size distribution of asperities needs to be characterized. The pressure dependence of reaction rate constant is usually weak in the range of pressures normally encountered in the laboratory. On the other hand, reactions under high-temperature and high-pressure are, in principle, both temperature and pressure dependent, although the pressure dependence might vanish under certain conditions¹¹. The calculated local stress at the asperity contacting area is in the order of 100 MPa. Some organic reactions were shown to double the reaction rate when the pressure was increased from atmospheric (0.1 MPa) to 50 MPa¹². The solubility of amorphous silica in seawater at 0°C and from 27 atm (~2.8 MPa) to 1,220 atm (~124 MPa) was found to be a linear function of pressure¹³. At the higher pressure range, the amount of dissolved silica in the solution is known to be supersaturated. The solubility and dissolution rate constants used to obtain model predictions in Figure 6 are assumed to depend on temperature, but not pressure. These two assumptions, one single representative asperity and pressure-independent reaction constants, may have resulted in the five order of magnitude lower predictions of the pressure

dissolution contribution to the outflow Si concentration in Figure 6.

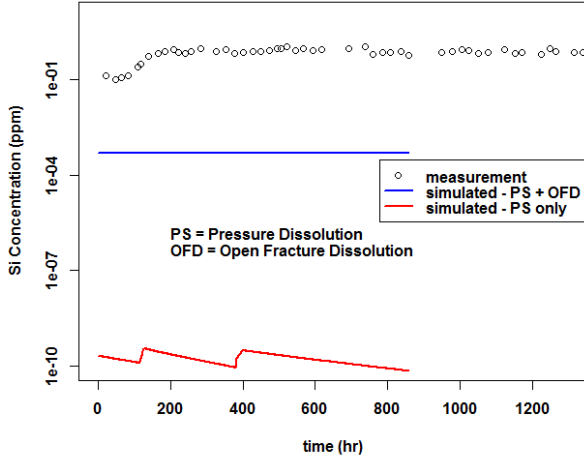


Fig. 6. Outflow dissolved silica concentrations from laboratory measurements (open circle, Yasuhara et al.¹) and model predictions (lines, this study). The blue line includes dissolution of silica in the open fracture space and the asperity contact areas. The red line accounts for pressure dissolution only.

We are also interested in determining the relative sensitivity of the mechanical, hydrological and geochemical processes to the perturbation of thermal field. To this end, a simple sensitivity analysis using the first leg of the Yasuhara experiment was conducted. The temperature of the system was elevated to 80 and 150 °C and the geochemical reaction constants were adjusted accordingly by using the van't Hoff equation. Diffusion coefficient is a function of temperature and pressure, but in general does not vary to a large extent in the range of 20 to 150 °C, assuming the fluid remains in liquid form under large confining pressure. The equivalent aperture of the fracture as well as the internal source term caused by pressure dissolution were recalculated. Thermal perturbation to the fracture flow system apparently has a relatively large impact to hydraulic parameters, in this case the aperture that controls the hydraulic conductivity of the fracture (Figure 7, dotted and dashed lines), under the proposed conceptual framework.

Temperature perturbation also has relatively large impact to open fracture wall silica dissolution as comparing with pressure dissolution (Figure 8, blue and red lines, respectively). Dissolved silica concentration as contributed by the pressure dissolution process is not sensitive to thermal perturbation (Figure 8, red lines), according to the proposed conceptual framework in this study. We nevertheless note that this latter observation is obtained with the understanding of the uncertainties discussed above.

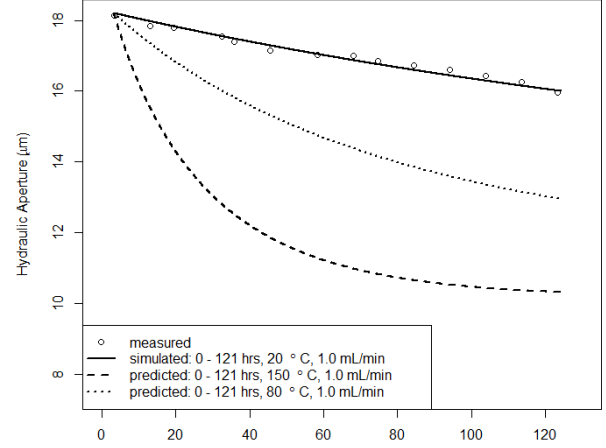


Fig. 7. Effect of temperature on hydraulic aperture, as predicted by the proposed conceptual framework (dotted and dashed lines). The open circles are laboratory measurements¹ and the solid line is model fitted.

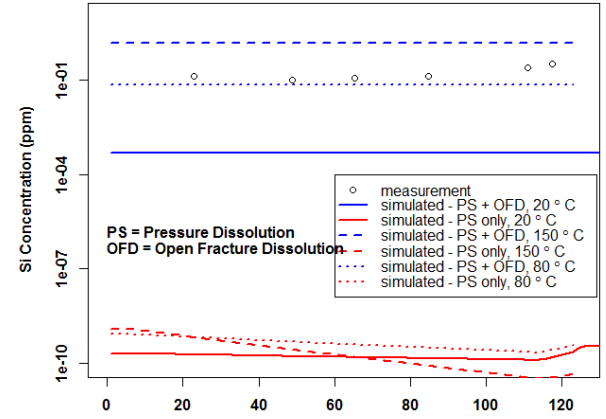


Fig. 8. Dissolved silica concentration sensitivity to temperatures. Model fitted parameters were used to calculate total outflow silicate concentration at three different temperatures, 20, 80 and 150 °C, using inputs from pressure dissolution results at the three respective temperatures. Open circles in the figure are measurements¹ at 20 °C.

VI. SUMMARY

This paper describes a research to study the coupled thermal (T), hydrological (H), mechanical (M), and chemical (C) processes within open fractures in novaculite rock columns. Novaculite is a microcrystalline rock composed of primarily quartz or silica. Two laboratory experiments on novaculite rock columns, each with an open fracture, were conducted by previous investigators.^{1,3} A multiscale conceptual and numerical model was developed in this study to understand the coupled processes and to determine the relative significance of the coupling among individual THMC processes. Model parameters were obtained by fitting the

pore scale pressure dissolution and silicate diffusion equations to the laboratory results. The dissolved silica thus calculated was used as internal source term in a hydrogeochemical model to predict dissolved silica concentration in the discharge fluid. These model calculations were compared with laboratory observations from previous investigations.^{1,3} A sensitivity analysis was also conducted to determine the relative strength of the coupling among the THMC processes.

The modeling results indicate that the multiscale conceptual framework was able to simulate the laboratory observations of fracture aperture reduction, with only two fitting parameters. Hydrogeochemistry modeling results at the column scale, however, indicate that additional processes and parameters may need to be accounted for. In particular, the conceptual framework did not include the stress corrosion cracking process and the likelihood of generation and transport of mineral fragments from the process. It does not consider fracture characteristics such as rough open fracture surface, and the potential existence of dissolved CO₂ in the incoming fluid.

The sensitivity analysis suggests that mechanical stress induced either by external loading or thermal perturbation to the system, has a relatively larger impact to hydrological characteristics of the system, as opposed to the production of dissolved silica from pressure dissolution. For the two geochemical processes, pressure dissolution and open fracture wall silica dissolution, thermal perturbation appears to exert larger control over the latter, under the proposed conceptual framework and the assumption that chemical reaction constants are pressure-independent. The causes of higher Si concentrations in the outflow fluid observed in the laboratory, as compared with model prediction (Figure 6), may need to be identified by further laboratory and modeling studies to reduce model uncertainty.

DISCLAIMER

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

REFERENCES

1. H. Yasuhara, A. Polak, Y. Mitani, A. Grader, P. Halleck, and D. Elsworth. "Evolution of fracture permeability through fluid-rock reaction under hydrothermal conditions," *Earth and Planetary Science Letters* 244: 186–200 (2006).
2. A. E. Bond, N. Chittenden, R. Fedors, P. S. Lang, C. McDermott, I. Neretniecks, P. Z. Pan, J. Šembera, N. Watanabe and H. Yasuhara. "Coupled THMC modelling of a single fracture in novaculite for DECOVALEX-2015," International Discrete Fracture Network Engineering Conference, October 19 - 22, 2014, Vancouver, Canada (2014).
3. 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).
4. H. Yasuhara, D. Elsworth, and A. Polak. Evolution of permeability in a natural fracture: Significant role of pressure solution, *J. Geophys. Res.* 109, B03204, doi:10.1029/2003JB002663 (2004).
5. H. Yasuhara and D. Elsworth. "Compaction of a Rock Fracture Moderated by Competing Roles of Stress Corrosion and Pressure Solution." *Pure and Applied Geophysics* 165: 1289-1306 (2008).
6. V. W. Lee, S. J. Mackwell, and S. L. Brantley. "The effect of fluid chemistry on wetting textures in novaculite," *J. Geophys. Res.*, 96, 10,023–10,037 (1991).
7. H. Yasuhara, N. Kinoshita, H. Ohfuji, D. S. Lee, A. Nakashima, and K. Kishida. "Temporal alteration of fracture permeability in granite under hydrothermal conditions and its interpretation by coupled chemo-mechanical model." *Applied Geochemistry* 26 (2011) 2074–2088 (2011).
8. Gwo, J. P., E. F. D'Azevedo, H. Frenzel, M. Mayes, G. T. Yeh, P. M. Jardine, K. M. Salvage, and F. M. Hoffman. "HBGC123D: A high performance computer model of coupled hydrogeological and biogeochemical processes," *Computers and Geosciences* 27: 1231-1242 (2001).
9. G.-T. Yeh, K. M. Salvage, J. P. Gwo, J. M. Zachara, and J. E. Szecsody. *HYDROBIOGEOCHEM: A Coupled Model of HYDROlogic Transport and Mixed BIOGEOCHEMical Kinetic/Equilibrium Reactions in Saturated-Unsaturated Media*, ORNL/TM-13668, Oak Ridge National Laboratory (1998).
10. C. M. Bethke and S. Yeakel. *The Geochemist's Workbench, Release 7.0, Reaction Modeling Guide*, University of Illinois (2007).
11. H.-H. Carstensen and A. M. Dean. "The Kinetics of Pressure-Dependent Reactions," *Comprehensive Chemical Kinetics* 42: 105–187 (2007).
12. N. S. Isaacs. *Physical Organic Chemistry*, 2nd edition, Adison Wesley Longman, Harlow UK (1995).
13. J. D. Willey. "The effect of pressure on the solubility of amorphous silica in seawater at 0°C," *Marine Chemistry* 2: 239–250 (1974).