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1 Coupled Thermo-Hydrological-Mechanical-Chemical Behavior of
2 Anisotropic Granite for Geologic Disposal of High-Level
3 Radioactive Waste: A Core-Scale Laboratory Investigation

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Abstract: The coupled thermo-hydrological-mechanical-chemical (THMC) behavior of rock within an Excavation Damaged Zone (EDZ) is critical for the safety and long-term performance of a geological repository for high-level radioactive wastes. While many laboratory experiments have been conducted to investigate EDZ rocks, the flow and deformation characteristics resulting from anisotropic rock textures and microcrack distribution under triaxial loading and elevated temperatures remain poorly understood. Particularly, cracks at various scales serve as fast paths for fluid flow and solute transport, and present as focal points of mechanical weakness, which complicate the coupled THMC processes in anisotropic EDZ rocks and challenge modeling predictions. In this study, a series of core-scale experiments was conducted on three granite samples under repository-relevant conditions. These rock samples were obtained from the Grimsel Underground Research Laboratory (URL), featured by anisotropic minerals (represented by bedding layers) and microcrack distributions and coarse cm-scale grain sizes. During the experiments, samples were subjected to an elevated temperature at 90 °C and different triaxial loading conditions either by radial (normal to bedding layers) or axial (parallel to bedding layers) compaction. Water was injected into the samples, and the rock permeability evolutions and effluent water chemistry were monitored closely. For intact samples, thermal expansion of minerals at 90 °C resulted in a large, 75% irreversible permeability reduction and rock strengthening under radial compaction, while thermal impact was limited to a 15% permeability reduction under axial compaction. In contrast, for a sample containing open cracks, the growth of fractures during the experiment resulted in an abrupt permeability increase and fast failure at 90 °C. The effluent water chemistry indicates much more considerable mineral dissolution from large shear sliding than that in

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37 rocks dominated by mechanical compaction. These results helped better understand the
38 coupled THMC processes in anisotropic rocks containing cracks, evaluate the behaviors of
39 EDZ rocks, and predict the long-term evolution of EDZ for the performance of the
40 repositories.

41 **Keywords:** High-level radioactive waste repository; Excavation disturbed zone; Rock
42 anisotropy; Coupled THMC processes; Permeability evolution

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46 **Highlights**

- 47 • The behavior of anisotropic granite was investigated under triaxial loading and at
48 temperatures up to 90 °C.
- 49 • Three samples (two intact and one fractured) from Grimsel Underground Research
50 Laboratory were studied.
- 51 • Various levels of thermal impacts on the permeability of intact core samples were
52 observed, depending on the anisotropy and stress conditions.
- 53 • Compaction of rough fracture accelerated shear failure of sample at 90 °C.

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54 **1. Introduction**

55 The long-term safety of nuclear waste disposal in geological formations relies on both
56 natural barriers formed by rock formations and an Engineered Barrier Systems (EBS) in
57 isolating the waste from biosphere (IAEA, 2003). For the natural rock barrier, the strength
58 and failure of a rock tunnel is time dependent (Lajtai et al., 1991). The strength,
59 geophysical, and hydrological properties of the rock surrounding the tunnel will change
60 with time, and affect the evolution of Excavation Damaged Zone (EDZ). This may lead to
61 the failure of rock formations as natural barriers and the worst case, the tunnel collapse
62 (Bieniawski, 1989). After the emplacement of the waste canister and bentonite buffer, the
63 evolution of EDZ rocks is impacted by multiple coupled factors including stress,
64 temperature, water activity, chemistry and mineralogy. These factors exhibit time-
65 dependent and complex interactions, resulting in significant and permanent rising of EDZ
66 rock permeability (Yasuhara et al., 2016).

67 The presence of fractures in rocks, from microcracks (e.g., Tenthorey and Fitz Gerald,
68 2003) to faults (Tadokoro and Ando, 2002) is critical to fluid flow and transport of
69 radionuclides at a variety of scales. The permeability of a fracture can be easily and quickly
70 changed as fractures undergo opening or closure induced by changes in normal stress, pore-
71 fluid pressure (Barton et al. 1985; Cook 1992; Vogler et al. 2016, 2018), temperature
72 (Morrow et al. 2001; Lima et al., 2019), and chemical equilibrium (Polak et al. 2003;
73 Yasuhara and Elsworth 2008). It has been widely recognized that when the fracture is
74 subjected to normal stress, a mechanical closure in the fracture can occur. The higher stress
75 concentrations at the contacting fracture asperities can cause reversible elastic compaction
76 or irreversible inelastic crushing (i.e., mechanical failure) of the propping asperities, and

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77 pressure solution that involves mineral dissolution at the contacting asperities, and/or
78 clogging of the fracture void by mineral precipitation (Yasuhara et al. 2004; Lang et al.
79 2016). When a fracture is subjected to elevated temperatures, thermal expansion can create
80 a thermal overclosure and enhanced pressure solution at the asperities (Yasuhara et al.,
81 2006; Peng et al., 2020). The effects of coupled Thermal-Hydrological-Mechanical-
82 Chemical (THMC) processes on fracture aperture and permeability have been examined by
83 a series of flow through experiments on artificially-fractured granite specimens at different
84 effective stresses and temperatures (Morrow et al., 2001; Polak et al., 2003; Yasuhara et al.,
85 2011; Kamali-Asl et al. 2018; Shu et al., 2020). Despite the extensive laboratory
86 investigations on permeability evolutions of single fracture at triaxial loading and
87 hydrothermal conditions, data is limited in granite with anisotropic textures and microcrack
88 distributions. As one of the “bottleneck” issues in repository design and performance
89 assessment (Tsang et al., 2005), the anisotropic fluid flow at different stress and thermal
90 conditions within the EDZ rock needs a systemic study.

91 When external loading is applied to rocks (including granitic and crystalline rocks),
92 pre-existing microcracks would propagate and coalesce along with creation of new ones,
93 resulting in ultimate failure (Bieniawski, 1967). The mechanical properties and
94 deformation behavior of rock containing pre-existing microcracks have been investigated
95 in a variety of experiments, including double torsion tests, uniaxial compression, triaxial
96 compression, and hydraulic fracturing tests. For example, Nara and Kaneko (2005) studied
97 the subcritical crack growth in andesite using the double torsion test and showed that crack
98 growth could be facilitated at higher water vapor pressures. Liu et al. (2022) systematically
99 reviewed the progressive failure process and mechanical properties of rock samples

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100 containing one, two and multiple flaws in uniaxial compression tests. The initiation and
101 propagation of new crack(s) were impacted by the inclination angle, length, width and
102 filling conditions of each pre-existing fracture(s). In triaxial loading tests, the presence of
103 confining stress typically increases the rock strength, and the failure pattern of rocks
104 gradually changes from axial tensile splitting to shear fracture. Ye and Ghassemi (2019)
105 examined the permeability enhancement through the propagation and coalescence of
106 preexisting fractures through triaxial-injection experiments under representative crustal
107 stress conditions on granite samples containing single or double preexisting
108 fracture(s)/flaw(s). Their results suggested that in addition to dilatant shear slip,
109 propagation and coalescence of preexisting fractures significantly contribute to granite
110 reservoir permeability creation during low injection pressure stimulation in fractured rocks.
111 The temperature impact on granite microstructure and strength can be more complicated.
112 The granite microstructure is commonly significantly changed after heating, which alters
113 both the physical and mechanical properties due to thermally-induced grain expansion.
114 Distinct thermal expansion of different mineral grains typically generates a nonuniform
115 distribution of thermal stress, which could change the microstructure and thus the
116 macroscopically-observed properties of granite in two aspects: (1) strengthening effect by
117 closing and healing of pre-existing microcracks, or (2) weakening effect through expanding
118 pre-existing microcracks and creating new ones (Wong et al., 2020). To the best of our
119 knowledge, it has not been well addressed in terms of the interactions between open
120 fractures and anisotropic microcracks under triaxial loading and hydrothermal conditions,
121 as well as the consequences in rock strengthening or weakening and permeability changes.

In this study, laboratory core-scale experiments were conducted in three anisotropic granite rock samples subject to triaxial loading and fluid flow at 25 and 90 °C. Although the temperatures used in the experiments are relatively low, they are expected to be applicable to the conditions in the rock surrounding a radioactive waste repository tunnel in many cases (Rutqvist, 2020). The samples were provided by the Grimsel Underground Research Laboratory (URL), Switzerland, featured by coarse mineral grains and a strong anisotropic texture and a large distribution of microcracks with a preferred orientation. Permeability evolution of the anisotropic samples under different axial and confining stresses was measured by applying transient differential pressures between the sample inlet and outlet, and monitoring the resulting changes in the water flow rates. During the experiment on each sample, effluent water samples were collected and the changes in the concentration of major aqueous chemical species were analyzed. Results from this study will help better understand the fluid flow and mechanical evolutions in anisotropic granites with open fractures and microcracks, when subject to triaxial loading and elevated temperatures.

2. Materials and methods

2.1 Rock core samples

Three granite core samples with a diameter of 3.80 cm and lengths ranging from 8.0 to 8.4 cm were used in this study. These samples were cut from a large slab obtained from the side wall drilling (with a diameter of 300 mm) of BK cavern in the Grimsel URL. The Grimsel Test Site is located in the central Swiss Alps and was established in 1983 by the National Cooperation for Radioactive Waste Disposal (NAGRA) for feasibility studies of the nuclear waste repositories in crystalline rock. It is located at around 450 m depth below

the eastern flank of the Juchlistock in the southern part of the Central Aare Massif (Keusen et al., 1989). A series of experiments has been carried out, ranging from site characterization-specific experiments to the engineered barrier system and finally to experiments designed to develop and test monitoring systems and study rock independent processes such as corrosion of canister materials (Schneeberger et al., 2019). The formation rock surrounding this cavern has been reported to be highly fractured, which hosts fault networks and water-conducting features critical for studying radionuclide migration and rock stability (Bossart and Mazuret, 1991). The rock is mainly composed of quartz, K-feldspar, plagioclase and biotite as reported by Selvadurai et al. (2019) in Table S1 of the supporting information (SI).

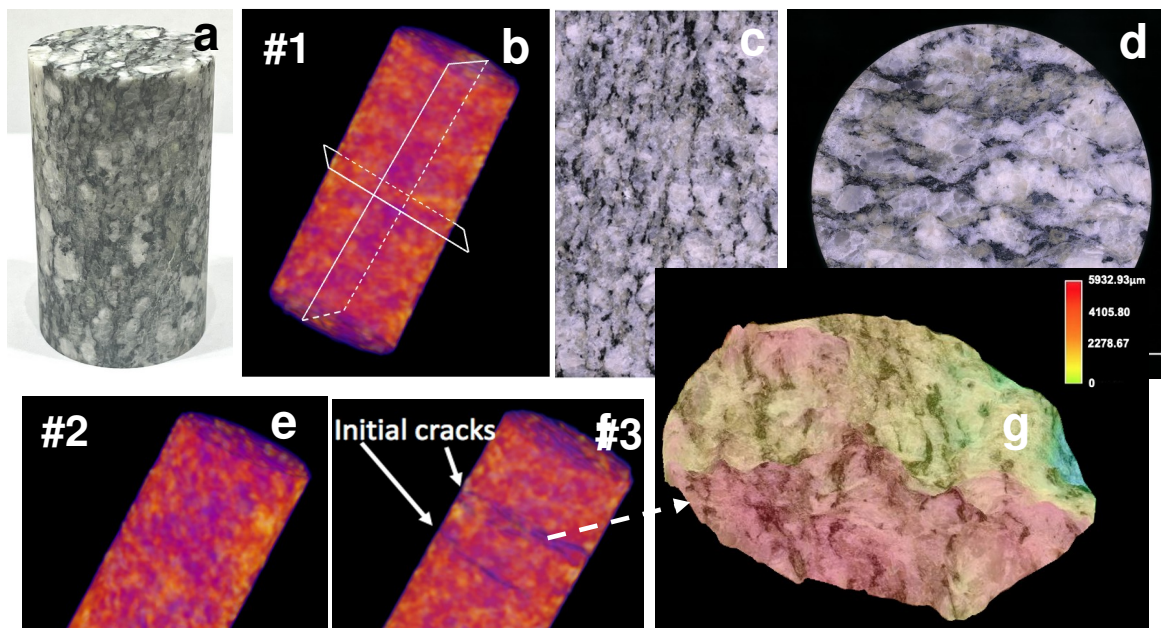
Figure 1a is a photograph of a representative core sample characterized by large grain sizes (up to ~1 cm) and foliations along the long-axis of the sample. Figures 1b-d are the 3-D X-ray CT images (obtained by a GE Lightspeed 16 medical X-ray CT scanner) of Sample #1, and the optical microscope images of the two cross-sections along the radial and long-axial directions (marked by the white boxes in Figure 1b), revealing the anisotropic textures. Figure 1e is the 3-D X-ray CT images for Samples #2. The porosity of Sample #1 and #2 were measured gravimetrically (Lin et al., 2015) using a vacuum pump, from the weights of the same samples that were first oven-dried at 90 °C and then saturated by deionized (DI) water. The samples were saturated in water by applying -101 kPa of vacuum to purge the trapped air out of the pores, over 6 hours until the water saturated mass exhibited no changes. Based on the mass changes, the porosity for Sample #1 and #2 was determined to be 0.0057 and 0.0055, respectively. The absolute water permeability of each water-saturated sample was measured by using the system shown in Figure 2. At room

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temperature, a constant confining stress (5 MPa) was applied, then water was injected at a range of flow rates from 0.8 to 5×10^{-3} mL/min. During water injection, the confining stress was selected as a common effective confining pressure encountered in EDZ rocks at Grimsel URL (David et al., 2018a). No axial stress was applied to the sample, except for the small reaction forces from the end plugs where the displacement was restricted during the test. While the outlet was open to the atmosphere, the inlet pressure was then closely monitored, ranging from 1 to 4.6 MPa. The average pore pressure over the core thus ranged from 0.5 to 2.3 MPa, 50% less than the applied confining pressure. No considerable impacts of pore pressure on permeability measurement are expected. Once the steady state was reached, the pressure difference between the inlet and outlet (ΔP) and water flow rate (Q) was used to determine the absolute water permeability (k) of the sample for the applied flow rate, according to the Darcy's law:

$$Q = \frac{kA}{\mu L} \Delta P \quad \text{Eq. (2)}$$

Where μ is water viscosity and L refers to the length of the core sample. For samples #1 and #2, the average permeability measured from different water injection rates was 1.45 and 1.27 μD , respectively.



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Figure 1. (a) Photograph of a representative granite sample provided by the Grimsel Underground Research Laboratory. (b) X-ray CT image and (c-d) cross-sectional microscope image of Sample #1. (e) X-ray CT image of Sample #2. (f) X-ray CT image of pre-cracked Sample #3. (g) Microscope image of the fracture surface geometry. The white boxes in (b) bound the axial and radial cross-section image in (c) and (d). The color bar in (g) represents the fracture surface roughness.

To better quantify the rock anisotropy, ultrasonic P-wave velocities were measured for the three samples under atmospheric conditions by using a custom apparatus. The measurement was conducted using a pair of 1-MHz P-wave transducers (Panametrics) by transmitting pulses through the core samples. Table 1 shows the results for the three core samples along the Z (the long axis), X (the radial direction that is parallel to the foliation) and Y (the radial direction that is perpendicular to the foliation) directions, under dry and water-saturated conditions (same to the conditions for measuring absolute water permeability). Velocities measured along the X axis were higher than those measured along the Y axis under oven-dried conditions, reflecting the effect of the texture anisotropy. Once saturated with water, the largest velocity increase was observed along the Y direction for Sample #1 and #2, indicating the presence of microcracks along the foliations and perhaps

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the large grain boundaries.

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Table 1. P-wave velocity (m/s) measured for the granite core samples prior to the loading tests

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*Note the P-wave velocity measurements was on Sample #3 conducted without the fractures.

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For Sample #3, an initial application of the radial confining stress up to 5MPa without restricting the axial deformation created several core-perpendicular tensile fractures, with

	Sample #1			Sample #2			Sample #3		
Diameter (cm)	3.81								
Length (cm)	8.0			8.3			8.4		
Porosity	0.0057			0.0055			0.0059		
Intrinsic water permeability (μD)	1.45			1.27			1.49		
P-wave velocity* (m/s)									
Direction	Dry	Water-saturated	Velocity increase	Dry	Water-saturated	Velocity increase	Dry	Water-saturated	Velocity increase
Z (long axis)	4145	5838	41%	3964	5327	34%	3858	5611	45%
X (parallel to foliation)	4363	5784	33%	4232	5754	36%	4459	5916	33%
Y (orthogonal to foliation)	3094	5554	80%	3285	5474	67%	3292	5575	69%

one fracture going through the sample. During excavation of tunnels for high-level radioactive waste disposal, creation of a free surface can lead to spalling failure (or, borehole breakout) involving tensile fractures in EDZ rocks. Thus investigating the sample and assessing the impact of such fractures on the mechanical strength and the evolution of the core-parallel permeability could help better understand the behavior of fractured granite at the repository conditions. This is because, Figure 1f shows the X-ray CT image of the sample with two induced fractures, while Figure 1g presents an image of the open fracture

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surface obtained using a digital stereo microscope (Keyence VHX-6000). The pre-cracked sample was reassembled and jacked in a Viton rubber sleeve, and the permeability measured with axial restriction was 1.49 μD , similar to Samples #1 and #2 because both fractures in Sample #3 are normal to the flow direction. Table 1 also lists the P-wave velocity and porosity measured for Sample #3 before inducing the tensile fractures, indicating similar anisotropic microcracks along the foliations.

Before the experiment, the aperture of the open fracture in Sample #3 was quantified based on the X-ray CT image and the calibration-free missing attenuation (*CFMA*) method proposed by Huo et al. (2016):

$$d = \frac{R \cdot \sum_{i=1}^N (CT_{mat} - CT_i)}{CT_{mat} - CT_{air}} \quad \text{Eq. (2)}$$

where d is the aperture size (μm), R is the resolution of the CT image ($=187.5 \mu\text{m}$), CT_{mat} refers to the CT number of surrounding rock matrix, CT_i is the CT number of fracture voxels, and $CT_{air}(= -1000)$ is the CT number of air. To determine the aperture, Sample #3 was reassembled and jacketed in a Viton rubber sleeve and CT scanned. On a CT image, we draw a line perpendicular to the fracture and read the CT_i number of each voxel along the line. Based on Eq. (2), the measured fracture apertures range from 0 (fully contact of asperities) to 330 μm . The aperture of the second fracture that propagates half way through the sample is expected to fall in this range as well.

2.2 Experimental setup and procedures

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Figure 2 shows schematic of the custom triaxial loading apparatus that allows long-term (days to months) experiments on rock core samples (designed for samples with a diameter 3.81 cm and a length up to 10 cm) under controlled temperature, mechanical, and chemical conditions. The apparatus is composed of two independent loading systems, allowing conducting experiments simultaneously on two samples. Each loading system enables a maximum axial stress of 200 MPa provided by a hydraulic ram (jack) and a confining stress of 70 MPa. The pressure vessels were made of titanium (grade 23) for its low weight and the corrosion resistance in addition to the high strength. Heat was applied to the rock sample and the vessel using two mica heating bands, up to the maximum designed

temperature of 200 °C. Three high-performance dual-cylinder syringe pumps (Vindum Engineering, CA) were used to control the radial confining pressure (Pump #1 using deionized water) and the axial stress (Pump #2 using hydraulic oil) and the injection flow rate and pressure (Pump #3). Each pump enabled fluid flow with a pressure up to 83 MPa, and a flow rate ranging from 0.0001 ml/min up to 29 ml/min. A back-pressure regulator, operated by compressed nitrogen gas, controlled the downstream pore pressure at the outlet of the sample. The actual outlet pressure was monitored independently via a pressure sensor, to determine the accurate pressure drop of the flow through the sample during a permeability measurement.

Before each experiment, a sample was first wrapped in a Viton sleeve with a tight tolerance for applying confining pressure. This sleeve was then inserted into a detachable sample pedestal with a tapered end, which was screwed onto the vessel's bottom end plug. Subsequently, the vessel hull was bolted onto the bottom end plug with eight high-strength steel bolts. The top end plug was then bolted to the vessel hull, again using eight steel bolts. This plug had an attached hollow cylinder with a tapered edge that was inserted into the end of the Viton sleeve. The contacts between the Viton sleeve and the top and bottom end plugs formed internal hydraulic seals. The hollow cylinder in the top end plug allowed insertion of a piston plug that was in direct contact with the rock sample. The surface near the rock-plug contact is in direct contact with the Viton sleeve, again forming a hydraulic seal. The assembled vessel was then emplaced in the loading frame on top of the hydraulic ram. The elevated temperature of the vessel and the rock sample was isolated from the exterior environment by using a fiberglass insulation and a cooling bath circulating cold water through aluminum cooling plates located at the top and the bottom of each vessel. A

short titanium cylinder was also used as a thermal isolation block to reduce heat dissipation from the vessel.

During the experiment, a constant axial stress of 14 MPa and a starting confining stress of 5 MPa were applied to Sample #1. These stresses are similar to the field conditions reported by Pahl et al. (1989) and David et al. (2018a, 2018b). Brine was then injected into the sample at a constant flow rate of 0.002 mL/min. The change paths in confining stress, pore pressure and temperature are schematically shown in Figure 3 (a) for Sample #1. Sample #2 and #3 were conducted simultaneously by applying a constant confining stress of 5 MPa and a constant brine injection rate of 0.0015 mL/min. The axial stress was increased step wisely from 14 MPa to 102 MPa, followed by pore pressure and temperature increases (Figure 3b). Temperature of the rock sample was closely monitored by a thermocouple emplaced in the confining fluid. For sample #1, the temperature was kept constant at 90 °C for 20 days before changing the stress conditions, while the stress conditions were maintained constant after heating for Sample #2 and #3 unless failure of the sample was observed (see Section 3.3). The average axial displacement of all samples was measured by a high-temperature LVDT mounted on the piston plug, with error at $\pm 0.05\%$.

Table S2 of SI shows the geochemical composition of the injected brine, which is a synthetic solution based on the chemical composition of Grimsel groundwater (ENRESA, 2000). During brine injection, the inlet and outlet fluid pressures as a function of the confining pressure under a selected temperature (25 or 90 °C) were closely monitored for calculating the changes in the sample permeability. Effluent brine was collected periodically, acidified by 2% volumetric fraction of HNO₃ for detailed dissolved elemental

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analysis by Agilent 8900 QQQ ICP-MS as a function of time, loading conditions and temperature.

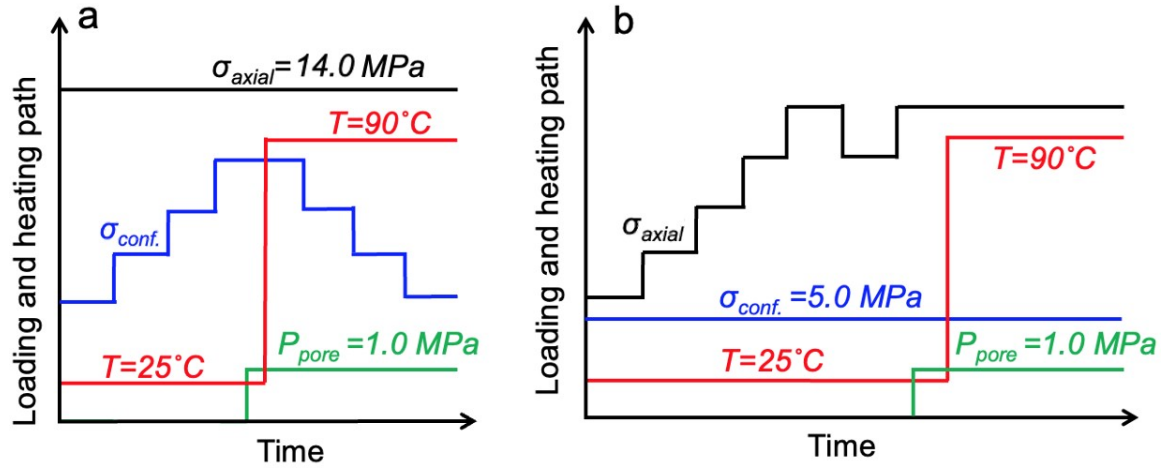


Figure 3. Loading and heating path for (a) Sample #1, (b) Sample #2 and #3.

3. Results

The permeability evolution of Sample #1 as a function of increasing confining stress and pore pressure at two temperatures (25 and 90 °C) is presented in Section 3.1. In Section 3.2, permeability evolution in Sample #2 resulting from increasing axial stress, pore pressure and temperature is investigated with a constant confining stress. At the same conditions to Sample #2, Section 3.3 describes the time-dependent permeability evolution and fast failure of the pre-cracked Sample #3.

3.1 Steady-state permeability vs. radial compaction, pore pressure and temperature

The relative changes of the steady-state permeability, k/k_i , at different loading and heating conditions in Sample #1, are shown in Figure 4a, where k_i is the intrinsic

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permeability (1.45 μD) in Table 1. The viscosity of deionized water was used for calculating k and k_i . Viscosity changes at elevated temperatures were considered when calculating k . Considerable impacts of salinity in the injected brine on viscosity is not expected from Kwak et al. (2005). The loading and heating path for Sample #1 can be described in 3 Stages: In Stage 1, when the axial stress was increased to 14.0 MPa (9 MPa differential stress), k/k_i decreased slightly to 0.95. k/k_i continued to decrease from 0.95 to 0.55 as the differential stress decreased from 9 to 4 MPa. In Stage 2, adding a 1.0 MPa pore pressure resulted in a slight increase of k/k_i from 0.55 to 0.57.

In Stage 3, the triaxial stress and pore pressure were maintained constant while the temperature was increased to 90 °C. This led to a dramatic reduction of k/k_i to 0.14 that was only 25% of the permeability measured at 25 °C (Stage 2). Also note when the differential stress was increased back to 6 and 9 MPa, the permeabilities at 90 °C were 24% and 22% of those measured at 25 °C under the same stress conditions.

Figure 4b presents the transient permeability evolution with temperature increase from 25 to 90 °C. During the first 4 hours of heating, k/k_i decreased quickly from 0.55 to 0.22, followed by a slow decrease to 0.14 for over 500 hours. During this transition, the axial and confining stresses as well as the pore pressure were held constant. Effluent brine was collected periodically, acidified by 2% volumetric fraction of HNO_3 for dissolved cations analysis using the standard Inductively Coupled Plasmas–Mass Spectrometry (ICP-MS) protocols. More detailed discussion is given in Section 4.3 on the effluent water chemistry analysis.

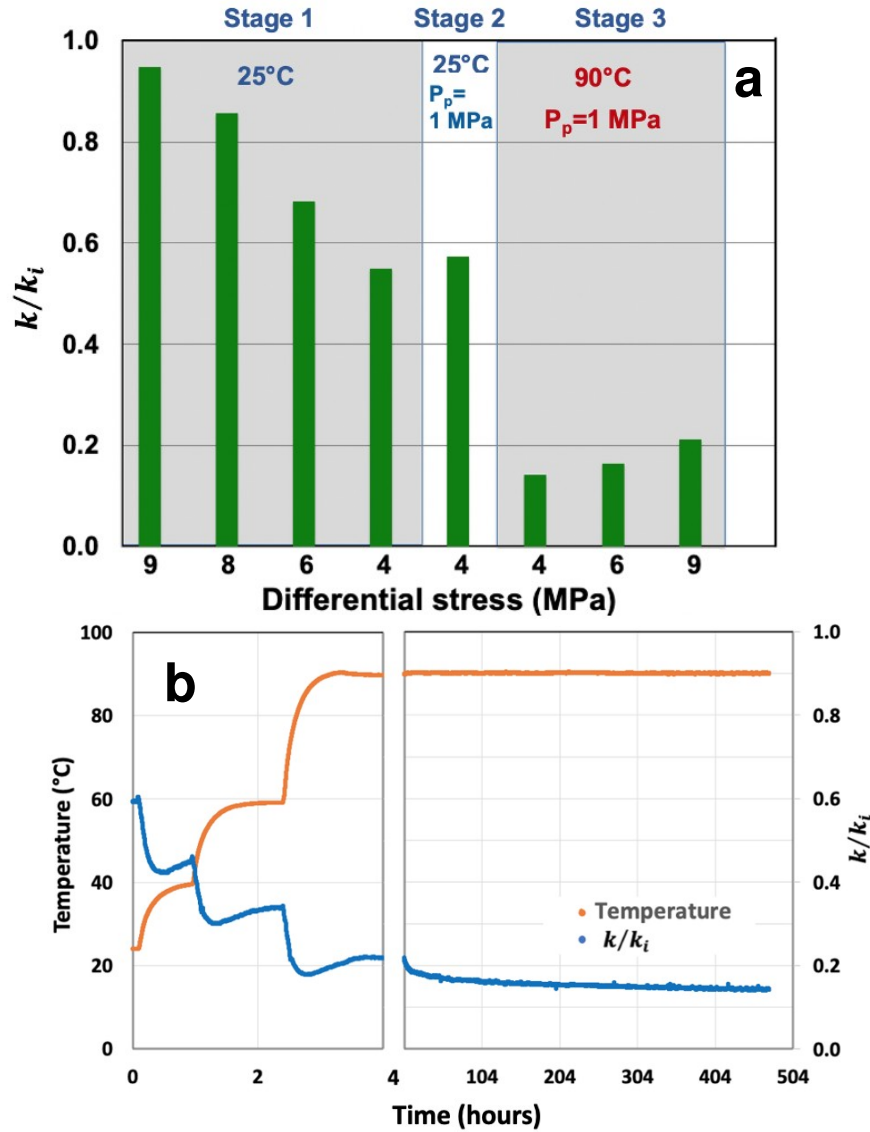


Figure 4. (a) The variation of steady-state permeability (k/k_i) as a function of applied differential stress and (b) dynamic changes of k/k_i vs. time after heating in Sample #1. During the experiment, the confining stress is step-wisely increased while the axial stress was maintained constant at 14 MPa.

3.2 Steady-state permeability vs. axial compaction, pore pressure and temperature

Different from Sample #1, Samples #2 and #3 were subject to constant 5 MPa confining stress and the axial stress increased by steps starting from 14 MPa to 102 MPa (Figure 3).

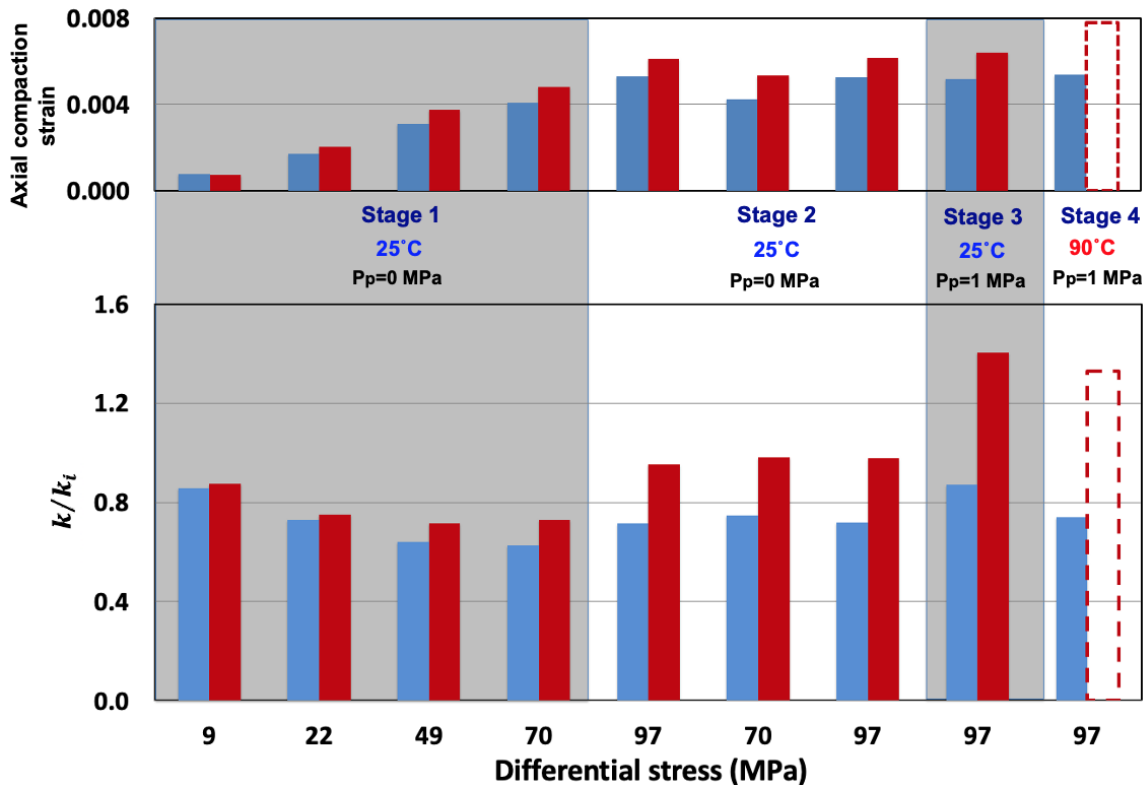
Figure 5 depicts the variations of steady-state k/k_i and axial compaction strain for Sample #2 (marked by blue column) in different stages of applied differential stress, pore pressure and temperatures.

Stage 1: Axial compaction. The steady-state permeability measured at 9 MPa differential stress was 1.1 μD and the corresponding $k/k_i = 0.86$. The axial compaction strain was 0.08%. Increasing differential stress up to 70 MPa resulted in continuous decreases of k/k_i from 0.86 to 0.63, meanwhile the axial strain decreased from 0.08% to 0.41%.

Stage 2: Cyclic loading. Further increasing the differential stress to 97 MPa induced compaction of the sample with a strain at 0.53%. Different from Stage 1, however, the permeability showed increase from $k/k_i = 0.63$ to 0.71. To better understand this permeability change by axial compaction, the axial loading was cyclically applied. As the differential stress was decreased to 70 MPa, axial compaction reduced accordingly to 0.42%, whereas permeability increased slightly to $k/k_i = 0.75$. Note that under the same differential stress, k/k_i during Stage 1 was smaller at 0.63, indicating potential opening of existing and/or developing new microcracks when a higher 97 MPa differential stress was applied in Stage 2. As the differential stress was recovered to 97 MPa, the compaction strain recovered elastically to 0.52%, and k/k_i recovered to 0.72. The cyclic loading at this stage indicated the potential development of microcracks that contributes to the permeability increase.

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379 **Stage 3: Increased pore pressure.** In Stage 3, the axial and confining stresses were kept
380 constant, and a 1 MPa pore pressure was applied by the injected water and controlled by a
381 back-pressure regulator. As shown in Figure 4, for Sample #2 (the blue column), the axial
382 strain was unchanged at 0.52%, while the permeability increased by 21% up to $k/k_i = 0.87$.
383 Such permeability increases in granite by increasing pore pressure (thus decreasing
384 effective stress) can be attributed to further opening of intrinsic and/or creating new
385 microcracks after Stage 2 (Liu et al., 2016).



386 **Figure 5.** Steady-state permeability evolution and axial strain changes for Sample #2 (blue
387 column) and #3 (red column) during triaxial loading and heating tests. The red dotted
388 column represents the transient axial compaction strain and k/k_i right after temperature was
389 increased to 90 °C.

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392 **Stage 4: Increased temperature.** In the final Stage of the experiment, the temperature
393 was increased to 90 °C over 3.5 hours while the axial and confining stresses and pore
394 pressure were kept constant. For Sample #2, steady-state permeability, k/k_i was reduced
395 from 0.87 at 25 °C to 0.74 at 90 °C (Figure 5) , which was accompanied by slight axial
396 compactions from 0.52% to 0.54% in strain. Once these initial changes occurred, the
397 permeability and axial strain remained constant for over 1 month.

398 **Impacts of pre-existing fractures.** Figure 5 also presents the steady-state permeability
399 changes in Sample #3 (marked by red columns) subject to the same loading, heating and
400 fluid flow conditions as Sample #2. In Stage 1, the steady-state k/k_i and compaction strain
401 at 9 MPa differential stress in Stage 1 were measured at 0.88 and 0.07%, respectively,
402 similar to those in Sample #2. However, when differential stress further increases and axial
403 compaction and permeability continuously decrease, the pre-cracked Sample #3 showed
404 larger compaction displacements and less permeability reductions comparing with Sample
405 #2. For instance, when the differential stress increased to 70 MPa, k/k_i and compaction
406 strain of Sample #3 was 0.73 and 0.48%, respectively, compared to 0.63 and 0.41% for
407 Sample #2. The pre-existing fractures perpendicular to the maximum principal stress
408 (compression) direction may contribute to the larger compaction in Sample #3. However,
409 they have less impacts on permeability as the two cracks are perpendicular to water flow
410 direction.

411 In Stage 2, the increase in the differential stress from 70 to 97 MPa resulted in the
412 increase of k/k_i in Sample #3 from 0.73 to 0.95 and the changes in the axial compaction
413 strain from 0.48% to 0.61%. Compared to Sample #2, the presence of horizontal fractures
414 continued to contribute to the larger axial compaction strain in Sample #3. The more

pronounced permeability increase may indicate more shear-induced sliding along microcrack surfaces. The cyclic axial loading between 70 to 97 MPa had very limited impacts on permeability variations (k/k_i varied from 0.95 to 0.98) in Sample #3, and, similar to Sample #2, the axial strain is mostly attributed to elastic deformation.

In Stage 3, Increasing pore pressure led to more considerable permeability increases in Sample #3 from $k/k_i = 0.78$ to 1.40, which then stayed constant for 12 days. The axial compaction strain changed slightly from 0.62% to 0.64%. The pore pressure further induced opening of existing and/or new microcracks after Stage 2. The cracks initiated at the open fracture asperities have the potential to grow sub-critically and coalesce with pre-existing microcracks in the background, eventually leading to catastrophic rock failure. In Section 3.3, we present an evolution of permeability over time and fast failure of Sample #3 after heating at 90 °C, which may reflect such a process.

3.3 Dynamic permeability evolution and fast failure of fractured granite

In Stage 4 of the experiment on Sample #3, the rise of temperature to 90 °C within 2.5 hours was accompanied by decrease in k/k_i from 1.40 to 1.33 (dash-line column in Figure 5). Figure 6 further presents the very dynamic changes of permeability (red triangles) and axial compaction strain (blue squares) vs. time while the temperature was maintained at 90 °C, which can be categorized in multiple phases.

In Phase 1 (0-49.7 hours), k/k_i first increased relatively slowly from 1.33 to 6.27 during the first 42.5 hours, followed by fast, large increases from 6.27 to 100 during 42.5-49 hours. During the latter period, k/k_i exhibited occasional spikes at 44, 48 and 48.6 hours, indicating generation of new microcracks accompanied by quick water redistribution and pressure equilibration. The strain during the first 49 hours, however, remained constant at

0.079%. During 49-49.7 hours, extremely large k/k_i variations over almost 5 orders of magnitudes from 1 to 10,000 were observed (see the insert in Figure 6a), concurrently with axial compaction strain increases from 0.79% to 0.90% and pore pressure drops to 0 MPa (atmospheric pressure) in the sample. It can be inferred that significant opening cracks developed during this period of time to the extent that the 1 MPa pore pressure could not be maintained because of the large permeability of the sample. In the subsequent experiment, the back pressure regulator was removed so that the pore pressure was reduced to 0 MPa. The confining and axial stresses, and water injection rate, were kept unchanged.

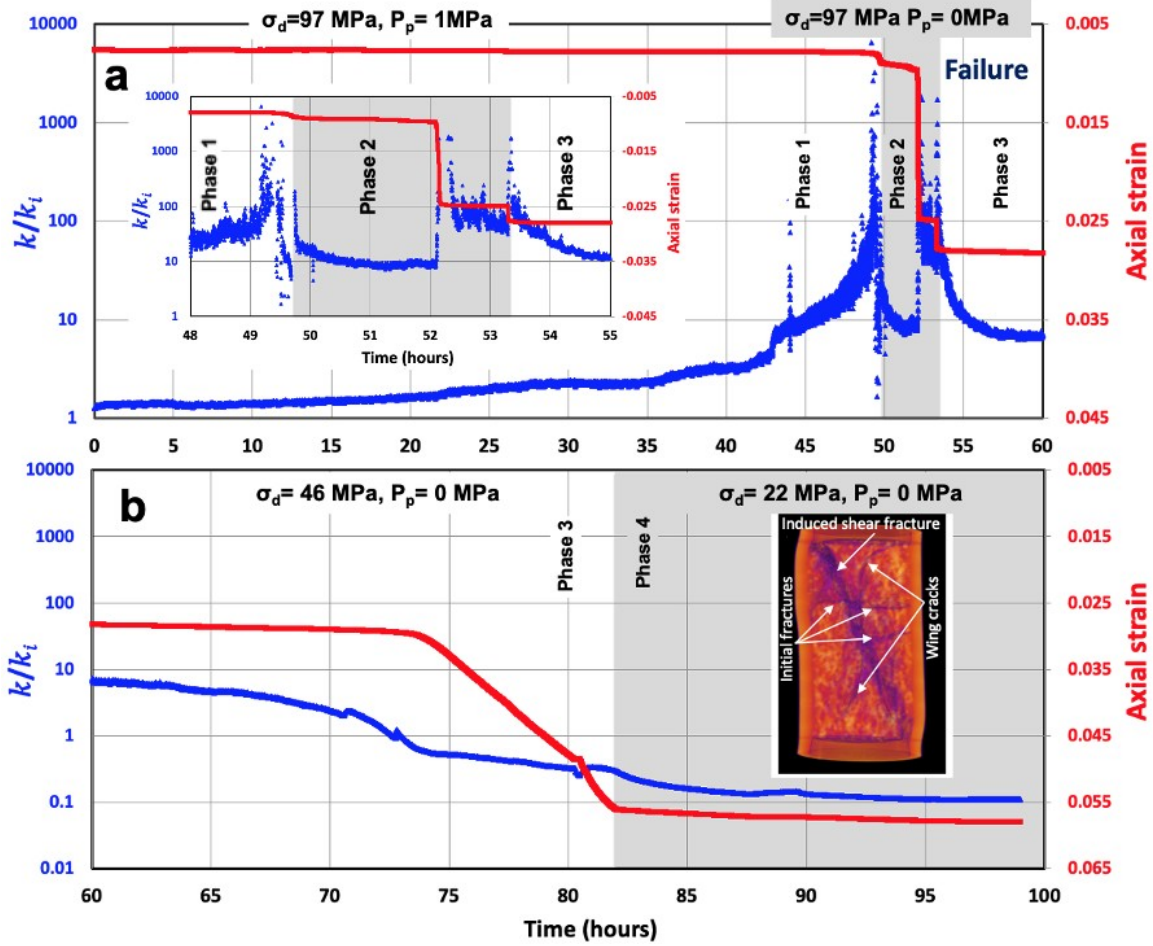
In Phase 2 (49.7-53.3 hours), k/k_i and axial strain were relatively constant at 10 and 0.92%, indicating the minimal shear sliding and temporal closure of cracks driven by the confining stress. This short-term stability was followed by strong k/k_i variations and more pronounced shear deformations with the changes in axial compaction strain from 0.96% to 2.5% at 52.1 hours. From 52.1 to 53.3 hours, the axial strain remained constant along with relatively stable k/k_i . At the end of this phase (53.3 hours), another spike of k/k_i and axial strain change from 2.5% to 2.7% occurred. These periodic variations in strain and permeability suggested frictional shear sliding that required time-dependent breakage of asperities at the fracture surfaces, accompanying sequential growth and arrest of microcracks (Sano, 1981, Brantut et al., 2013). Compared to Sample #2, it seems the horizontal fractures considerably accelerated the subcritical crack growth and creep failure in Sample #3 tested at 90 °C.

In Phase 3 (53.3-82 hours), the differential stress was reduced to 46 MPa, which slowed the compaction axial strain rate between 53.6 to 73 hours. Between 73 to 81.5 hours, however, another large axial strain change from 2.97% to 5.61% occurred. The reduced

461 differential stress did not stop the axial compaction and shear sliding, indicating the
462 development of pervasive shear fractures and complete failure of the sample. In contrast,
463 k/k_i decreased continuously from 100 to 0.3, without any spike or noticeable transit
464 variation, despite the shear sliding. In Phase 4 (82-100 hours), the differential stress was
465 reduced further to 22 MPa, resulting in relatively stable k/k_i and the axial strain. At the end
466 of the experiment, k/k_i and the axial strain were 0.11 and 7.5%, respectively.

467 After the experiment, Sample #3 was recovered while still secured in a viton jacket for
468 X-ray CT imaging. The insert image in Figure 6b shows clearly an oblique shear fracture
469 and two wing cracks induced by shear deformations, which coalesced with two pre-existing
470 horizontal cracks. Heating of the sample under axial compaction may have caused
471 thermally induced stress concentrations at the contacting asperities of the horizontal open
472 fracture (see Figure 1g), which resulted in sub-critically driven crack initiation, growth, and
473 coalescence with pre-existing microcracks in the sample. The propagation and coalescence
474 of the microcracks eventually led to a large shear fracture penetrating through the
475 horizontal fracture under triaxial loading (see the insert CT image in Figure 6b). Also note
476 that there are the crushed debris and fine powders on the shear fracture surface (Figure S1 in
477 SI), resulting from the shear sliding and failure of contacting asperities. During Phases 3
478 and 4, these debris and fine particles may have migrated by the water flow and clogged the
479 apparatus pipeline, which resulted in k/k_i smaller than 1 (i.e. the permeability became
480 smaller than the initial intrinsic state) in Figure 6b.

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483 **Figure 6.** Dynamic permeability evolution (red) and axial strain (blue) changes for the pre-
484 cracked Sample #3 under triaxial loading and heating at 90 °C for periods of (a) 0-60 hours,
485 and (b) 60-100 hours. Note: σ_d represents applied differential stress, P_p is pore pressure.

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487 4. Discussion

488 4.1 Thermally induced microcrack closure by radial compaction

489 Results from Sample #1 showed larger (75%) irreversible permeability reductions at 90
490 °C when microcracks in the sample was subject to compaction at 10 MPa confining stress,
491 when compared to the 15% permeability reduction in Sample #2 at 90 °C when

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492 microcracks were dominated by shear sliding at a lower 5 MPa confining stress. While
493 laboratory data on microcracks is limited, permeability reduction and closure of a single
494 open fracture at constant confining stress by elevated temperatures has been reported. For
495 instance, Lima et al. (2019) and Peng et al. (2020) presented fast fracture aperture reduction
496 in granite samples when temperature was increased to 90 °C, similar to that in Figure 3b.
497 Shu et al. (2020) concluded that at a high constant confining pressure (e.g. greater than 10
498 MPa), the fracture hydraulic aperture and permeability decrease rapidly with increase in
499 temperature, due to a decrease in either elastic modulus or bond strength of the mineral
500 grains, pressure dissolution at contacting asperities, or a combination of these effects. At
501 low constant confining pressures (e.g. 5 MPa), as the temperature increases, the hydraulic
502 aperture and permeability can eventually be increased by free-face dissolution at the
503 fracture faces while the impacts of pressure dissolution may be ignored. In Sample #2, we
504 did observe such permeability increase at 5 MPa confining stress and elevated temperature.
505 Different from a large open fracture, the limited microcrack surfaces and larger flow
506 tortuosity in Sample #2 may hinder for such dissolution. Instead, the pressure dissolution at
507 some asperities could potentially lead to slight permeability reduction and microcrack
508 closure after heating, as shown in Figure 4.

509 **4.2 Thermally induced fast failure of granite with fractures and microcracks**

510 Extensive laboratory studies have shown obvious rock strength reduction in granite
511 occurs at about 300–400 °C and around 600 °C at which the mechanical behavior of rock
512 transforms from brittle to ductile along with a significant drop of strength. However, very
513 few detailed attempts have been made to elucidate the temperature influence on rock
514 strength in the lower temperature range between room temperature and 200 °C.

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515 Controversy remains in this mild temperature regime. Chen et al. (2012) and Singh et al.
516 (2015) suggested that rock strength monotonically decreases as the temperature increases.
517 Gautam et al. (2018), Yin et al. (2016), and Zhang et al. (2017) observed that there is little
518 variation of rock strength below a certain temperature. Conversely, Ferrero and Marini
519 (2001), Gautam et al. (2018) and Kumari et al. (2017) reported the opposite phenomenon of
520 strength enhancement in the mild temperature regime.

521 It is widely accepted that brittle creep of granite is controlled by the growth of cracks
522 and eventually results in macroscopic faulting of the rock, and there exists a critical level of
523 microcrack density (damage) that can be sustained by a rock. Any further increase in
524 damage above the critical state will result in strain acceleration and rock failure. Based
525 upon this interpretation, the pre-existing microcracks in Sample #2 are expected to be less
526 than this critical level, because the rock did not fail during the experiment at 90 °C for over
527 1 month. In contrast, the presence of the horizontal rough fractures accelerated subcritical
528 crack growth and resulted in the fast rock failure at 90 °C within 60 hours. The higher stress
529 concentrations at the contacting fracture asperities, under axial compaction, can lead to new
530 microcrack generation, growth and coalescence with existing ones. Elevated temperature
531 may further contribute to this and accelerated rock failure by thermal cracking induced by
532 non-uniform thermal expansion of different minerals. For instance, Du et al. (2022)
533 reported ~2.5 times higher in thermal expansion of quartz than feldspar at 90 °C. In
534 addition, rock natural microcracks were formed at elevated temperatures (Barton 2007).
535 When these microcracks cool down, some mismatch in microscale across is expected to
536 occur, due to the different thermal expansion coefficients of their constitutive minerals.
537 When subject to elevated temperature, natural fractures should undergo heterogeneous

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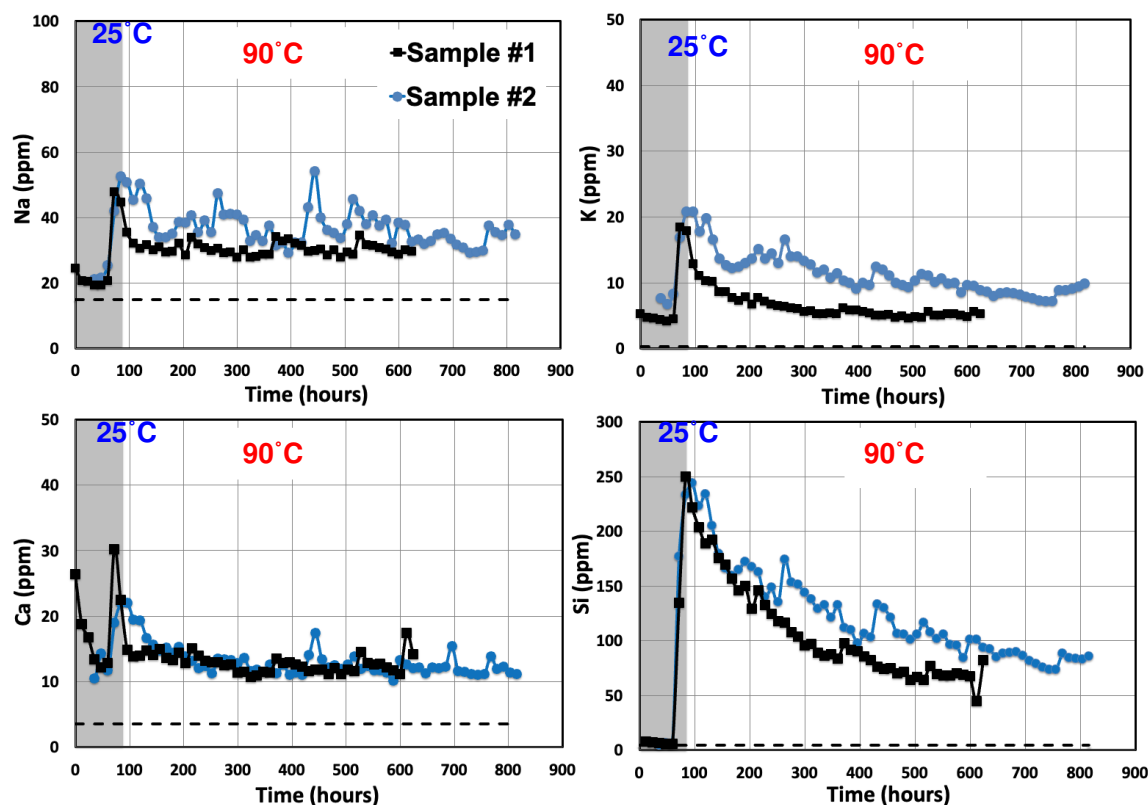
538 thermal dilation that promotes overclosure and new cracks development (Barton 2007).
539 Reduced fracture toughness by the chemical reaction with water may also contribute to the
540 accelerated rock failure.

541 **4.3 Effluent water chemistry**

542 During the experiments, effluent water samples were collected for major dissolved cations
543 from the three granite samples. Figure 7 shows their variations as a function of time in
544 Sample #1 and #2 collected at 12-hour intervals. The black dash lines represent the
545 concentration of each cation in the injected water before experiments. As shown in the
546 figure, heating of both samples up to 90 °C resulted in rapid increase of Na, K, Si in effluent
547 water samples from dissolution of K-feldspar and plagioclase, followed by gradual
548 decreases until the end of the experiments. The initial increase in cation concentrations in
549 both samples can be attributed to the fast dissolution of fine-grained minerals enhanced by
550 the elevated temperature. The faster concentration decrease in Sample #1 may be attributed
551 to the higher confining stress applied, and higher water flow rate (thus less residence time
552 of water in the sample) in Sample #1 (0.002 mL/min) than that in Sample #2 (0.0015
553 mL/min). It is also noted a higher Ca concentration from Sample #2 at 25 °C, which might
554 be attributed to higher plagioclase dissolution when the sample was subject to a higher
555 confining stress and pressure solution was more considerable. The average Na, K, Ca and
556 Si concentrations by the end of the experiments for Sample #1 and #2 were 30.33, 5.12,
557 12.93 and 67.73 (ppm) and 33.70, 8.61, 12.01 and 85.15 (ppm), respectively.

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563 **Figure 7.** The variations of major cations vs. time in the effluent water samples for
564 experiments on Sample #1 and #2. The black dash lines represent the concentration of each
565 cation in the injected water.

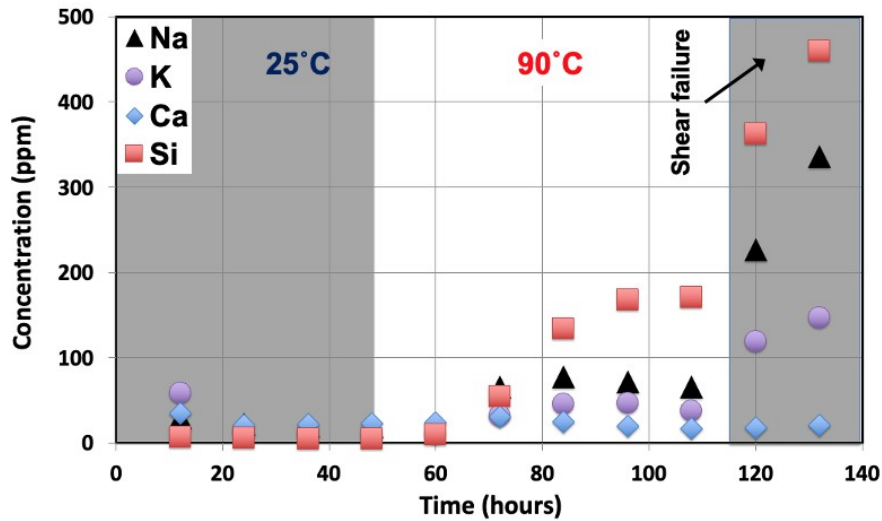
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567 Figure 8 shows the temporal variations of major cations in the effluent water for Sample
568 #3. Different from Samples #1 and #2, once heated up to 90 °C, the overall concentrations
569 of Si, Na and K in the effluent water samples for sample #3 continuously increased with
570 time. The concentrations of Si, Na and K increased even more pronouncedly after shear
571 failure of the sample in Phase 3 was observed, resulting from the growth of existing and/or
572 newly formed microcracks, which facilitated more mineral dissolution and subsequently
573 higher solute concentrations. New fractures and cracks after sample failure provided new
574 surfaces available for mineral dissolution, including the large surface area provided by the

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575 debris and mineral powders. Because the water used in the experiment is undersaturated
576 with respect to the minerals contained in the rock, these new surfaces and the crushed debris
577 from large shear sliding permit fast dissolution with longer time. In contrast, for Sample #1
578 and #2, the contribution of the direct dissolution from the pre-existing microcracks is
579 expected to be much smaller because of the overall smaller surface area. In Sample #3, the
580 relative importance of the pressure dissolution would be larger due to the newly created
581 fracture surfaces contacting fluid. Figure 8 also shows some attainment of relatively stable
582 Na, K and Si concentration between 85 to 108 hours, indicating potential steady-state
583 mineral dissolution at the constant flow rate, despite permeability during this period of time
584 increased continuously (see 0-50 hours in Figure 6a). The lower Ca concentration in the
585 effluent water might be attributed to the lower Ca abundance in the rock compared to Na, K
586 and Si (Selvadurai et al., 2019). It may also indicate the new microcracks, during the shear
587 failure of the sample, were developed most likely in biotite, K-feldspar and quartz and/or
588 their boundaries. It is also noted that the Si concentrations after shear failure of the sample
589 in Figure 8 are higher than a typical solubility of quartz reported by Morey et al. (1962).
590 This may be attributed to solid residuals (from crushed debris and fine powders of the failed
591 sample) in the effluent water and after acidified, further dissolution of these solids led to a
592 concentration higher than Si solubility under the experimental conditions.

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594 **Figure 8.** The variations of major cations vs. time in the effluent water samples for
 595 experiments on Sample #3.

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597 5. Conclusions and implications

598 The anisotropic nature of granite at Grimsel URL imposes great challenges in
 599 predicating EDZ rock behaviors and the long-term performance of a repository for
 600 radioactive waste. Results from the intact samples (Sample #1 and #2) showed small
 601 difference in permeability when samples were dominated by either compaction (Sample
 602 #1) or shearing of microcracks (Sample #2). Sample #1, however, experienced more
 603 considerable thermal impacts with large (75%) irreversible permeability reduction after
 604 temperature was raised to 90°C. By comparison, thermal impacts on permeability in
 605 Sample #2 is limited to 15% reduction at the same temperature. This observation indicates
 606 the importance of interactions between rock anisotropy, original microcracks and stress and
 607 thermal conditions in determining EDZ behaviors. In geologic repositories exhibiting
 608 similar rock anisotropy like that in the Grimsel URL, the swelling of hydrated EBS

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609 bentonite can impose additional compression stress (up to 5 MPa) to the surrounding EDZ
610 rock and when this stress is normal to microcracks (similar to Sample #1), elevated
611 temperature could lead to large permeability reduction that slows down radionuclide
612 transport.

613 Results from the test on Sample #3 exhibited the impacts of open fracture on the
614 degradation of rock mechanical strength at elevated temperature. After excavation, tensile
615 cracks may develop in EDZ rock at different orientations relative to the maximum principal
616 stress, i.e., the strata stress. The fast failure of Sample #3 with open fractures in less than 2
617 days at 90°C, even normal to the maximum principal stress, emphasizes that extreme
618 caution is needed in the presence of open fractures for tunnel security and repository
619 design. For the long-term performance and security of repository, tunnel geometry should
620 be designed to minimize stress concentrations in the tunnel walls, along with optimized
621 tunnel support considering pre-existing fractures. Continuous monitoring of the
622 surrounding rock mass during excavation is also critical to detect any signs of instability or
623 excessive fracturing. In multiple cavern systems, it is suggested to ensure adequate spacing
624 between caverns to prevent large-scale interconnected fracture networks developed in
625 EDZ. Results from the effluent water samples also imply that water chemistry could
626 potentially be an early warning signal of extensive subcritical crack growth and severe rock
627 failure. The observed transient steady-state behaviors in solute concentration profiles
628 followed by large increase after rock failure indicate further dissolution of minerals, which
629 may lead to either further opening or closure of fracture depending the stress state. Long-
630 term tests and further mineralogical characterizations are needed to better understand the

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631 chemical feedbacks in response to the mechanical behavior/failure of EDZ rocks, as well as
632 the permeability evolutions for radionuclide migration.

633 **Supporting Information (SI)**

634 More detailed information on the water composition and photograph of Sample #3 after the
635 experiment are provided in the SI.

636 **Conflicts of interest**

637 The authors declare no competing financial interest.

638

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