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## **Effects of chemical alteration on frictional properties in a deep, granitic, geothermal system in Cornwall: Direct shear experiments at near in-situ conditions**

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### **Key Points:**

- 22 • Large fault systems are targeted at depth as geothermal reservoirs in high-heat producing  
23 granites in Cornwall
- 24 • Direct shear experiments were conducted on a series of successively more naturally  
25 altered granites from a fault in Carnmenellis granite
- 26 • Alteration makes sliding more likely but also more stable, while higher temperatures  
27 destabilise shearing  
28

## 29 Abstract

30 Geochemical alteration of host rocks might affect the productivity and the potential for induced  
31 seismicity of geothermal systems. In addition to natural alteration, following production and heat  
32 extraction, re-injected fluids at lower temperatures and different pressures may be in chemical  
33 disequilibrium with the rock, impacting mineral solubility and dissolution / precipitation  
34 processes. In this study, we investigate the effect of geochemical alteration on the frictional  
35 behaviour of granites, and their seismogenic potential, by conducting direct shear experiments  
36 using samples with varying degrees of alteration. The samples originate from the Carnmenellis  
37 granite in Cornwall, SW England, and represent the formation used in the United Downs Deep  
38 Geothermal Power Project for heat extraction. Experiments were conducted on granite powders  
39 (referred to as gouges) at room temperature and 180°C, at simulated in-situ confining and pore  
40 pressures of 130 MPa and 50 MPa, respectively (~5 km depth). With increasing degree of  
41 alteration, the frictional strength of the gouges decreases while frictional stability increases. At  
42 high temperature, frictional stability is reduced for all samples while maintaining the trend with  
43 alteration stage. Microstructural investigation of the sheared gouges shows alteration delocalises  
44 shear by reducing grain size and increasing clay fraction, which promotes the formation of  
45 pervasive shear fabrics. Our work suggests that, within the range of tested pressures, more  
46 alteration of granite initially causes more stable shearing in a fault. This behaviour with  
47 alteration is sustained at high temperatures, but the overall frictional stability is reduced which  
48 increases the potential for induced seismicity at higher temperatures.

## 49 1 Introduction

50 Transitioning towards renewable and carbon neutral energy generation is one of the  
51 major tasks during the 21<sup>st</sup> century. In addition to wind or solar energy, harnessing the Earth's  
52 geothermal heat can help diversify national green energy portfolios, particularly as geothermal  
53 energy projects can deliver a continuous supply of heat and/or power. In the United Kingdom,  
54 one of the most promising areas for deep geothermal energy production is Cornwall, SW  
55 England [Busby and Terrington, 2017]. This region is underlain by the Cornubian batholith, with  
56 several large granitic plutons now revealed by erosion. Granites often contain higher  
57 concentrations of uranium, thorium and potassium relative to their host rocks [Abesser *et al.*,  
58 2020]. The decay of these elements causes enhanced heat flow which, in the case of Cornwall, is  
59 close to double the UK average [Busby and Terrington, 2017]. This makes the region a target for  
60 geothermal operations like the United Downs Deep Geothermal Power project (UDDGP; see  
61 Figure 1 and Figure 2) in the Carnmenellis pluton or the Eden Project in the St. Austell pluton  
62 [Ledingham *et al.*, 2019, and edengeothermal.com, 2022]. These projects target fractures and  
63 fault zones in crystalline rock that provide pre-existing pathways for the production of hot fluids  
64 (~180°C) and then the re-injection of cooler fluids following heat extraction [e.g. Ledingham *et*  
65 *al.*, 2019].

66 The sustainability and reliability of these systems depends on the stability of the fault  
67 transport properties throughout the lifetime of the system. After heat is extracted, fluids are re-  
68 injected into the subsurface at temperatures and pressures that may not be in equilibrium with the  
69 surrounding rock. Consequently, the fluids may not be in chemical equilibrium with the  
70 surrounding rock, which may promote geochemical reactions to occur [e.g. Kamila *et al.*, 2021;  
71 Stefansson, 1997]. Mechanical and/or geochemical rock property changes occurring over  
72 extended periods of time (i.e., years), can impact the system's productivity [e.g. Bischoff *et al.*,

73 2024; *Forbes Inskip et al.*, 2023]. Furthermore, given that fault frictional properties depend on  
74 mineralogical composition [e.g. *Fang et al.*, 2018a; *Ikari et al.*, 2011; *Summers and Byerlee*,  
75 1977; *Tembe et al.*, 2010], this might impact the risk for induced seismicity [*Scholz*, 1998].

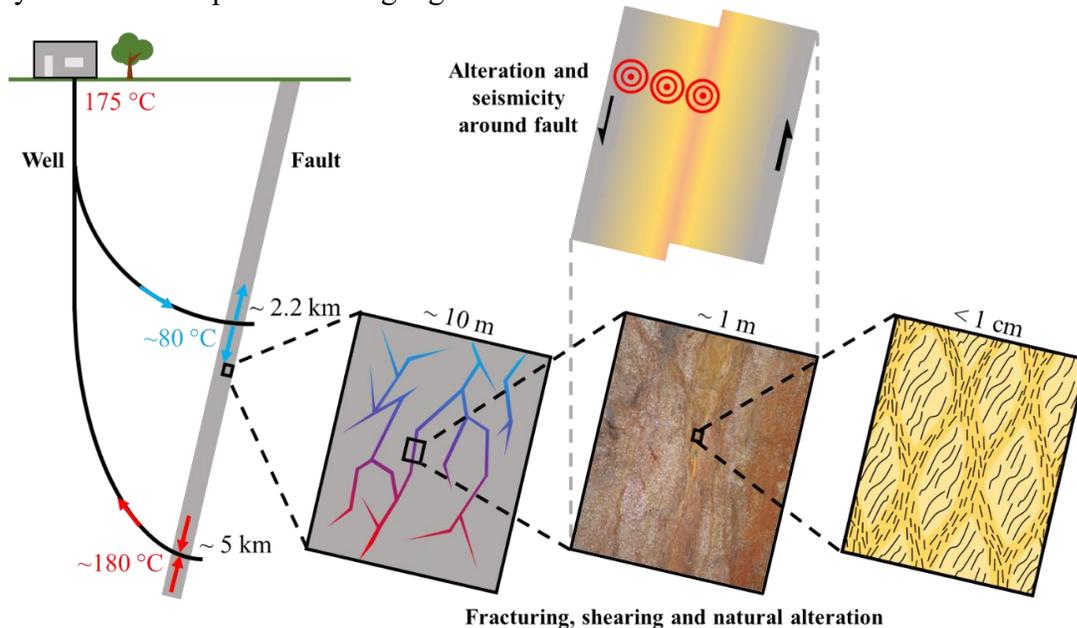
76 The frictional properties of granite are relatively well understood over a wide range of  
77 conditions [e.g. *Biegel et al.*, 1992; *Blanpied et al.*, 1995; *Byerlee*, 1967; *Hadizadeh et al.*, 2015;  
78 *Ishibashi et al.*, 2018; *Mitchell et al.*, 2016; *Wang and Scholz*, 1994]. However, in most previous  
79 studies unaltered granite was used. At room temperature, unaltered granite gouges were found to  
80 exhibit stable sliding under a wide range of conditions. These only transition into potentially  
81 unstable sliding at high sliding velocities under high effective pressures [*Kilgore et al.*, 1993] or  
82 at displacements of 10 to 100 mm in high displacement tests [*Beeler et al.*, 1996]. *Blanpied et al.*  
83 [1998] found that unaltered granite gouges transition from stable sliding to potentially unstable  
84 sliding between 90°C and 350°C at 400 Mpa effective pressure and 100 Mpa pore pressure. This  
85 temperature window is relevant to the UDDGP project which operates between ~180°C at the  
86 production well and ~80 °C at the injection well based on projected temperature gradients [*Busby*  
87 *and Terrington*, 2017; *Ledingham et al.*, 2019], but also applies to other geothermal systems  
88 which work at similar temperature ranges.

89 Natural fault systems, however, often show geochemical alteration, usually becoming  
90 more distinctive with increasing proximity to the fault core [e.g. *Nishimoto and Yoshida*, 2010].  
91 Studies that have considered granite alteration have focused on the alteration mechanisms [e.g.  
92 *Bartier et al.*, 2008; *Nishimoto and Yoshida*, 2010; *Savage et al.*, 1987] or other rock mechanical  
93 properties [e.g. *Chen et al.*, 2018; *Coggan et al.*, 2013] instead of friction. *Meller and Kohl*  
94 [2014] considered both chemical alteration and rock frictional behaviour and analysed the impact  
95 of hydrothermal alteration zones on the mechanical behaviour in granite at Soultz-sous-Forêts  
96 (France). They compared borehole logs and friction literature for different rock types and clast  
97 clay mixtures and related alteration state and bulk clay content of the rock to frictional strength.  
98 Furthermore, they observed on the geothermal reservoir scale that large seismic events were  
99 limited to unaltered granite, while clay-rich zones only hosted small magnitude events. However,  
100 *Meller and Kohl* (2014) only related this to friction coefficients from the literature and neglected  
101 frictional stability (defined here in terms of the rate and state friction parameters ( $a - b$ ), as  
102 described in the data processing section later) as important factor for nucleation of seismic  
103 events.

104 Trends in frictional behaviour have been linked to microstructural changes, for mono-  
105 mineral powders [e.g. *Bedford and Faulkner*, 2021; *Beynon and Faulkner*, 2020; *Leclère et al.*,  
106 2016], and synthetic multi-mineral powder mixtures [e.g. *Bedford et al.*, 2022; *Bos et al.*, 2000;  
107 *Tembe et al.*, 2010]. While this research provides important fundamental insights, natural rocks  
108 often contain more than three minerals and the minerals mechanically interact during shearing,  
109 creating complex microstructures. To investigate the links between friction and microstructure in  
110 nature, ground-up complex rocks were tested [e.g. *He et al.*, 2007; *Hunfeld et al.*, 2017; *Okuda et*  
111 *al.*, 2023], including granite [e.g. *Beeler et al.*, 1996; *Hadizadeh et al.*, 2015; *Ikari et al.*, 2011].  
112 Although, granite friction was analysed at hydrothermal conditions [e.g. *Blanpied et al.*, 1995]  
113 and with respect to geothermal systems [e.g. *Zhang et al.*, 2023], nobody has yet linked their  
114 microstructural observations to friction and seismicity in geothermal systems or to the effects of  
115 natural geochemical alteration on friction.

116 In this study, we investigate the effect of geochemical alterations on the frictional  
117 behaviour and microstructure in granitic fault systems (Figure 1). The key questions addressed

118 here are: Does natural alteration promote or inhibit unstable sliding and with this induced  
 119 seismicity? How is the friction behaviour related to structural changes triggered by alteration?  
 120 We conducted a series of direct shear experiments at room temperature (18°C) and at 180°C  
 121 under near in-situ stress conditions on Cornish granites, representative of the fault zone targeted  
 122 in the UDDGP project. We prepared gouges from a sequence of samples from the fault zone with  
 123 increasing proximity to the fault core exhibiting different stages of natural chemical alteration.  
 124 Following the shear-experiments, we prepared thin sections from the deformed gouges and  
 125 analysed the development of the gouge's microstructure.

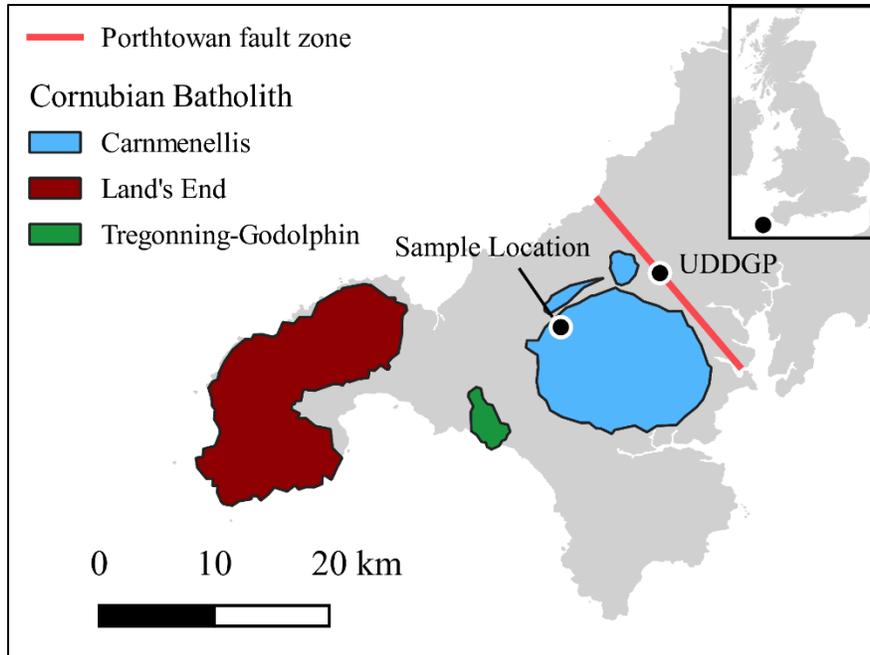


126 **Figure 1: (left) Schematic of the UDDGP project; Porthtowan fault zone as reservoir for fluid production and**  
 127 **reinjection. (right) Rock undergoes temperature and pressure changes under geothermal operation. This**  
 128 **potentially geochemically alters the rock along the flow paths changing the mineralogy around the fault. New**  
 129 **mineralogy causes the formation of other shear structures and changes frictional properties that affect the**  
 130 **potential of induced seismicity [modified after Ledingham et al., 2019].**  
 131

## 132 2 Materials and Methods

### 133 2.1 Sample Material

134 We collected granite samples from the Carnmenellis pluton in SW Cornwall, England.  
 135 The pluton is part of the Cornubian batholith that spans from the Isles of Scilly in the SW to the  
 136 Dartmoor pluton in the NE [Bott et al., 1958]. The batholith was emplaced during the early  
 137 Permian post-collisional extension and the Carnmenellis pluton was dated to be  $293.7 \pm 0.6$  Ma  
 138 [Chesley et al., 1993]. The pluton contains significant amounts of biotite and muscovite, is  
 139 medium- to coarse-grained and contains potassium feldspar phenocrysts (< 25 mm) that make up  
 140 <5 to 25 wt-% of the rock [Simons et al., 2016].



141  
 142 **Figure 2: Map of south-west Cornwall indicating the locations of three major plutons. The plutons are part of**  
 143 **the Cornubian Batholith emplaced during early Permian post-collisional extension. Sampling location**  
 144 **(Holman's Test Mine) is shown and the approximate position and strike of the Porthtowan fault zone**  
 145 **[modified after *Simons et al.*, 2016].**

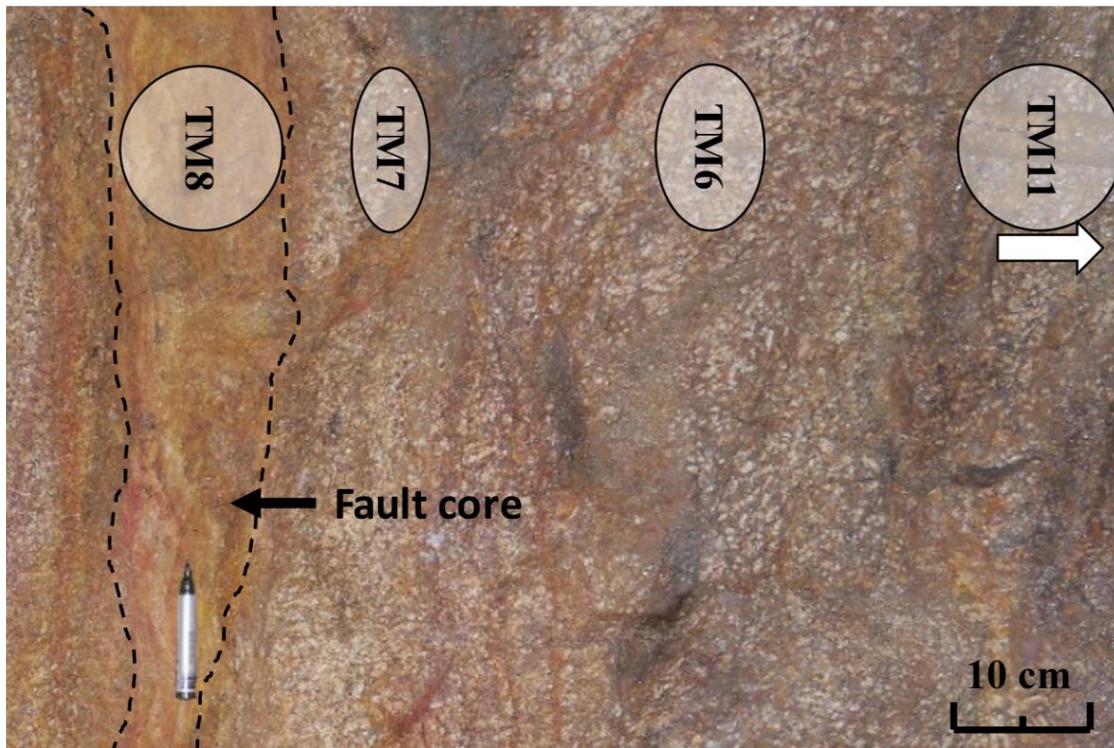
146 The granite samples used here have been collected in Holman's test mine which is  
 147 located about 1.5 km SE of Camborne (Figure 2) and reaches ~ 30 m deep into the Carnmenellis  
 148 pluton. The mine was selected for sampling because rocks from the mine are less weathered than  
 149 surface outcrop samples. The mine hosts several steeply inclined veins and faults in the granites  
 150 – known as cross-course structures – that have developed in Late Permian to Triassic due to an  
 151 ENE-WSW extensional regime in southern England [*Chadwick and Evans*, 1995]. These  
 152 structures usually crosscut and displace earlier mineral veins that formed during or shortly after  
 153 granite emplacement in Early to Middle Permian times, and their strike is usually N-S to NW-  
 154 SE. It is these structures that are targeted in the UDDGP (Porthtowan fault zone) and Eden  
 155 (Great cross-course) projects. We therefore consider the cross-course structures at Holman's test  
 156 mine to be a suitable analogue for our study, and relevant for geothermal energy projects in  
 157 Cornwall.

158 We here study the alteration across the cross-course structures in Holman's test mine as  
 159 analogue for alteration in the Porthtowan fault zone at depth. The samples were collected at  
 160 variable distances to the fault core (Figure 3) to capture increasing geochemical granite alteration  
 161 with decreasing distance to the fault core, which was confirmed by XRD compositional analysis  
 162 (Table 1). With increasing proximity to the fault core (see Figure 3), the samples show  
 163 increasing clay (up to 25 wt-%) and decreasing plagioclase contents (down to 0 wt-%). The  
 164 sample from within the fault core (TM8) shows the highest quartz (59 wt-%) and a slightly lower  
 165 clay content (22 wt-%). Smectite+chlorite contents increase (2 to 11%) closer to the fault core  
 166 while muscovite+biotite+illite contents decrease (18 to 10%), indicating progressive alteration  
 167 towards the fault core. Sample Sa6 was collected from another cross-course fault from the same  
 168 mine for comparison purposes. The fault strikes parallel to the main fault sampled but was

169 located approximately 50 m further East in a parallel drive. Sa6 contains higher amounts of  
 170 kaolinite and lower amounts of smectite+chlorite compared to TM samples. Sa6 indicates the  
 171 potential variability in the composition of rock that underwent argillic alteration, while the  
 172 overall mechanism of decreasing plagioclase and increasing clay content is the same.

173 Sample preparation involved crushing the samples mechanically to cm-sized pieces.  
 174 Samples were then milled in an agate disc mill for 30-90 sec. The powders obtained were  
 175 subsequently sieved to < 125  $\mu\text{m}$  to ensure a uniform maximum grain size. Milling and sieving  
 176 was repeated until all material passed through the sieve to avoid fractionation. This sample  
 177 powder represents fault gouge to study friction behaviour.

178



179

180 **Figure 3: Image of a cross-course structure in Holman’s test mine with markers of approximate sampling**  
 181 **locations. The position of the fault core is marked by dashed lines and the marker was placed for scale. The**  
 182 **width of the tags represents the approximate volume from where each sample was taken. TM11 was collected**  
 183 **one meter away and is considered unaltered. Sa6 was collected from another cross-course structure in the**  
 184 **mine (Table 1).**

185 **Table 1: XRD results for granitic samples used in this study in wt-%. TM8 to TM11 are ordered by proximity**  
 186 **to fault core, with TM8 having been collected within the core. Sa6 was collected at another fault for**  
 187 **comparison.**

Mineral	Sa6	TM8	TM7	TM6	TM11
Quartz	37	59	31	31	31
Plagioclase	1	0	2	8	20
K-Feldspar	38	18	37	36	28
Ankerite	< 0.5	< 0.5	< 0.5	< 0.5	0
Anhydrite	< 0.5	< 1	1.5	< 0.5	0

Tourmaline	< 1	< 0.5	< 0.5	< 1	< 1
Total Clay	20	22	25	23	21
Msc+Bio+Ill*	10	10	13	12	18
Kaolinite	7	1	2	2	1
Chl+Sme**	3	11	11	9	2
Unknown***	3	0	3	2	<1

\* Muscovite, Biotite and Illite

\*\* Chlorite and Smectite

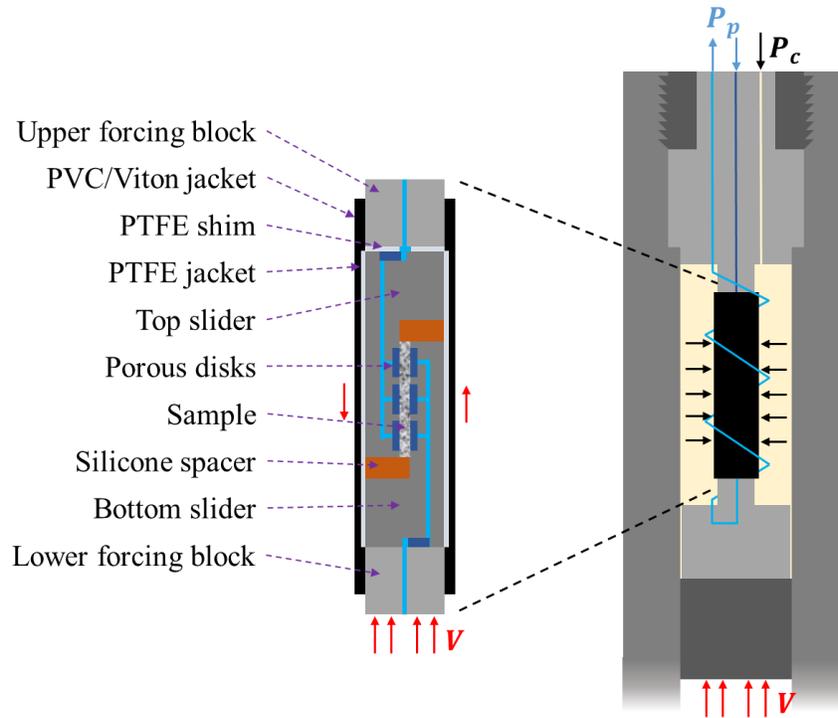
\*\*\* Amorphous and not identifiable components

188

## 189 **2.2 Triaxial apparatus and direct shear assembly**

190 Our experiments were performed using a triaxial deformation apparatus at the University  
 191 of Liverpool. The apparatus allows the application of confining pressures of up to 250 Mpa and  
 192 pore pressures up to 200 MPa using two servo-controlled fluid systems. The axial piston moves  
 193 via a servo-controlled gear train and can be operated by a force or displacement feedback loop.  
 194 An internal force gauge measures differential axial load of up to 300 kN. Three external furnaces  
 195 control experimental temperatures while the axial force gauge at the bottom of the vessel is  
 196 water-cooled to reduce thermal drift. The apparatus is described in detail in *Faulkner and*  
 197 *Armitage* [2013].

198 The direct shear assembly used in our experiments consists of two L-shaped sliders,  
 199 between which a 20x36 mm gouge slab is located (Figure 4). Pore pressure reaches the sample  
 200 through porous stainless-steel frits that are push fitted into the slider surfaces. 1.3 grams of dry  
 201 sample powder were evenly distributed on one of the sliders. We then pre-compressed the  
 202 powder onto the sliders at about 5 Mpa stress normal to the slider surface to create a gouge layer  
 203 of ~1 mm thickness. To ensure coupling between gouge and slider, the contact surface of the  
 204 slider is made up of a series of 100 µm deep machined grooves. Silicone rubber spacers  
 205 accommodate the displacement via elastic compression and stabilise the assembly. The  
 206 combined slider-sample-spacer set is wrapped in a thin sheet of Polytetrafluoroethylene (PTFE)  
 207 to reduce friction between sliders and the surrounding jacket. The jacket consists of Polyvinyl  
 208 chloride (PVC) in low temperature experiments and of Viton rubber in high temperature  
 209 experiments. A thin PTFE disk was fitted to the top of the jacketed sliders to allow for small  
 210 lateral displacements of the sliders during pressurisation and compaction of the gouge layer. A  
 211 detailed description and a schematic of the direct shear assembly are provided in *Faulkner et al.*  
 212 [2018].



213  
 214 **Figure 4: Schematic drawing of the triaxial rig and a detailed view of the direct shear assembly [modified**  
 215 **after Faulkner *et al.*, 2018].**

### 216 2.3 Experimental procedure

217 Most shear experiments were performed at a confining pressure  $P_c$  of 130 MPa and a pore  
 218 fluid pressure  $P_p$  of 50 MPa, leading to an effective pressure  $P_{\text{eff}} (= P_c - P_p)$  of 80 MPa on the  
 219 sample. These pressures correspond to the lithostatic and hydrostatic pressures in a granite body  
 220 at 5 km depth, respectively, and were chosen to be comparable to the conditions at the bottom of  
 221 the production well at UDDGP [Paulillo *et al.*, 2020]. We used room temperature experiments to  
 222 initially investigate the effect of gouge composition and then tested at 180°C to analyse  
 223 temperature effects. The axial piston was advanced at fixed rates, compressing the slider setup,  
 224 and causing shear deformation of the samples. Following a run-in phase of 0.3  $\mu\text{m/s}$  for 1.5 mm,  
 225 the axial displacement rate was stepped every 0.5 mm between 0.3  $\mu\text{m/s}$ , 1  $\mu\text{m/s}$  and 3  $\mu\text{m/s}$  until  
 226 a maximum displacement of 5 mm was reached (velocity sequence displayed in detail in Figure  
 227 5). A slow run-in velocity was chosen to ensure the shear-enhanced compaction of the gouge did  
 228 not result in pore-fluid pressure transients within the gouge layer that might affect mechanical  
 229 results [Faulkner *et al.*, 2018]. Load point displacement (resolution  $\pm \sim 0.5 \mu\text{m}$ ), confining  
 230 pressure ( $\pm 0.01 \text{ MPa}$ ), pore fluid pressure ( $\pm 0.01 \text{ MPa}$ ), axial load ( $\pm 0.05 \text{ kN}$ ), and temperature  
 231 ( $\pm 0.1^\circ\text{C}$ ) were measured, and the corresponding signals logged at a frequency of 10 Hz.

232 Following the experiments, the sheared gouge slabs were carefully taken out of the  
 233 assembly and left to dry at room temperature for several days. They were then impregnated with  
 234 epoxy resin to conserve structure and stabilise the thin slab for thin section preparation. Thin  
 235 sections were cut perpendicular to the shear surface and tangential to the shear direction. We  
 236 polished the section and analysed them using Scanning-Electron Microscopy (SEM) as well as  
 237 Energy-Dispersive X-ray Spectroscopy mapping (EDS mapping) using a Quanta 650 FEG SEM.

238 We used Avizo, a software application for data visualisation by ThermoFisher Scientific,  
 239 to analyse and quantify trends in the microstructure by processing the SEM images of the thin  
 240 sections. The images were denoised using a non-local means filter and the grey levels readjusted  
 241 using sigmoidal intensity remapping. We then segmented the images through thresholding and  
 242 analysed the segmented particles for grain size (as grain area  $a_j$ ), grain shape (as aspect ratio of a  
 243 fitted ellipsoid  $AR_j$ ), and grain orientation. The latter is calculated as the angle between the  
 244 gouge layer and the direction of the maximum Feret diameter of a grain in the 2D image. The  
 245 Feret diameter of a grain at a specific angle describes the distance between two parallel lines  
 246 perpendicular to that angle enclosing a grain at its outside boundary. It is comparable to the  
 247 diameter that would be measured with a calliper, if the calliper was fixed to the image plane with  
 248 the ruled scale of the calliper positioned parallel to the specific angle of interest. Furthermore, we  
 249 filtered out all grains smaller than 8 pixels because they are close to the image resolution,  
 250 limiting interpretation of shape and orientation. This filtering of the smallest grain portion and  
 251 everything below will be addressed in the discussion section.

## 252 2.4 Data processing

253 The axial load applied during each experiment was converted into shear stress  $\tau$  on the  
 254 gouge sample which we used to calculate the apparent coefficient of friction according to  
 255  $\mu = \tau / \sigma_{\text{eff}}$ . Effective normal stress  $\sigma_{\text{eff}}$  is the difference between normal stress  $\sigma$ , which in our  
 256 setup is induced by the confining pressure, and pore fluid pressure  $P_p$ , i.e.  $\sigma_{\text{eff}} = \sigma - P_p$ .  
 257 Following *Behnsen and Faulkner* [2012], cohesion and jacket strength are ignored. The data  
 258 were analysed using the Rate-and-State Friction (RSF) framework [*Dieterich*, 1978; 1979;  
 259 *Ruina*, 1983] to interpret the velocity dependence of the friction coefficient according to:

$$\mu = \mu_0 + a \ln\left(\frac{V}{V_0}\right) + b \ln\left(\frac{V_0 \theta}{D_{RS}}\right) \quad 1)$$

260 Here,  $\theta$  is a gouge state variable,  $a$  determines the magnitude of the instantaneous change  
 261 in  $\mu$  upon a given step change in sliding velocity from a reference value  $V_0$  to a new value  
 262  $V = eV_0$ ,  $b$  reflects the magnitude of the change in  $\mu$  during the evolution to a new steady state  
 263 value over a critical sliding distance  $D_{RS}$ , and  $\mu_0$  is the steady state friction coefficient measured  
 264 at the reference velocity  $V_0$ . For the state variable  $\theta$  we used Dieterich's aging law formulation  
 265 [*Dieterich*, 1979]:

$$\dot{\theta} = 1 - \frac{V\theta}{D_{RS}} \quad 2)$$

266 For changes in the steady state friction coefficient  $\mu_{ss}$  resulting from a stepwise change in  
 267 sliding velocity equations (7) and (8) yield [e.g. *Marone*, 1998]:

$$\frac{d\mu_{ss}}{d(\ln V)} = (a - b) \quad 3)$$

268 A positive  $(a - b)$  value indicates that the friction coefficient increases with higher  
 269 velocities while a negative value of  $(a - b)$  indicates a decrease in friction coefficient with an  
 270 increase in velocity. Systems that exhibit positive  $(a - b)$  values are therefore called “velocity-  
 271 strengthening” (VS) and slip in such systems is intrinsically stable. In contrast, systems that  
 272 show negative  $(a - b)$  values are called “velocity-weakening” (VW) and have the potential to  
 273 nucleate unstable slip [e.g. *Scholz*, 2019]. We used the MATLAB software package RSFit3000

274 by *Skarbek and Savage* [2019] to obtain the Rate-and-State Friction parameters. The software  
275 uses a nonlinear least-squares fitting routine for fitting a RSF model to a velocity step and  
276 outputs the related parameters.

### 277 **3 Results**

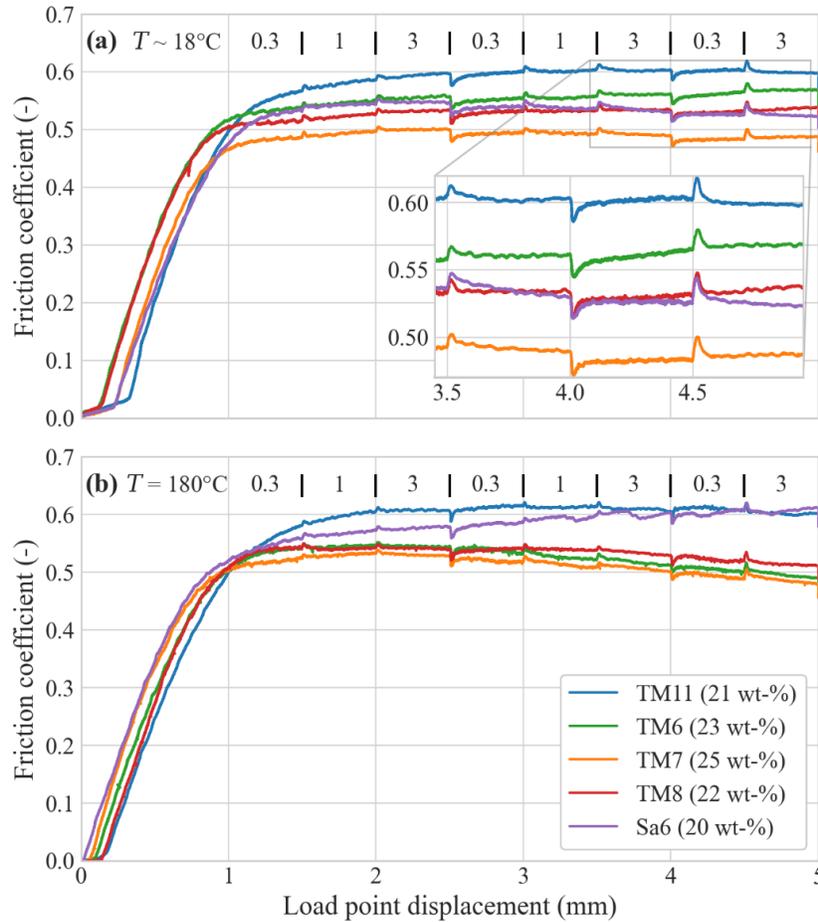
#### 278 3.1 Direct shear experiments

279 Figure 5a shows representative curves of friction coefficient  $\mu$  versus load point  
280 displacement for all samples at room temperature ( $\sim 18^\circ\text{C}$ ) and 80 Mpa effective pressure. From  
281 displacements of 0.1 mm to 0.9-1.2 mm,  $\mu$  increases steeply and linearly to values of 0.43 to  
282 0.52, when the apparent yield point is reached. Following yielding, the friction coefficients of the  
283 gouges generally continue to gently increase with displacement until a displacement of 2 mm.  
284 Thereafter, the samples continue to show similar behaviour as all reach a steady state before the  
285 final displacement of 5 mm. However, their final friction coefficients are different with TM11  
286 reaching the highest value of  $\sim 0.6$  and TM7 the lowest value of  $\sim 0.49$ .

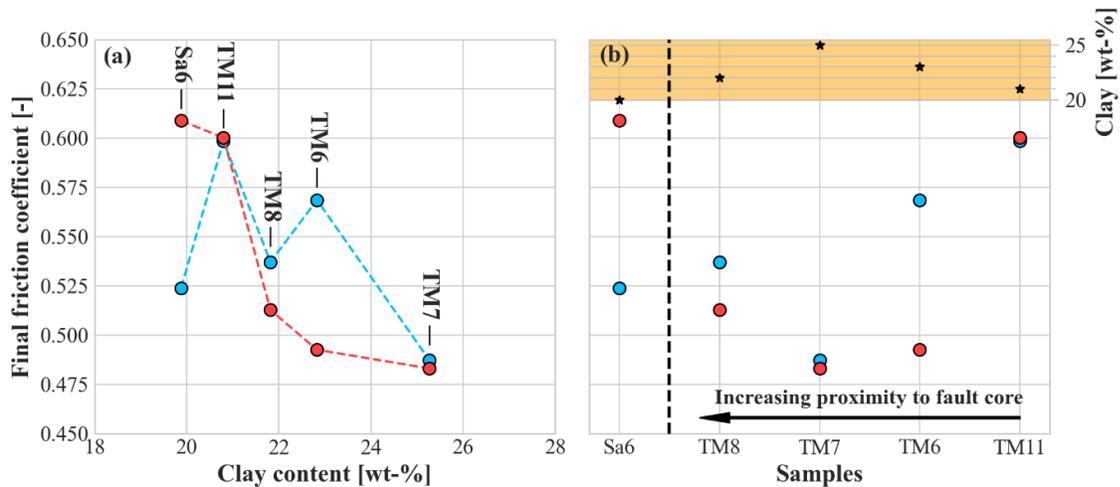
287 Figure 5b shows friction coefficient  $\mu$  versus load point displacement for the experiments  
288 performed at  $180^\circ\text{C}$ . Similar to measurements performed at  $18^\circ\text{C}$ ,  $\mu$  increases steeply and  
289 linearly from 0.1 mm to 0.9-1.2 mm to values of 0.48 to 0.56, after which the apparent yield  
290 point is reached. Thereafter,  $\mu$  of TM11 remains constant, for Sa6 it increases slightly, indicating  
291 minor strain hardening, and for TM6, TM7 and TM8 it decreases slightly with displacement,  
292 indicating minor strain weakening. We generally observe that for the high temperature  
293 experiments at  $180^\circ\text{C}$ , high clay contents ( $>21$  wt-%) lead to strain weakening (TM6, TM7,  
294 TM8), while strain hardening is observed for samples with a clay content of  $\leq 21$  wt-% (TM11,  
295 Sa6).

296 Figure 6 shows the final frictional coefficient, which is a mean of the friction coefficients  
297 over the last 0.4 mm displacement, versus clay content of the gouge (a) and the relative sampling  
298 position of the gouge in the fault (b). Friction coefficients range from 0.49 for sample TM7 to  
299 0.60 for sample TM11 at  $18^\circ\text{C}$ . At  $180^\circ\text{C}$ , they seem to decrease with increasing clay content  
300 from 0.61 at 20 wt-% clay for Sa6 to 0.49 at 25 wt-% for TM7 (see Figure 6a). This trend,  
301 however, is not supported by the low temperature data. TM11, TM6 and TM7 show that clay  
302 content increases while friction coefficient decreases with increasing proximity to the fault core  
303 (see Figure 6b). TM8 was collected from within the fault core but shows final friction  
304 coefficients at low and high temperature which are higher than for TM7, the next sample further  
305 away from the core.

306

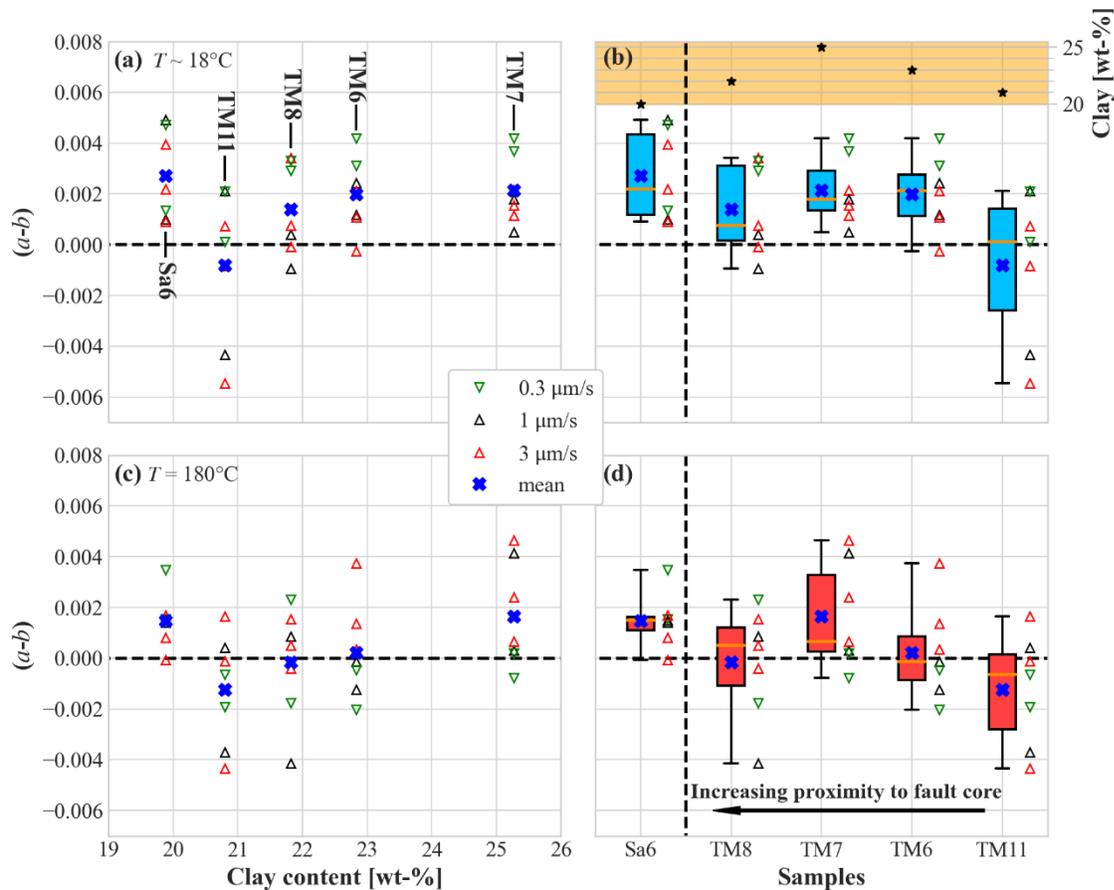


307  
 308 **Figure 5: Friction coefficient against load point displacement for all samples tested, at room temperature**  
 309 **~18°C (top) and at 180°C (bottom),  $P_{\text{eff}} = 80$  MPa. Velocity steps are in  $\mu\text{m/s}$  and are marked on top of the**  
 310 **graphs. Percentages in the legend indicate the clay content in the sample.**



311  
 312 **Figure 6: Final friction coefficient calculated as mean over the last 0.4 mm displacement versus (a) clay**  
 313 **content and (b) relative position in the fault. In (b) Samples are ordered by increasing proximity to fault core**  
 314 **while Sa6 was collected at another fault and used as a reference here. Blue dots indicate room temperature**  
 315 **results (~18°C); red dots represent high temperature results (180°C).**

316 Figure 7 relates the frictional stability of our samples to their clay content (a, c) and to  
 317 their relative position to the fault core (b, d). The combined parameter  $(a - b)$  at low  
 318 temperature (Figure 7a) ranges from -0.0057 to 0.0049. The relatively low  $b$  values of Sa6 (20  
 319 wt-% clay) result in  $(a - b)$  values that are on average higher than for the other samples. Values  
 320 of  $(a - b)$  for Sa6 are all positive and thus indicate velocity strengthening behaviour. Excluding  
 321 sample Sa6,  $(a - b)$  increases with increasing clay content. TM11 (21 wt-% clay) shows both  
 322 velocity strengthening and velocity weakening behaviour with values from -0.0057 to 0.0022.  
 323 Values of  $(a - b)$  range from -0.0012 to 0.0035 and -0.0003 to 0.0043 for samples TM8 and  
 324 TM6 with 22 wt-% and 23 wt-% clays, respectively, indicating increasing velocity strengthening  
 325 behaviour with increasing clay content. The sample with the highest clay content (TM7, 25 wt-  
 326 %) shows  $(a - b)$  values from 0.0003 to 0.0043, i.e. velocity strengthening behaviour at all  
 327 velocities tested. Clusters related to post-step slip velocity can be recognised, where  $(a - b)$  at 3  
 328  $\mu\text{m/s}$  are lowest and values at 0.3  $\mu\text{m/s}$  highest. At 180°C (Figure 7c), the ranges and the means  
 329 of  $(a - b)$  of all samples shifted to smaller and more negative values and with this towards more  
 330 velocity weakening behaviour. We generally observe increasing  $(a - b)$  values with increasing  
 331 clay content at high temperature for TM samples. Furthermore, the mean frictional stability  
 332 generally increases with increasing proximity to the fault core from -0.0008 to 0.0021 at low  
 333 temperature (Figure 7b) and from -0.0013 to 0.0017 at high temperature (Figure 7d).



334 **Figure 7: Frictional stability  $(a-b)$  against phyllosilicate content (left column) and ordered by proximity to**  
 335 **fault core (right column) at room temperature ( $\sim 18^\circ\text{C}$ ; top) and at high temperature ( $180^\circ\text{C}$ ; bottom). Blue**  
 336 **crosses represent mean values. Boxplots show mean value (blue cross), median (orange line), upper and lower**  
 337 **quartile (box) and overall data range (whiskers) for data of each sample.**  
 338

## 339 3.2 Gouge microstructures

340 In the following we describe the main characteristics of the gouge microstructures of all  
341 our samples at room temperature and of TM11, TM7 and Sa6 at high temperature. An overview  
342 and some example microstructures are given in this chapter, but more detailed images, as well as  
343 EDS-element-maps are attached in the supplementary materials. We use the terminology of  
344 *Logan et al.* [1992] in the description of the main shear structures.

345 *Unaltered sample TM11:* Figure 8a shows compressed but unsheared TM11. This was  
346 analysed to study the sample microstructure before and after shear deformation. The  
347 homogeneously distributed large clasts (<125  $\mu\text{m}$ ) are mostly quartz, albite, and orthoclase with  
348 occasional larger interparticle booklets of muscovite and biotite. In addition, we find other small  
349 clay minerals like kaolinite and smectite and small amounts of apatite and rutile, while the latter  
350 is forming needles of 10-20  $\mu\text{m}$  in length. Elongated grains partly orient between  $\sim 75^\circ$  and  $90^\circ$   
351 (normal) to the compression direction. Biotite and muscovite predominantly orient perpendicular  
352 to normal stress and partially form layers around larger clasts.

353 In comparison, the sheared TM11 sample contains significantly fewer clasts of larger size  
354 compared to the unsheared counterpart (Figure 8b). This makes it more heterogenous with  
355 respect to spatial grain size distribution and causes the formation of specific shear structures, of  
356 which examples are shown in Figure 9. Large clasts are mostly quartz, while feldspars (albite  
357 and orthoclase) are widely fractured leading to a decrease in grain size. Shear deformation did  
358 not only lead to grain size reduction but also to generally rounder grains as a result of wear  
359 (Figure 9b and c). Some clasts roughly align and orient along early P-shears. Biotite and  
360 muscovite booklets, that aligned perpendicular to the compression direction in the unsheared  
361 sample, partially maintain previous layers they formed around larger clasts while others have  
362 been crushed between larger and stronger grains. Most micas orientate in the direction of P-  
363 foliation and R1-Riedel-shears (Figure 9c). Furthermore, the shearing has crushed apatite  
364 crystals that form ‘tails’ (Figure 9d). These tails are crushed grains that align along the shear  
365 sense. Larger clasts are surrounded by fine to very fine-grained (< 20  $\mu\text{m}$ ) matrix which  
366 concentrates in dedicated ‘deformation bands’ and are lower in visible porosity compared to  
367 areas where larger grains dominate. Such deformation bands are found at the top and at the  
368 bottom of the gouge, indicating the formation of boundary Y-shears (Figure 8b), but are also  
369 along R1-shears (Figure 9a) where they promote the formation of fractures. Neither P-foliation  
370 nor R1-shears are very obvious throughout most of the sample. However, the gouge fractured  
371 along some R1-shears (Figure 9a) and R1- and P-orientation are indicated in mineral orientation  
372 and alignments.

373 *TM11 sheared at 180 °C:* The structure of the unaltered granite gouge TM11 tested at  
374 180°C differs significantly from the experiment performed at room temperature (Figure 8c). Like  
375 for the low temperature test, it contains large grains in a small-grained matrix. However, large  
376 grains are much less abundant, suggesting grinding to form smaller particles. Here, quartz and K-  
377 feldspar are fractured, while mainly fracturing of K-feldspar was observed for the low  
378 temperature experiment. The overall structure displays the shear sense well due to an easily  
379 identifiable P-foliation indicated by orientated and curved micas and apatite tails. The orientated  
380 and slightly ground micas form planes of weakness that promote fracture formation along shears.  
381 The P-foliation tails off into boundary Y-shears towards the top and the bottom of the gouge  
382 layer. In this gouge, P-foliation and Y-shears are dominant while R1-shears are present but less

383 developed. The shear structures are more comparable to the more altered samples described  
384 below (e.g. TM7) than to room temperature TM11.

385 *TM6 sheared at room temperature:* The gouge of TM6 (Figure 8d) is less dominated by  
386 larger grains than TM11. The large grains are K-feldspar and quartz, with some larger booklets  
387 of mica. EDS-mapping indicates small amounts of albite in the fine-grained matrix indicated by  
388 elevated Na<sup>+</sup> concentrations. The P-foliation is composed of orientated and bent micas as well as  
389 orientated elongated grains. It is most obvious in the central part of the gouge, interbedded  
390 between horizontal boundary Y-shears. The latter take the form of deformation bands, dominated  
391 by an accumulation of smaller grains. TM6 contains brighter areas in the central part of the  
392 gouge that can be identified as crushed K-feldspar.

393 *TM7 sheared at room temperature:* The gouge made from TM7 is highly altered and is  
394 characterised by the highest clay content of all samples tested (Figure 8e). This results in a  
395 significantly different gouge structure in comparison to TM11. It contains less larger grains  
396 (quartz, rounded or fractured K-feldspar) that are surrounded by a fine-grained matrix. EDS-  
397 mapping shows little Na<sup>+</sup> within the gouge which confirms the XRD results suggesting small  
398 amounts of plagioclase. Muscovite is partially ground to form particles < 50 µm (some forming  
399 ‘tails’ like apatite) and either orientated normal to stress direction or forming oscillating P-  
400 foliations. Al<sup>3+</sup> in the EDS maps further indicates that the P-foliation contains higher amounts  
401 muscovite and illite compared to surrounding areas. Only small amounts of biotite are found in  
402 TM7, suggesting dissolution, transformation into chlorite, or grinding to fine particles. The fine-  
403 grained matrix in TM7 is dominated by pervasive P-foliation rather than distinct shears. P, R,  
404 and Y-shear structures developed more localised in the sheared TM11 gouge. Like for TM11, the  
405 boundary Y-shears in TM7 form towards the upper and lower boundary of the gouge, while  
406 fractures have formed along P- and R1-shears. In general, the SEM image shows brighter and  
407 darker areas; their distribution highlights the anastomosing foliation in the gouge structure.  
408 Brighter minerals are related to high K-feldspar contents following grinding to smaller particles.  
409 In comparison, porosity is filled with a less dense epoxy resin, leading to generally darker  
410 colours. As such, the SEM image appears darker towards the centre of P-, R1-, and Y-shears  
411 which is potentially related to a concentration of clays and slightly higher porosity due to the  
412 onset of fracturing.

413 *TM7 sheared at 180 °C:* In contrast to TM11, the structure of TM7 sheared at 180°C  
414 (Figure 8f) is very similar to the structure following the room temperature test on TM7. The big  
415 grains are slightly more damaged and more mid-sized grains are present at high temperature. The  
416 bright and dark zones described in TM7 are slightly more obvious, indicating shear direction and  
417 grain displacement. The undulating P-foliation fabric described before at low temperature is  
418 more defined at high temperature due to the more obvious P-foliation. This is because clays are  
419 generally better oriented, leading to the development of more bright zones.

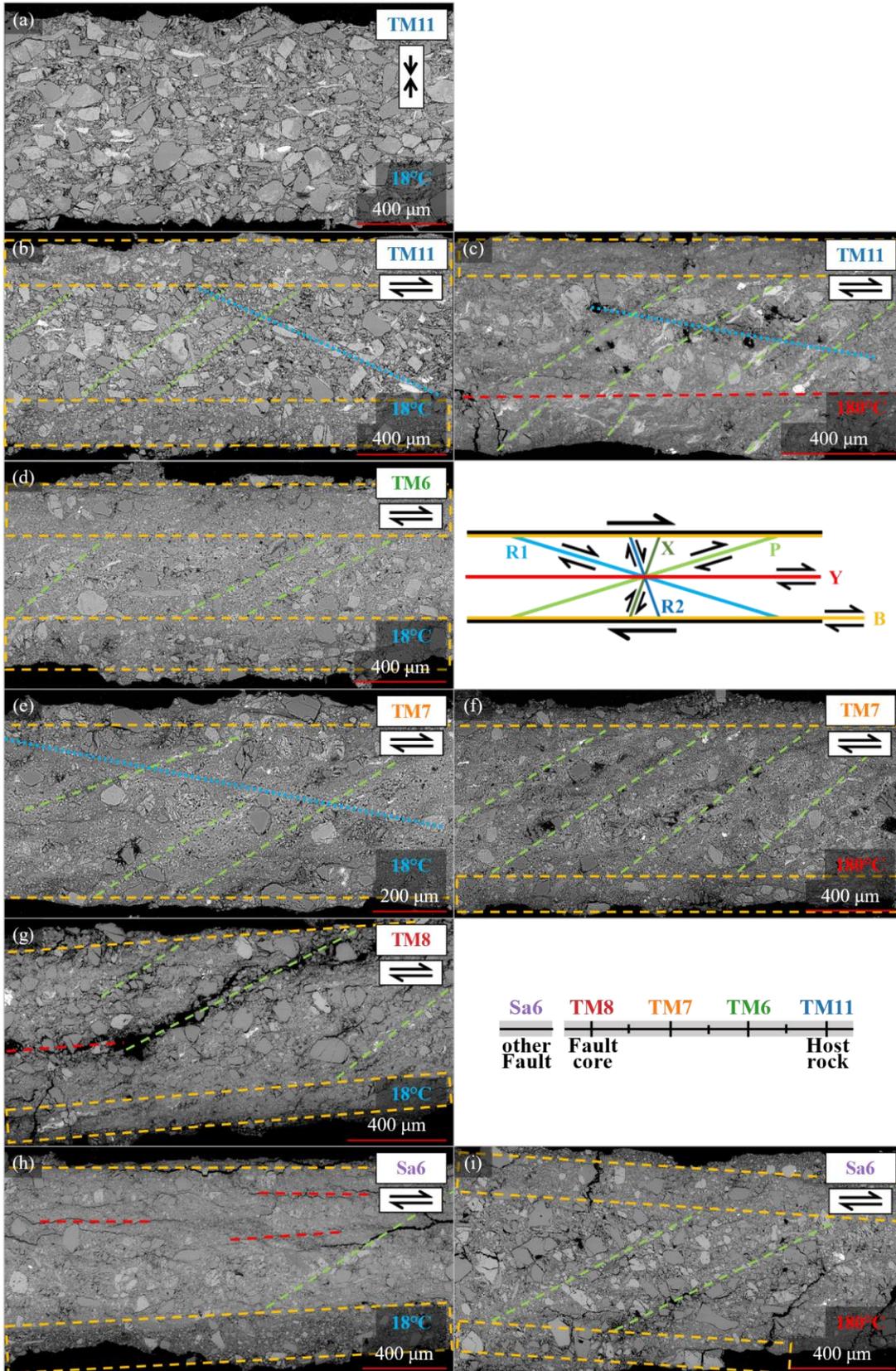
420 *TM8 sheared at room temperature:* The fault core sample TM8 contains almost twice as  
421 much quartz as the other samples. Therefore, the large grains in the sheared gouge are dominated  
422 by quartz with some minor amounts of K-feldspar (Figure 8g). As before, these are surrounded  
423 by a small-grained matrix. The large grain fraction is higher than in TM6 and TM7, and lower  
424 than in TM11. The dark gouge is likely caused by low feldspars and high quartz contents.  
425 Crushed apatite embedded within the dark gouge can be found, creating some brighter areas.  
426 Furthermore, very bright fine grains are present throughout the gouge, likely being haematite,  
427 resulting in a reddish colouration. Apatite indicates the shear sense by forming ‘tails’ of crushed

428 particles that follow the P-foliation. The gouge shows fracturing along the P-foliation and  
429 slightly darker traces have formed where the gouge expanded as these areas are more porous  
430 (darker). Orientated micas are important to identify shear structures in previous gouges.  
431 However, only few large pieces of muscovite are left, and biotite is completely absent. The  
432 oscillating P-foliation transitions into horizontal boundary Y-shears towards the upper and lower  
433 boundary of the gouge. While P- and Y-shearing is obvious, R1-shears are more difficult to  
434 identify because they mainly take the form of aligned grains.

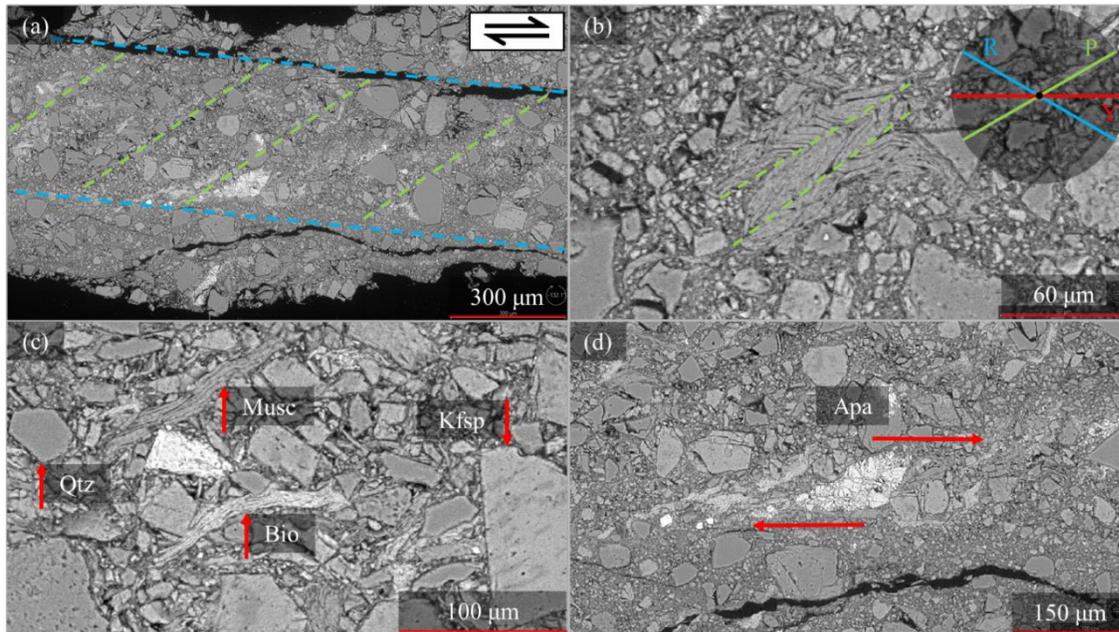
435 *Sa6 sheared at room temperature:* The mineralogical composition of Sa6 is similar to  
436 TM7 but contains higher kaolinite and quartz contents. The gouge from Sa6 is more  
437 heterogeneous than TM7. The structure is similar to TM6, with a similar content of large grains  
438 (Quartz, K-feldspar), embedded in a small-grained matrix (Figure 8h). The feldspars are  
439 fractured and have started to disperse into the matrix. Apart from the variable grain size, the  
440 gouge also seems to contain more porous areas with distinct boundaries separating them from  
441 areas of low porosity. The matrix orientation appears chaotic, not clearly representing the  
442 imposed shear sense in contrast to the other gouges. Large grains orientate in both, P- and R1-  
443 direction but the gouge is mainly fractured along P- and Y-direction as planes of weakness. The  
444 most dominant shears have formed along the P-foliation, underlining sinistral shear sense.

445 *Sa6 sheared at 180 °C:* The microstructure following shearing at high temperature shows  
446 quite similar structure to its low temperature equivalent (Figure 8i); however, shears are better  
447 developed. Unlike for TM7 and TM11, Sa6 shows a higher share of large grains in the high  
448 compared to the low temperature measurement. The matrix is less chaotic with elongated grains  
449 mostly oriented in direction of the P-foliation.

450



452 **Figure 8: SEM images of gouges following experiments. Sample ID, shear sense and test temperature are**  
 453 **indicated on the right side of each image. The gouge in (a) was only compressed without lateral displacement.**  
 454 **Next to (d), a schematic describes typical shear structures as defined by Logan *et al.* [1992] adding B for**  
 455 **boundary Y-shears. with a colour code that is then used in the pictures to mark the related structures. Next to**  
 456 **(g), a schematic indicates the relative origin position of the samples in the fault.**



457 **Figure 9: Examples for structural interpretation on sheared unaltered granite (TM11) at 18°C. (a) Overview**  
 458 **of sheared unaltered gouge in which direction and approximate position of Riedel-shears (blue) and P-**  
 459 **foliation (green) are marked with dotted lines. (b) Double folded muscovite with fold planes parallel to the**  
 460 **direction of the P-foliation. Due to initially having been oriented against shear direction and the relatively low**  
 461 **strength of the mica compared to quartz or feldspar minerals, the grain is folded rather than rotated. (c)**  
 462 **Platy biotite (Bio) and muscovite (Musc) arranging towards P-foliation next to quartz (Qtz) and K-feldspar**  
 463 **(Kfsp) grains. The feldspar contains darker lines indicating perthitic unmixing. (d) Soft apatite grains ground**  
 464 **to smaller particles due to shear, forming ‘tails’ with shape defined by the shear sense.**  
 465

## 466 4 Discussion

### 467 4.1 Alteration stage of samples

468 Our samples were collected at increasing proximity from and within the fault core of a  
 469 cross-course structure in the Carnmenellis granite and exhibit different stages of granite  
 470 alteration (Table 1). Alteration of a host rock gradually increases towards the fault core due to  
 471 enhanced access to fluids circulating through the damage zone [Faulkner *et al.*, 2010]. In  
 472 Carnmenellis granite the main alteration mechanism observed around cross-course structures is  
 473 argillic alteration which implies the formation of clay minerals (mainly kaolinite and smectite) at  
 474 the cost of plagioclase [e.g., Bevins *et al.*, 2010]. With time, due to sericitisation, the kaolinite  
 475 and smectite transform into more stable secondary white mica like illite [Cuadros, 2012]. K-  
 476 feldspar also degrades, showing signs of sericitisation of the sodium-rich mineral cores, but this  
 477 process acts slower than the alteration of plagioclase. Argillic alteration can be observed in  
 478 samples Sa6, TM6, TM7 and TM8, clearly showing a reduction in plagioclase with an increase  
 479 in kaolinite, smectite and chlorite in comparison to TM11 (see Table 1). In addition, the mafic

480 minerals in the rock are transformed into chlorite. In Carnmenellis granite, this affects mainly  
481 biotite which is found in addition to or replaced by chlorite in more altered aggregates [Charoy,  
482 1986]. As a consequence, our samples show a significant loss in muscovite, biotite and illite  
483 from the unaltered (TM11) to the altered state by at least 5 wt-%. As muscovite is relatively  
484 stable under hydrothermal conditions and illite successively forms with time, the main phase lost  
485 is biotite - either transforming into chlorite or lost in solution.

486 As a consequence of transformation of tectosilicates into phyllosilicates, the overall rock  
487 strength decreases and porosity increases [Coggan *et al.*, 2013]. It should be noted that porosity  
488 can only increase until alteration causes major parts of the rocks mineral structure to break down.  
489 At this point compaction can occur which reduces the pore space again. The TM samples  
490 become successively more altered with proximity to the fault core. From TM11 (furthest away  
491 from core) to TM8 (from within core), the amount of plagioclase decreases while the amount of  
492 clay minerals increases.

493 TM8 shows a small reduction in overall phyllosilicate and a significant increase in quartz  
494 content (59 wt-% see Table 1). One possibility to explain the reduction of clay and higher  
495 proportion of quartz in TM8 relative to all other samples is the removal of small clay particles  
496 associated with circulation of fluids along the fault. Alternatively, the fault could have hosted a  
497 quartz vein before shearing which would also increase the amount of quartz in the fault gouge.  
498 There was no obvious evidence for the presence of a former quartz vein in this fault, but it is also  
499 difficult to identify small quartz veins in heavily altered rock, especially after shear  
500 displacement. Furthermore, quartz veins are commonly present in cross-course structures [Bevins  
501 *et al.*, 2010]. Either explanation or even a combination of both appear reasonable. Sa6 was  
502 collected at a different fault in the same mine and based on the amount of plagioclase, it would  
503 be positioned in an alteration stage between TM7 and TM8. However, the combined amount of  
504 clay (kaolinite + smectite + chlorite) in Sa6 is relatively low and the fact that it contains most  
505 kaolinite in all samples makes a position towards the less altered end possible (e.g., between  
506 TM11 and TM6).

#### 507 4.2 Frictional strength and stability of altered granite

508 The alteration sequence with proximity of our samples to the fault core is partly reflected  
509 in the final friction coefficients measured (Figure 6). Notably, the least altered sample TM11  
510 showed the highest final friction coefficient (~0.6), while the lowest final friction coefficient of  
511 ~0.49 was exhibited by TM7, the second closest to the core. Fang *et al.* [2018a] analysed the  
512 frictional behaviour of natural and synthetic rocks with variable composition and clay content.  
513 They reported that frictional strength decreases with phyllosilicate content and increases with  
514 tectosilicate content which is similar to observations made on artificial gouge mixtures with  
515 variable amounts of clay [e.g. Crawford *et al.*, 2008; Takahashi *et al.*, 2007; Tembe *et al.*, 2010].  
516 In our experiments, we make similar observations for TM11, TM6 and TM7, showing the lowest  
517 final friction coefficient for TM7, containing the highest phyllosilicate content of 25 wt-%.  
518 However, the final friction coefficients observed for the other samples are not in full agreement  
519 with a decrease in the friction coefficient with an increase in phyllosilicate content. This may be  
520 related to the narrow range in phyllosilicate content in our samples (20-23 wt-%). Behnsen and  
521 Faulkner [2012] analysed the frictional behaviour of different pure sheet silicates and found that  
522 at an effective pressure of 90 MPa, pure muscovite has a higher friction coefficient at the yield  
523 point than kaolinite, illite or smectite. Micas in TM11 are mostly muscovite and biotite due to the

524 unaltered stage of the sample while the more altered samples contain increasing amounts of illite.  
525 This supports the higher friction coefficient of TM11 even though it contains a higher amount of  
526 phyllosilicates than Sa6.

527 Sa6 and TM6 (intermediate alteration) show similar frictional behaviour at room  
528 temperature with the difference that Sa6 shows strain weakening behaviour, and TM6 shows  
529 strain hardening. The main difference between these two samples lies in the phyllosilicate  
530 composition. While the amount of combined muscovite, biotite and illite is relatively similar,  
531 Sa6 contains more kaolinite and TM6 contains more chlorite and smectite. This does not seem to  
532 explain the observed strength difference as (i) *Behnsen and Faulkner* [2012] found that pure  
533 kaolinite gouge has an almost three times higher friction coefficient at the yield point than pure  
534 smectite and (ii) *Tembe et al.* [2010] found that an increase in the smectite content rather than the  
535 total clay content relates to lower friction coefficients. We hypothesise that this inconsistency is  
536 caused by the small differences in clay contents of our samples, which is reflected in only small  
537 differences in friction coefficients.

538 At high temperature, frictional strength decreases with increasing clay content across all  
539 samples. In the low temperature tests, Samples Sa6 and TM8 deviate from this trend with clay  
540 content. This could be caused by quartz and the feldspars in wet systems becoming more  
541 susceptible to thermally activated deformation mechanisms with increasing temperature [like  
542 stress corrosion cracking and pressure solution, e.g., *den Hartog and Spiers*, 2014]. When the  
543 previously strong tectosilicates become easier to shear and break, there is less dilatational  
544 movement between grains necessary to accommodate horizontal shear [*den Hartog and Spiers*,  
545 2013; *Niemeijer and Spiers*, 2007]. Rather than moving grains relative to other grains potentially  
546 causing lateral movement, the grains can deform or break. The weaker the tectosilicates become,  
547 the more the friction on horizontal clay minerals will contribute to the shear strength of the  
548 gouge. This shift in contribution to shear strength at high temperature could then also explain the  
549 trend of frictional strength with clay content in our high temperature tests.

550 Another possible effect of high temperature in granite friction we partially see in our  
551 samples (e.g., TM6, TM7 and TM8) is a reduction of frictional strength with increasing  
552 temperature. While the frictional strength of TM11 stays almost the same and TM7 shows only a  
553 very small reduction, Sa6 shows a significant increase in frictional strength from low to high  
554 temperature. A potential reason relates to the friction against displacement curve that records the  
555 high temperature experiment on Sa6 (Figure 5b). In contrast to all other curves, the friction  
556 coefficient of Sa6, that reached steady-state conditions below 2.5 mm displacement, starts to  
557 divert from this state at a displacement of 2.5 mm, as the friction coefficient increases, and the  
558 frictional resistance starts oscillating. To analyse the potential origin of this oscillation we looked  
559 at the recorded pressures, the temperature, the axial displacement rates, and the pump volumes.  
560 None of them showed cyclic behaviour with displacement like the friction coefficient. A quick  
561 permeability test following the experiment also showed the sliders to be permeable, ruling out  
562 pore pressure build-ups during deformation. As we did not observe any machine related effect,  
563 we assume the oscillation could have been caused by variations in the microstructure. However,  
564 proving that is difficult as we only analyse the microstructure at the end of the experiment.

565 Frictional stability shows the opposite behaviour compared to the frictional strength for  
566 our alteration sequence (Figure 7). ( $a - b$ ) and thus, frictional stability is velocity weakening  
567 (VW) for TM11 that was sampled the furthest from the fault core and then becomes velocity  
568 strengthening (VS) with increasing proximity to the fault core for Sa6, TM6 and TM7. In TM8,

569 which was sampled from within the fault core,  $(a - b)$  decreases slightly. This can again be  
 570 related to higher amounts of quartz in the sample caused by an additional process. When trying  
 571 to relate the frictional stability behaviour to the overall clay content, TM11, TM6, TM7 and TM8  
 572 also seem to form a trend towards higher  $(a - b)$  values with higher phyllosilicate content. This  
 573 observation agrees with previous work that reported a stabilizing effect of phyllosilicates on slip  
 574 [e.g. *Fang et al.*, 2018a; *Ikari et al.*, 2011; *Tembe et al.*, 2010]. Again, Sa6 is not consistent with  
 575 this trend as it only shows velocity strengthening behaviour while having the lowest  
 576 phyllosilicate content.

577 In the high temperature tests, frictional stability again changes from velocity weakening  
 578 (VW) from TM11 to velocity strengthening (VS) towards the fault core for Sa6, TM6 and TM7  
 579 while decreasing slightly for the core sample TM8. This is the same behaviour of the altered  
 580 samples compared to their low temperature equivalent, indicating that the same or at least similar  
 581 processes control frictional stability at both temperatures. Previous work reports that  $(a - b)$  of  
 582 granular gouges decreases with increasing temperature, changing from velocity strengthening to  
 583 velocity weakening behaviour where the trend turns and then increases with further increase in  
 584 temperature, back from velocity weakening to velocity strengthening behaviour [e.g., *Blanpied et al.*  
 585 *et al.*, 1998; *den Hartog and Spiers*, 2013]. This creates a velocity weakening temperature window  
 586 that *Blanpied et al.* [1998] reported for wet granite gouge at a temperature range between  $\sim 90^\circ\text{C}$   
 587 and  $\sim 350^\circ\text{C}$ . The exact temperature limits of the window vary dependent on other conditions  
 588 [see *den Hartog and Spiers*, 2013], but our results are in general agreement with the observations  
 589 as the frictional stability for all of our samples is reduced at  $180^\circ\text{C}$  compared to the room  
 590 temperature tests. This leads to a more velocity neutral and for some even velocity weakening  
 591 behaviour, which is related to less stable sliding and a higher potential for stick slip behaviour.

#### 592 4.3 Gouge structure and frictional behaviour

593 Argillic alteration is the main process relevant for our suite of samples, by which large  
 594 feldspar crystals are replaced by clay particles. This can be seen in the grain size distributions of  
 595 the increasingly altered samples (TM6, TM7) containing more smaller grains than the less  
 596 altered sample (TM11, quantitative structural analysis in supplementary information). This  
 597 suggests that, in addition to the phase change, argillic alteration also indirectly causes a  
 598 significant reduction in grain sizes, which is affecting frictional behaviour [e.g., *Anthony and*  
 599 *Marone*, 2005; *Bedford and Faulkner*, 2021; *Higashi and Sumita*, 2009; *Jiang et al.*, 2016; *Mair*  
 600 *et al.*, 2002]. *Bedford and Faulkner* [2021] tested the effects of grain size on shear localisation  
 601 and sliding behaviour on quartz powders of different initial grain sizes between 40 and 120 MPa  
 602 normal stress. They report that the smaller the initial grain size of the gouge, the more  
 603 deformation will localise, and unstable sliding will become more likely. We cannot report this  
 604 size effect in our test, but we also observe localisation of shear in our samples.

605 Although grain sizes are larger, the sheared unaltered sample (TM11) contains zones  
 606 where smaller grains accumulate towards the boundaries, indicating the beginning of formation  
 607 of boundary Y-shear zones (Figure 8b). In comparison, grain sizes in the more altered samples  
 608 (TM6, TM7) are homogeneously distributed over the gouge layers, and shearing affects the  
 609 width of the gouges more evenly (Figure 8c and d), dispersing deformation over a larger volume.  
 610 This difference in localised boundary shearing in the unaltered sample against pervasive layer  
 611 deformation in the altered samples is a first effect promoting more stable sliding in our altered  
 612 samples. However, this is opposite to *Bedford and Faulkner* [2021] who reported smaller grain

613 sizes to promote shear localisation and less stable sliding. A potential explanation of this  
614 difference to *Bedford and Faulkner* [2021] could be the initial maximum grain sizes of their  
615 samples of 5 - 30  $\mu\text{m}$ . As we start from a significantly larger maximum grain size of 125  $\mu\text{m}$ ,  
616 more strain is needed to accommodate similar degrees of localisation.

617 In addition to the grain size effect on localisation, grains that align towards R1-shears  
618 form force chains over the thickness of the gouge layer. The development characteristics of these  
619 chains were also found to affect frictional properties [e.g., *Anthony and Marone*, 2005; *Jiang et al.*,  
620 2016]. As the unaltered material contains fewer large grains over the same thickness  
621 compared to the altered material, the force chains can accommodate higher stresses, for which a  
622 stress release (e.g., due to mechanical failure of a grain in the chain) is related to less stable  
623 sliding [*Anthony and Marone*, 2005]. In contrast, the force chains in the altered material need to  
624 consist of significantly more grains to span the same layer thickness as the unaltered material.  
625 *Anthony and Marone* [2005] report, that such chains, composed of more, smaller grains, are  
626 weaker and more compliant to deformation. Stresses cannot build up as high and creep in the  
627 gouge happens more stably.

628 The development of friction parameters and microstructure are further affected by the  
629 chemical phase transformation from feldspars to clays which increases the clay fractions in the  
630 gouge. An increase in clay fraction has been found to stabilise sliding at fixed conditions [e.g.  
631 *Ashman and Faulkner*, 2022; *den Hartog and Spiers*, 2013; *Fang et al.*, 2018a; *Ikari et al.*, 2011;  
632 *Tembe et al.*, 2010]. With the phase change, the large feldspar grains are replaced by small platy  
633 clay grains. Although there are only small differences in the grain aspect ratios of our samples,  
634 the more altered rocks (TM6 and TM7) show tendencies towards more elongated grains than the  
635 least altered sample (TM11, see supplementary information). These elongated clays promote the  
636 formation of foliations and clay shear fabrics, which initially stabilise shear and allow easier  
637 orientation of grains in the direction of major shear structures. This is due to the low bond  
638 strength between the phyllosilicate sheets whose separation may further suppress dilatancy in the  
639 material [*Ikari et al.*, 2011].

640 In our experiments, we see more distinct preferred grain orientation in the direction of  
641 shear structures from least altered (TM11), over the intermediate altered (TM6 and Sa6), to the  
642 most altered stages (TM7 and TM8). Outside the fault core, the long axes of the grains orient  
643 clearer to shear structures with a higher degree of alteration (in the order of TM11 to TM6 and  
644 TM7), indicating an increase in pronounced shear fabrics. The least altered sample (TM11)  
645 contains the lowest amount of clays and shows orientations that are most independent of shear  
646 structures and closest to simple compression. The more altered the sample, the broader the grains  
647 orientate away from horizontal compression and towards P- and R1-shear directions (see TM6).  
648 The formation of pervasive undulating P-foliation fabric with shear is promoted for samples with  
649 higher clay content (Figure 8c). Finally, the sample containing the most clays (TM7) also  
650 contains the most distinct grain orientations towards P- and B/Y-orientation (see quantitative  
651 analysis in supplementary information). In this sample the fabric is most developed, stabilising  
652 sliding and decreasing frictional strength compared to TM11. The estimated porosity based on  
653 black values in the SEM images is lowest, hinting to least dilatational shear. As a result, we see  
654 the present argillic alteration promoting anisotropy of gouges by promoting the formation of  
655 shear fabrics and foliation, and with further displacement promoting formation of shear  
656 structures.

657 The fault core sample (TM8) diverges from the trends we found for the frictional  
658 behaviour from TM11 (unaltered) to TM7 (close to core). As reported earlier, TM8 contains the  
659 highest amount of quartz for all our samples because of an additional alteration process. This  
660 affects the grain size distribution, as the relative amount of stronger and potentially larger grains  
661 is increased while the relative amount of small clay particles is reduced. In addition to the size  
662 effect, the quartz grains are more rounded than clays and feldspars (also visible in aspect ratios  
663 analysed in supplementary information) and cause more evenly distributed grain-orientations  
664 than in the sample with the highest clay content (TM7). However, shear structures and clay  
665 fabric are still relatively well developed in the fault core sample (TM8) in comparison to the  
666 unaltered sample (TM11) due to the higher amount of small-grained matrix. This also affects  
667 frictional strength because the force chain network will develop similar to the other altered  
668 samples (TM6 and TM7). However, the fabrics are more localised in TM8 than in TM7,  
669 anastomosing around other grains and causing less stable sliding and higher frictional strength.  
670 In conclusion, larger quartz grains are probably responsible for higher strength and the less stable  
671 sliding than in the other highly altered sample (TM7), but sliding is still more stable in the fault  
672 core sample than in the unaltered sample (TM11) due to the larger proportion of clays.

673 A comparison of gouge samples from low and high temperature tests shows for the  
674 unaltered sample (TM11) that the post shear grain size distributions at high temperature contain  
675 more smaller grains and that the grains are more oriented towards the major shear structures. The  
676 difference in grain size could be related to granite becoming weaker with increasing temperature  
677 [e.g., *Yin et al.*, 2016]. This could indicate that during shear at higher temperatures more brittle  
678 failure occurs, generally resulting in more smaller grains. As this is simple grain size reduction  
679 without a phase transition (as it would be caused by argillic alteration), such a reduction  
680 promotes shear localisation and with this less stable sliding [e.g., *Bedford and Faulkner*, 2021].  
681 In contrast to the unaltered sample (TM11), this effect of temperature is not as visible in the high  
682 clay sample (TM7) because the grain sizes are mostly similar after shearing at low and the high  
683 temperature. However, the narrower distribution of orientations of grains in the 180°C gouge  
684 hint at a more developed fabric/foliation which could then cause more localised shear and  
685 therefore less stable sliding (see quantitative analysis in paper supplements).

686 The effects of temperature on the frictional behaviour have formerly been related to the  
687 competition of different microstructural processes. *Niemeijer and Spiers* [2007] explain  
688 temperature driven changes in friction in halite-muscovite mixtures by the competition of  
689 thermally activated compaction and granular flow causing dilation, for which temperature  
690 controls the mechanism that dominates deformation. In their three-regime-model pressure  
691 solution is suggested as thermally activated compaction mechanism. *den Hartog et al.* [2012]  
692 applied the *Niemeijer and Spiers* [2007] model to their experiments on crushed illite-rich  
693 Rochester shale and found close resemblance to their observed temperature dependence of  
694 friction and microstructure. However, *den Hartog et al.* [2012] also state that pressure solution  
695 would be insufficient to accommodate the required compaction at high temperatures under their  
696 used conditions. They suggest thermally activated stress corrosion cracking (SCC) as dominant  
697 deformation process in their experiments. The grain size distributions in our high temperature  
698 tests are shifted to smaller grain sizes compared to our room temperature equivalents which could  
699 imply a contribution of a thermally activated deformation mechanism. Similar to *den Hartog et*  
700 *al.* [2012], we assume that pure pressure solution would be insufficient to accommodate shear  
701 deformation under our used conditions as we see no evidence for it in our microstructures.  
702 Although we observe tailed apatite clasts in our samples, the tails consist of crushed material and

703 show no signs of dissolution or precipitation. The feldspars and quartz grains are more fractured,  
704 more angular, and smaller in our high temperature gouges which would imply SCC as acting  
705 process.

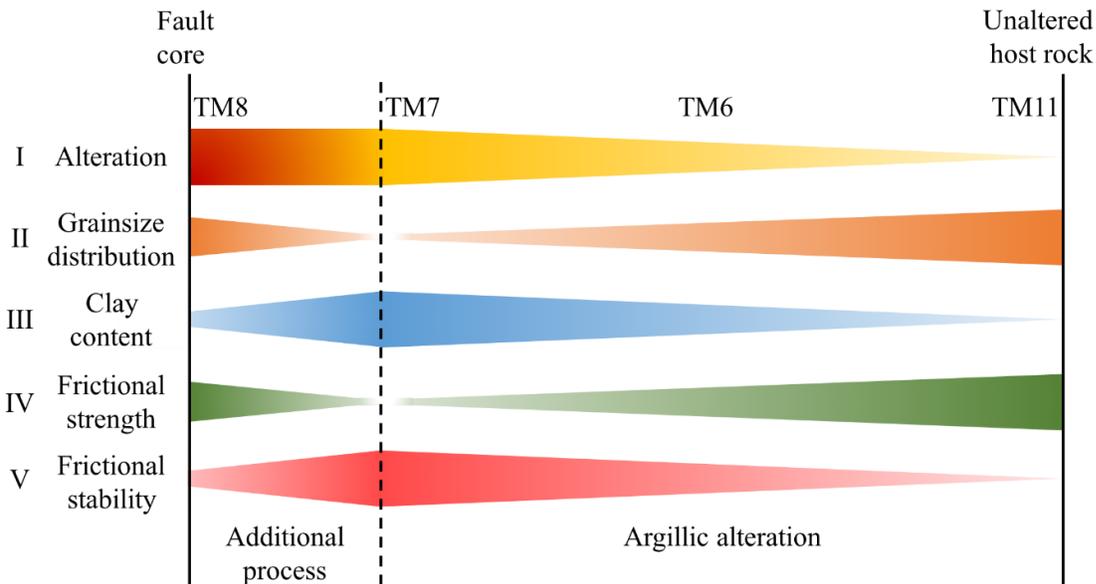
#### 706 4.4 Implications for fault behaviour in geothermal settings

707 Our room temperature results suggest that geochemical alteration stabilises slip in  
708 granitic fault systems. The closer the proximity to the fault core, the more altered it is likely to  
709 be, reducing frictional strength but increasing frictional stability (see TM11, TM6 and TM7 in  
710 Figure 10). A granitic gouge that initially exhibits velocity weakening behaviour is expected to  
711 shear more stably if plagioclase is replaced by clays, but due to the reduced strength, shear also  
712 becomes more likely. As more stable shear means a decrease in the likelihood of stick-slip events  
713 which are related to seismicity, shearing in more altered material is more likely to happen  
714 aseismic, contributing to destressing the system with less seismicity. The increase in clay content  
715 due to argillitic alteration also promotes the formation of shear fabrics and therefore the  
716 orientation of grains. These fabrics potentially make fault gouge more anisotropic and affect  
717 other physical and hydraulic properties [Kenigsberg *et al.*, 2019; Kenigsberg *et al.*, 2020].

718 We further found the trends with alteration to still be valid at up to 180°C, which is  
719 comparable to the bottom hole temperature at the UDDGP project. However, if alteration  
720 continues and additional processes reduce the amount of clays (like the integration of a quartz-  
721 vein or loss of fines due to flow along the), the remaining gouge will be enriched in stable grains  
722 like quartz (TM8). This could reduce fault stability over longer time scales. This accumulation of  
723 stable grains is limited by the fault permeability. A lower permeability will reduce overall  
724 geochemical reactions (i.e., dissolution and precipitation) by providing less fluid that is out of  
725 equilibrium with the host rock. Furthermore, fault stability is also controlled by the strength of  
726 the rock surrounding the core. If this rock is weaker due to higher amounts of clay, then slip and  
727 deformation will be concentrated where the rock is the weakest. The potential effects of  
728 temperature on fault friction were reported earlier [e.g., Blanpied *et al.*, 1998; den Hartog and  
729 Spiers, 2013]. Our findings mainly support these, as our tests at higher temperature showed  
730 sliding to become less stable from low temperature towards the velocity weakening window,  
731 while frictional strength decreased as well. Temperature had a similar effect on all samples,  
732 regardless of the composition, i.e. a higher or lower degree of alteration does not enhance or  
733 dampen any temperature effects. In turn this general temperature related shift of our data  
734 suggests that the stabilising effect of alteration on frictional stability is temperature independent,  
735 because the magnitude of the increase in stability with increasing alteration (from TM11, over  
736 TM6 to TM7) is about the same for our low and high temperature tests.

737 With respect to the UDDGP project and its suspected temperature range from 80 to  
738 180°C from injection to production well, the injection of new fluids or the reinjection of  
739 produced fluids could trigger further argillic alteration as it usually occurs between 100 and  
740 300°C [Pirajno, 2009]. Although the overlap of these temperature ranges does not include the  
741 80°C at the injection well, related lower temperature processes like kaolinization [Pyrillos *et al.*,  
742 2003] will have the same effect of replacing feldspars with clay minerals closer to the injection  
743 well. Over longer periods of time, this can then widen the alteration zones around conductive  
744 faults and gradually reduce frictional strength while increasing frictional stability. Assuming a  
745 wider alteration zone also means a wider fault or potentially thicker fault gouge layer involved in  
746 the accommodation of displacement, this widening has an additional stabilising effect as thicker

747 gouge layers were found show more stable shearing than thin ones [Byerlee and Summers, 1976;  
748 Marone *et al.*, 1990].



749 **Figure 10: Schematic of the implications of our results on frictional behaviour. The wider the beam: (I) the**  
750 **more developed the rock alteration state, (II) the higher the amount of large grains, (III) the higher the clay**  
751 **content, (IV) the higher frictional strength and (V) the higher frictional stability. The colour change in**  
752 **alteration (I) represents an additional process necessary to explain frictional changes towards the fault centre.**  
753

## 754 5 Conclusions

755 A series of direct shear experiments at room temperature and in-situ pressure conditions  
756 were conducted on granite gouges to investigate the effects of chemical alteration on the  
757 frictional behaviour in granitic fault systems. We used Carnmenellis granite samples from a fault  
758 (cross-course structure) in Cornwall that exhibit different degrees of natural alteration. Samples  
759 that underwent a higher degree of alteration show higher amounts of clay minerals (kaolinite,  
760 smectite, chlorite) and lower amounts of plagioclase. Frictional strength of the samples decreases  
761 with increasing proximity to the fault core except for the sample closest to it. The sample  
762 collected from within the fault core contains higher amounts of quartz in comparison to the other  
763 samples which we relate either to removal of other minerals dissolved as small particles in fluid  
764 or the integration of material from a small quartz vein into the gouge. Frictional stabilities of the  
765 samples change from velocity-weakening in the least altered sample to velocity-strengthening in  
766 the more altered samples. The fault core sample shows less velocity-strengthening behaviour  
767 than two less altered samples, presumably due to the increase in quartz content. Microstructural  
768 analysis of the samples relates alteration and increasing frictional stability to delocalisation of  
769 shear fabrics due to reduction of grain size and increase in clay content. Tests at 180°C showed a  
770 general decrease in sliding stability compared to the low temperature tests, while the intensity of  
771 the decrease was independent of alteration stage. From our results we suggest that argillic  
772 alteration around faults generally increases the likelihood of slip to occur but stabilises slip  
773 towards the fault core, destressing the system by promoting aseismic shear. However, additional  
774 effects of altered or dissolved material may complicate this pattern.

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782 **Conflict of interest**

783 The authors have no conflicts to disclose.

784 **Availability Statement**

785 The data that supports the findings of this study are available within the article and the  
 786 supporting information.

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