# Grain fragmentation and frictional melting during initial experimental deformation and implications for seismic slip at shallow depths

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- 4 Chien-Cheng Hung<sup>1#†</sup>, Li-Wei Kuo<sup>1\*#</sup>, Elena Spagnuolo<sup>2</sup>, Chun-Chieh Wang<sup>3</sup>, Giulio
- 5 Di Toro<sup>2,4</sup>, Wen-Jie Wu<sup>1</sup>, Jia-Jyun Dong<sup>5</sup>, Wayne Lin<sup>6</sup>, Hwo-Shuenn Sheu<sup>3</sup>, En-Chao
- 6 Yeh<sup>7</sup>, Pei-Shan Hsieh<sup>6</sup>
- 7
- 8 <sup>1</sup>Department of Earth Sciences, National Central University, Taoyuan, Taiwan
- 9 <sup>2</sup>Istituto Nazionale di Geofisica e Vulcanologia, Rome, Italy
- 10 <sup>3</sup>National Synchrotron Radiation Research Center, Hsinchu, Taiwan
- <sup>4</sup>Dipartimento di Geoscienze, Università di Padova, Padua, Italy
- <sup>5</sup>Graduate Institute of Applied Geology, National Central University, Taoyuan, Taiwan
- 13 <sup>6</sup>Industrial Technology Research Institute, Hsinchu, Taiwan
- <sup>7</sup>Department of Earth Sciences, National Taiwan Normal University, Taipei, Taiwan
- 15
- 16 \*corresponding author
- 17 Address: No. 300, JhongDa Rd., Jhongli District, Taoyuan City 32001, Taiwan
- 18
   Tel: +886-3-4227151
   ext:65628
   Fax: +886-3-4222044
- 19 E-mail: liweikuo@ncu.edu.tw; liweikuo@gmail.com
- 20

21 <sup>†</sup> now at Earth Simulation Laboratory, Department of Earth Sciences, Utrecht

- 22 University, Utrecht, The Netherlands
- 23 <sup>#</sup> equal contribution

24

#### 25 Abstract

26 During seismic slip, the elastic strain energy released by the wall rocks drives grain 27 fragmentation and flash heating in the slipping zone, resulting in formation of 28 (nano)powders and melt droplets, which lower the fault resistance. With progressive 29 seismic slip, the frictional melt covers the slip surface and behaves as a lubricant 30 reducing the coseismic fault strength. However, the processes associated to the 31 transition from grain fragmentation to bulk frictional melting remain poorly 32 understood. Here we discuss in-situ micro-analytical investigations performed on 33 experimentally produced solidified frictional melts from the transition regime 34 between grain fragmentation and frictional melting. The experiments were performed 35 on granitic gneiss at seismic slip rates (1.3 and 5 m/s), normal stresses ranging from 3 36 to 30 MPa. At normal stresses < 12 MPa, the apparent friction coefficient  $\mu_{app}$  (shear 37 stress vs. normal stress) evolves in a complex manner with slip:  $\mu_{app}$  decreases 38 because of flash weakening, increases up to a peak value  $\mu_{p1} \sim 0.6-1.0$ , slightly 39 decreases and increases again to a second peak value  $\mu_{p2} \sim 0.44-0.83$ , and eventually 40 decreases with displacement to a steady-state value  $\mu_{ss} \sim 0.3-0.45$ . In-situ synchrotron 41 observations of the solidified frictional melt show abundance of ultra-fine quartz 42 grains before  $\mu_{p2}$  and enrichment in SiO<sub>2</sub> at  $\mu_{p2}$ . Because partial melting occurs on the 43 ultra-fine quartz grains and, as a consequence, it suggested that the second 44 re-strengthening  $(\mu_{p2})$  is induced by the higher viscosity of the melt due to its 45 enrichment in Si from melting of the ultra-fine quartz grains derived from grain 46 fragmentation.

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48 Keyword: frictional melting, grain fragmentation, ultrafine quartz, viscosity,

49 synchrotron analysis.

#### 50 1. Introduction

51 Numerous physical and chemical processes have been theoretically and 52 experimentally proposed to justify fault lubrication during seismic slip, for example, 53 flash heating (Rice, 2006; Beeler et al., 2008; Goldsby and Tullis, 2011), powder 54 lubrication (Han et al., 2010; Reches and Lockner, 2010), frictional melting (Spray, 55 2005; Di Toro et al., 2006a), silica gel formation (Di Toro et al., 2004), 56 elastohydrodynamic lubrication (Brodsky and Kanamori, 2001; Cornelio et al., 2019), 57 grain size- and temperature-dependent processes (Green et al., 2015; De Paola et al., 58 2015; Spagnuolo et al., 2015; Rowe et al., 2019) and thermal decomposition (Han et 59 al., 2007; Collettini et al., 2013). In particular, on a fault patch in silicate-built rocks, 60 flash heating and melting (Goldsby and Tullis, 2011) or grain fragmentation of the 61 rock (Green et al., 2015; De Paola et al., 2015; Spagnuolo et al., 2015; Rowe et al., 62 2019; Chen et al., 2017a) may occur during the initial stages of seismic slip at the 63 passage of the earthquake rupture propagation front. With progressive slip, the 64 continuously generated melt droplets can accumulate to form a continuous melt layer 65 possibly resulting in melt lubrication (Spray, 1995; Hirose and Shimamoto, 2005). 66 Therefore, studies on powder generation, flash heating and melting, and the formation of a continuous melt layer provided criteria for understanding fault lubrication 67 68 (Reches and Lockner, 2010; Chen et al., 2017a; Spray, 1995; 2005; Di Toro et al., 69 2006; Rice, 2006; Beeler et al., 2008; Goldsby and Tullis, 2011).

However, the transition between fault rock comminution and frictional melting remains unclear. More relevantly, how do the associated frictional properties derived from both fault comminution and frictional melting affect the fault strength during fault rupture is not well-constrained. Here, we address these questions by conducting rock friction experiments on granitic gneiss at seismic rates and further discuss how the presence of fragmented grains and the chemical evolution of the friction melt influence the frictional behavior of the experimental fault.

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#### 78 2. Materials and Methods

#### 79 2.1 Starting materials

80 The exhumed pseudotachylyte-bearing fault, which crosscuts the granitic gneiss 81 of the Tananao Metamorphic Complex (TMC), crops out in the Hoping area, northeast 82 Taiwan (Chu et al., 2012; Fig. 1a). The fault zone rocks are mainly cataclasites and 83 mylonites overprinted by pseudotachylytes. Pseudotachylytes were generated at depths > 4 km about 1.6 Ma ago (Chen et al., 2017) in faults accommodating 84 displacements by up to 220 mm (corresponding to earthquakes of  $M_W 6.4 \pm 0.4$  if all 85 86 the displacement would be associated to a single seismic slip event; Korren et al., 87 2015). Pseudotachylytes form vein and injection veins networks that intrude the host

88 granitic gneiss, and they were used to infer earthquake source parameters (e.g., slip 89 vectors, Korren et al., 2015; brushlines, Ferré et al., 2016). We used the granitic gneiss 90 from the Hoping borehole cores drilled by the Industrial Technology Research 91 Institute (ITRI) as the starting materials for the rotary shear experiments (Fig. 1b and 92 1c). The mineral composition of the core granitic gneiss is 35-40% quartz, 15-25%93 feldspar, 20–25% micas (muscovite dominant) and 5–10% other accessory minerals 94 (estimated by optical microscope analysis of thin sections). This estimated 95 composition is similar to the modal content of the granitic gneiss cropping out in the 96 area with 30-35% quartz, 20-25% feldspar, 30-35% micas (both muscovite and 97 biotite), 3-5% clinozoisite-epidote and titanite, as well as 2% of other accessory 98 minerals (Chu et al., 2012). The bulk chemical composition of the granitic gneiss is 99 also shown in Table 1. The granitic gneiss has a heterogeneous mineralogical 100 distribution due to the presence of a foliation enriched in muscovite or in quartz and 101 feldspar (Fig. 1b and 1c). To achieve more homogenous mineralogy of the sliding 102 surface, the samples prepared for the experiments were drilled with the long axis 103 oblique (approximately  $25^{\circ}$  to  $30^{\circ}$ ) to the gneissic foliation.

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#### 105 2.2 Rock deformation experiments

Rock deformation experiments were conducted with the low-to-high velocity
rotary shear friction apparatus (LHVR) installed at the National Central University
(NCU), Taiwan (Yang et al., 2014; Kuo et al., 2015), and the slow-to-high velocity
apparatus (SHIVA) at the Istituto Nazionale di Geofisica e Vulcanologia (INGV,
Rome), Italy (Di Toro et al., 2010). The latter was used to extend the applied normal
stresses up to 30 MPa and perform an experiment under vacuum conditions.

112 For LHVR, the samples were solid cylinders (25 mm external diameter) which 113 were tightly confined with iron wires (Fig. 1b). For SHIVA, the samples were hollow 114 cylinders (50 mm external, 30 mm internal diameters) which were jacketed with an 115 external aluminum ring (Fig. 1c; Niemeijer et al., 2011; Nielsen et al., 2012). To 116 improve the alignment of two mounted samples before the experiments, for LHVR, 117 we gently knocked the edge of the upper one until the measured un-centered degree 118 was lower than 10  $\mu$ m, and, for SHIVA, we pre-ground the slip surface of the samples at 1 cm/s and 1 MPa (Niemeijer et al., 2011). The pre-grinding lasted until the 119 120 recorded torque achieved a constant value which is associated with the parallelism of 121 the two opposite sliding surfaces.

Because of the rotary configuration of the two machines used, both slip and slip rate increase with sample radius. As a consequence, we define the equivalent slip rate  $V_e$  (Shimamoto and Tsutsumi, 1994; Hirose and Shimamoto, 2005):

125 
$$V_e = \frac{4\pi R(r_{ext}^2 + r_{ext}r_{int} + r_{ext}^2)}{3(r_{ext} + r_{int})} \left[\frac{m}{s}\right]$$
 Eq. 1

126 where R is the revolution rate of the motor (i.e., the target R in the experiments 127 presented here were 1500 rpm for LHVR and 3000 rpm for SHIVA), rext the external 128 radius (12.5 and 25 mm for LHVR and SHIVA, respectively), and  $r_{int}$  the internal 129 radius (0 mm and 15 mm for LHVR and SHIVA, respectively) of the samples (Fig. 1). 130 The experimental conditions above resulted in target  $V_{\rm e}$  of 1.3 m/s and 5 m/s for 131 LHVR and SHIVA, respectively. Hereafter we refer to the "equivalent slip rate" as 132 slip rate. The measured torque (T) allows us to determine the shear stress ( $\tau$ ) by 133 assuming that  $\tau$  is constant over the entire slip surface (Hirose and Shimamoto, 2005):

134 
$$T = \int_{r_{int}}^{r_{ext}} 2\pi \tau r^2 dr = \frac{2\pi \tau}{3} (r_{ext}^3 - r_{int}^3) [N m]$$
 Eq. 2

Given the strong assumption of a constant shear stress over the entire slip surface, which cannot be the case as the shear stress varies with slip rate (e.g., Di Toro et al., 2011) and in the cylindrical configuration the slip rate increases with the sample radius, mechanical data obtained with hollow cylinders are more accurate than those obtained with solid cylinders. From Eq. 2, the friction coefficient  $\mu$  is:

140 
$$\mu = \frac{\tau}{\sigma} = \frac{3T}{2\pi\sigma(r_{ext}^3 - r_{int}^3)}$$
 Eq. 3

141 hereafter we refer to  $\mu$  as apparent friction coefficient  $\mu_{app}$  for the following 142 description.

143 The LHVR experiments were conducted at normal stresses of 3, 6, 9, 12, and 19 144 MPa, at a target slip rate of 1.3 m/s, and under room humidity conditions. The SHIVA 145 experiments were conducted at the normal stresses of 3 and 30 MPa, respectively, and 146 at a target slip rate of 5 m/s under both vacuum and room humidity conditions (Table 147 2). All mechanical data (torque, angular rotation, and axial displacement) were 148 acquired at a frequency up to 1 kHz and 25 kHz for LHVR and SHIVA, respectively. 149 The friction coefficient of the granitic gneiss evolved in a complex manner with slip. 150 Because of this, we conducted slip-stepping experiments or experiments that were 151 stopped at the about the maximum or minimum values of the friction coefficient.

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#### 153 2.3 Analytical methods

 We impregnated the experimental products of LHVR with epoxy resin and prepared petrographic thin sections and rock slices, both cut across the diameter of the sample and perpendicular to slipping surface. The petrographic thin sections were prepared with a thickness of 30 μm, mounted on silica glass and polished by 0.3 μm thick alumina powder for optical and scanning electron microscopy analysis. The thickness of rock slices were ~ 1 mm for *in-situ* synchrotron X-ray diffraction (XRD) and micro-Raman spectroscopy analysis.

161 2. Three-lens optical microscopy (OM) (Leica DMLP) coupled with an image

162 capture system, and field emission scanning electron microscopy, coupled with 163 an energy dispersive spectrometer (FESEM/EDX) (JSM-7000F model) equipped 164 at the National Central University, Taiwan, were used to investigate the 165 microstructures and determine the semi-quantitative chemical composition of the 166 solidified frictional melt. For SEM-EDX, the petrographic sections were 167 sputtered with platinum at 5 nm thickness and semi-quantitatively analyzed at 15 168 kV with a focused beam of  $\sim 1 \ \mu m$  in diameter.

169 The *in-situ* synchrotron XRD installed at the beamline BL01C2 in the National 3. 170 Synchrotron Radiation Research Center (NSRRC), Taiwan, was used to 171 determine the mineral assemblages of both the solidified frictional melt and the 172 host rock. The analyses were operated at a wavelength of 0.774910 Å, with an 173 electron beam energy of 1.5 GeV and a beam size of 150 µm in diameter. 174 Because of the presence of large grains ( $\sim 1.0$  mm in size) of quartz, the samples 175 of gneiss, differently from the solidified frictional melts, were reduced into 176 powders and analyzed. This prevented the presence of intensity anomalies in the 177 XRD spectra due to the occurrence of crystals (e.g., quartz) larger than the X-ray 178 beam (Kuo et al., 2014a, b). However, the intensity anomalies can still be 179 observed in the results of the glass matrix owing to the presence of large survivor 180 quartz or feldspar grains. To reduce the effect of intensity anomalies and 181 representatively present the synchrotron XRD data, the spectra were reported as 182 the average values of three analyses on the same sample.

4. The micro-Raman spectroscopy (Horiba Jobin Yvon UV–VIS Labram HR) at the
National Taiwan Museum was used to determine the presence of water within the
solidified frictional melt. We focused on the region from 100 to 4,000 cm<sup>-1</sup> on
the Raman spectrum. The analysis was operated with a 532 nm laser as an

187 excitation source with a beam size of  $1-2 \mu m$  in diameter under a  $\times 100$  objective

with a final laser power of 100 mW on the sample surface. A charged-coupled
device (CCD) detector was installed in the microscope used to focus the
excitation laser beam to collect the backscattered Raman signal. We collected
three data of the matrix per deformed sample for the solidified frictional melt at
an acquisition time of 15 s per analysis. We added the Raman spectra of pure
water and quartz for comparison.

5. The synchrotron transmission X-ray microscopy (TXM) at the beamline BL01B1
(Song et al., 2007a, b; Yin et al., 2006) at the NSRRC, Taiwan, was used to
obtain X-ray radiographies to determine the particle size distribution (PSD) of
the survivor clasts hosted in the solidified frictional melt. The analysis was
conducted on a 30-µm-thick sample collected from the petrographic thin sections

- over an area of 375 μm x 75 μm. The beam X-ray energy was 8 keV and the
  analysis had a spatial resolution of 50-nm (see Song et al., 2007a, b).
- 201 The software ImageJ (available at https://imagej.nih.gov/ij/) was used to process 6. 202 the images of both the back scattering electron (BSE) and the TXM to estimate 203 the PSD of the survivor grains within the solidified frictional melt. ImageJ 204 allowed us to measure the size of quartz grains by exploiting the gray contrast 205 between the glass matrix and the quartz grains in the BSE images (Kuo et al., 206 2015). On the contrary, we draw by hand the survivor grains from the TXM 207 images due to the difficulty of the contrast identification between glass matrix 208 and grains. By doing this, the overlapping of the survivor grains could be also 209 clearly identified.
- 210 7. One experiment (LHVR1097) performed on granitic gneiss from the outcrop was 211 recorded with an infrared thermal-sensing camera (thermoIMAGER TIM160, 212 MICRO-EPSILON) to measure the temperature evolution of the experimental 213 fault with slip. The infrared camera, integrated with the TIMConnect software, 214 provided an instantaneous measurement of the temperature from 150°C to 900°C 215 over an area of 0.6 mm in diameter at an acquisition rate of 10-500 Hz. Since the 216 slipping zone is < 0.3 mm in thickness and the temperature is measured from 217 outside, the measured temperatures yielded a minimum estimate of the 218 temperatures achieved in the slipping zone.
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### 220 **3. Results**

### 221 3.1 Frictional behavior and temperature evolution during fast shear experiments

The mechanical data exhibit a complicated evolution of  $\mu_{app}$  with slip, which varied with normal stress (Fig. 2a). At slip initiation, once the static friction  $\mu_s$  was overcome,  $\mu_{app}$  decreased rapidly to a first minimum value (possibly associated with flash heating and weakening, see discussion). Then the frictional evolution varied with normal stress:

227 <u>At large normal stresses</u> ( $\geq$  12 MPa, e.g., experiment s1289, Fig. 2a)  $\mu_{app}$ 228 increased to a peak value  $\mu_{p1} = 0.66$  and then dramatically decreased to a low value of 229  $0.16 \pm 0.01$  (an average value of friction coefficient after peak friction and before the 230 sample was broken) defined in the literature as steady state friction  $\mu_{ss}$  (e.g., Di Toro 231 et al., 2011). At the end of slip, during sample deceleration,  $\mu_{app}$  increased again. In 232 general, this frictional evolution with slip is consistent with previous experiments 233 performed on silicate-built cohesive rocks sheared at seismic slip rates (Hirose and 234 Shimamoto, 2005; Di Toro et al., 2006a, b; Niemeijer et al., 2011; Passelègue et al., 235 2016).

236 <u>At low normal stresses</u> (< 12 MPa, e.g., experiments LHVR 0438 and

LHVR0452, Fig. 2a and 3), after slip initiation and initial flash weakening,  $\mu_{app}$  first increased to a peak value at  $\mu_{p1}$ ~1.0, then decreased to 0.8-0.5, then increased again to a second peak value at  $\mu_{p2} \sim 0.44$ -0.83 and eventually decreased with displacement to  $\mu_{ss} \sim 0.3 \pm 0.01$ -0.45 ± 0.08. Then, similarly to the large normal stress experiments,  $\mu_{app}$  increased again at the end of slip (Fig. 2a). This complicated evolution, though observed in previous experiments, has been only partly discussed (e.g., Hirose and Shimamoto, 2005; Di Toro et al., 2006).

With the aim of investigating the process responsible for the  $\mu_{p1}$  and  $\mu_{p2}$ , we analyzed the slipping zone produced in dedicated experiments stopped, with increasing slip (slip-stepping experiments, Fig. 3), after:

247 (1) 1.5 m of slip corresponding to  $\mu_{p1}$  (State A, experiment LHVR0487);

248 (2) 2.6 m of slip corresponding to the frictional strength decrease between  $\mu_{p1}$  and  $\mu_{p2}$ 

249 (State B, experiment LHVR0534);

250 (3) 2.1 m of slip corresponding to ca.  $\mu_{p2}$  (State C, experiment LHVR0452);

(4) 5.7 m of slip during the final weakening (State D, "steady-state" conditions,
experiment LHVR0438).

253 To illustrate the temperature evolution with slip for loading conditions identical 254 to those imposed in the reference experiment LHVR0438 where "steady-state" 255 conditions were achieved, experiment LHVR1097 was equipped with an infrared 256 thermal-sensing camera. Because the emissivity  $\varepsilon$  of melt varies with temperature, we 257 set two values of  $\varepsilon$ , 0.55 and 0.9 for high and low temperature, respectively (Abtahi et 258 al., 2002). For  $\varepsilon = 0.55$ , the temperature of the slipping zone immediately increased to 259  $\sim$ 430°C (ca. 0.6 m of slip in Fig. 3e), slightly rose to  $\sim$ 500°C at State A (ca. 1.2 m of slip), slightly decreased and remained a constant value of ~450°C during State B 260 261 (between 1.2 and 2 m of slip), steadily rose to  $\sim$ 750°C at State C (between 2 and 3 m 262 of slip), and eventually increased to a constant value of ~850°C during State D (from 263 3 m to the end of slip). There was a loss of temperature measurements (between  $\sim$ 3.5 264 to 4 m of slip) because of connection issues with the infrared camera. For  $\varepsilon = 0.9$ , 265 with respect to the estimated temperatures for  $\varepsilon = 0.55$ , temperatures achieved in the 266 different States were lower by about ~100°C for States A and B, ~200°C for State C and  $\sim 250^{\circ}$ C for State D. 267

Regarding the entire experimental dataset, the initial peak (corresponding to  $\mu_{p1}$ ) and steady-state (corresponding to  $\mu_{ss}$ ) shear stress increased linearly with normal stress, resulting in an effective friction coefficient of ~0.3 and ~0.11 for  $\mu_{p1}$  and  $\mu_{ss}$ , respectively (Fig. 2b). Instead, the second  $\mu_{p2}$  disappeared with increasing normal stress (Fig. 2a).

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274 3.2 Microstructures, mineralogy and chemical composition of the solidified

#### 275 frictional melt

276 We recovered the experimental products of the slip-stepping experiments and 277 investigated with OM, FESEM-EDX, synchrotron XRD and synchrotron TXM. The 278 mineralogy and microstructures of the slipping zones are shown in Figures 4, 5, and 6. 279 In general, the thickness of the experimental slipping zones increases from the edge 280 (were the slip rate is the highest) to the center (were the slip rate is nominally zero) of 281 the circular slip surface (Fig. 4a to d). In the experiment stopped at State A, the 282 solidified melt patches were discontinuous and meniscus-shaped (Fig. 4). In the 283 experiments stopped at states B and C, the solidified melt formed a continuous and 284 dark-coloured donut-shaped feature. In the experiment stopped at State D, the 285 solidified melt covered the entire slip surface (Fig. 4). In addition, the boundary 286 solidified melt / host rock was irregular (presence of embayments, indicated by 287 arrows in Figs. 4a-d) due to selective melting of the low-melting-point minerals (e.g., 288 muscovite) of the granitic gneiss (see Hirose and Shimamoto, 2003; Fig. 4d). The 289 average thickness of the solidified frictional melt layers was determined by averaging 290 the value of the real melt thickness obtained at different horizontal positions of the 291 solidified melt on the slip surface. Although this measurement might result in an 292 underestimate value as it includes the extrusion of melt at the end of slip, the melt 293 thickness still notably varied at each state from  $\sim 270 \ \mu m$  in State A,  $\sim 330 \ \mu m$  in 294 State B,  $\sim 170 \,\mu\text{m}$  in State C, and  $\sim 230 \,\mu\text{m}$  in State D (Table 2).

295 The *in-situ* synchrotron XRD data show that the mineral assemblage of the host 296 rock (black line) was composed of quartz, feldspar, calcite, and muscovite (Fig. 4e). 297 Instead, the clasts suspended in the solidified frictional melts produced in the 298 slip-stepping experiments were mainly composed of quartz and minor feldspar 299 (Fig.4e). As observed in both natural and experimental pseudotachylytes, this 300 indicates that quartz is more resistant than feldspar and muscovite during 301 comminution and melting (see Spray, 2010, for a review). Unfortunately, the signals 302 of the presence of amorphous materials (= glass matrix of the pseudotachylytes) 303 between 15° to 40° of  $2\theta$  were not detected in the synchrotron XRD spectra because 304 of the strong signal depression due to the high peaks of quartz.

305 Under the FESEM-BSE, the grains of the undeformed Hoping granitic gneiss are 306 angular and elongated in shape, with grain size varying from  $\sim 0.1$  to  $\sim 1.0$  mm (not 307 shown). The representative BSE images of the solidified melts under identical 308 magnification show a substantial amount of sub-angular quartz grains and the 309 presence of vesicles distributed within the glass matrix (Fig. 5). It is clear that the 310 grain size distribution of the survivor grains of quartz varies significantly over the 311 four states of the slip-stepping experiments, e.g., grains  $< 5 \mu m$  in size in State C are 312 more abundant than in both States B and D (Fig. 5 and section 3.5).

313 The TXM images show the overlapping of rounded to sub-rounded survivor 314 grains which ranged from 50 nm to 10 µm in diameter within the solidified melts 315 representative of the four states of the slip-stepping experiments (Fig. 6). For grains < 316  $5 \,\mu\text{m}$  (size range that is difficult to be clearly observed under SEM-BSE), we find that 317 the shapes of the survivor grains in the TXM images were in rounded or spherical 318 shape, similar occurrence to the survivor quartz grains reported from Kuo et al. (2015). 319 These results suggest that partial melting on the edge of grains likely occurred, in 320 particular, for ultra-fine grains (Kuo et al., 2015).

321 We utilized FESEM/EDS to analyze the matrix of the solidified frictional melts 322 and adopted the average value of 40 points as representative of the chemical 323 composition of the matrix (Table 1). The analysis showed no significant chemical 324 variation of the composition of the solidified frictional melts from any of the four 325 states of the slip-stepping experiments, except for the enrichment in SiO<sub>2</sub>-content 326 from State A to State C and particular high CaO-content in State D. The Raman 327 spectra of the slip-stepping experiments showed negligible water content in the 328 solidified frictional melts (Fig. 7).

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## **330 3.3** Frictional melt viscosity and temperature estimate

331 In addition to the measured temperature of the frictional melt during the 332 experiment LHVR1097, we also estimated the possible viscosity and temperature of 333 the friction melt in the four states following the method suggested by Wallace et al. 334 (2019). The non-Arrhenian Newtonian temperature-dependence viscosity of the 335 silicate melt can be estimated by the GRD viscosity calculator (available online at 336 https://www.eoas.ubc.ca/~krussell/VISCOSITY/grdViscosity.html) by inputting the 337 melt composition (table 1; Giordano et al., 2008; Lavallée et al., 2012). This 338 calculation is based on the Vogel-Fulcher-Tamman equation:

339 
$$Log \eta = A + \frac{B}{T(K) - C}$$
 Eq. 5

340 where the  $\eta$  is viscosity (Pa s), A, B, and C, are the modelled parameters based on 341 viscosity measurement of geochemical composition (Giordano et al., 2008), and T the 342 silicate melt temperature (K). We obtained the viscosity curves for the slip-stepping 343 frictional melts (Fig. 8) by giving at the constant A the value of -4.55 (optimal value 344 based on Giordano et al. (2008)), and the values of B and C were obtained from the 345 chemical composition of the glasses and 17 adjustable parameters (see demonstration 346 calculation in Giordano et al. (2008), Table 1). The presence of vesicles suggests that 347 some volatiles (e.g., OH from muscovite) were present in the melt, but the water 348 content in the glass was likely negligible and assumed to be zero based on the absence 349 of water signal from Raman analysis (Fig. 7).

Based on the mechanical data, the apparent viscosity of the frictional melts ( $\eta_{app}$ ) can be directly calculated by the ratio of the measured shear stress to the strain rate as:

352 
$$\eta_{app} = \frac{\tau}{\dot{\varepsilon}} = \frac{\tau w}{v_e} [Pa \ s]$$
 Eq.6

where  $\tau$  is the measured shear stress (Pa),  $\dot{\varepsilon}$  is the strain rate (s<sup>-1</sup>), *w* is the thickness of the melt layer (an underestimate of the thickness during sliding as it includes the extrusion of melt at the end of slip) and  $V_e$  is the equivalent slip velocity (m/s). Because solid particles (= survivor clasts) are present in the frictional melts, the calculated apparent viscosity could be overestimated (e.g., Metzner, 1985). To correct the effect of clasts on the apparent viscosity, we calculated the relative viscosity ( $\eta_r$ ) using an empirical equation (Kitano et al., 1981):

360 
$$\eta_r = (1 - \frac{\phi}{A})^{-2}$$
 Eq. 7

with A = 0.54 - 0.0125r, where  $\phi$  is the volume fraction (volume particles/total volume)

362 of the clasts, A is the parameter related to the packing geometry of the solid particles 363 and r is the average aspect ratio of solid grains (about 2 in our case). We measured the 364  $\phi$  using BSE images from the center to the edge of the experimental pseudotachylyte 365 and averaged the value. The value of  $\phi$  ranged from ~0.11 at State C to ~0.20 at State 366 A, B, and D (Table 2). Moreover, the presence of vesicles should lower the apparent 367 viscosity of the frictional melt (e.g. Manga et al., 1998). However, the volume fraction 368 of the vesicles is small (< 0.1) and shows no significant variation in the four states. 369 Therefore, the contribution of the vesicles to the apparent viscosity should be 370 negligible (Lejeune et al., 1999). Based on Eqs. 6 and 7, the mechanically-constraint 371 apparent viscosities corrected with the presence of clasts ( $\eta_c = \eta_{app} / \eta_r$ ) are ~229 Pa s 372 at State A, ~232 Pa s at State B, ~125 Pa s at State C, and ~99 Pa s at State D (Table 373 2).

According to the model for Non-Arrhenian temperature-dependence of melt viscosity (Eqs. 5; Wallace et al., 2019), the chemically- and mechanically-constraint apparent viscosities for the four states are consistent with the temperature of ~1370°C (State A), ~1300°C (State B), ~1456°C (State C), and ~1265°C (State D) (Fig. 8). These chemo-mechanical-constraint temperature estimates are at odds with the temperature measured estimated with the infrared camera, as it will be discussed in section 4.3.

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#### **382 3.4 Particle size distribution (PSD)**

383 The PSD was obtained by measuring all clasts (mainly made of quartz) within

384 the solidified frictional melt from the images of both SEM and TXM. With the 385 FESEM, for each state, we collected eight to twelve BSE contiguous images to cover 386 almost entirely slipping zone. Moreover, we collected some images at higher 387 magnification (1,300X, similar to Fig. 5) to obtain more accurate size distribution on 388 finer grains ( $< 10 \, \mu m$ ). With the TXM, for each state, we analyzed identical areas of 389  $375 \,\mu\text{m}$  in length and 75  $\mu\text{m}$  in width, corresponding to the central part of the slipping 390 zone. Since the TXM images included all the grains over the entire thickness (30  $\mu$ m) 391 of the thin-section, the number of particles measured from the TXM images has different unit compared to that from BSE images (i.e. number of particles per  $\mu m^2$  for 392 SEM and per  $\mu m^3$  for TXM). Therefore, we further divided the obtained data from the 393 TXM images by the sample thickness (µm) to integrate it with the PSD data from the 394 395 BSE images. To increase the accuracy of the density data in size range of  $1-3 \mu m$ 396 (lower limit of SEM analysis), we extend the TXM results to overlap this size range. 397 The density data in this overlapping range is reported as the average value of both 398 TXM and SEM results.

The PSD data were plotted as a cumulative frequency diagram for the mean diameter, *r*, using a logarithmic scale. Our results (solid dots) were fitted and plotted with solid curves for mean diameters > 1  $\mu$ m and as dashed lines for diameters < 1  $\mu$ m. The particle density, *N*(*r*), which represents the cumulative number of particles per  $\mu$ m<sup>2</sup>, follows a function of mean diameter (*r*), defined as the geometric mean of the

404 major and minor axes of an irregular grain, in the form of a power-law for frictional

405 melting (not comminution):  $N(r) = N'(1 + r/r')^{-D}$ , where N is the cumulative number 406 of grain sizes greater than r, that is, N' (which depends on the total number of 407 measurements), r' is constant, and D is the fractal dimension (Shimamoto and 408 Nagahama, 1992; Tsutsumi, 1999).

409 The fitted curves for each state are almost identical, i.e., the curves are steep in 410 the coarse-grained size ranges (> 10  $\mu$ m) and gently flatten towards fine size < 10  $\mu$ m, 411 and to some degree the PSDs obey to a power-law (Fig. 9). Interestingly, we find that 412 the measured particle density for the grain sizes smaller than 1 µm is larger than the 413 fitted curves, in particular, at states A and B (Fig. 9). Based on the previous PSD 414 studies of the experimentally produced pseudotachylytes, the slope of the PSD curves 415 shows a linearly steep to horizontal evolution in the size range from about 100 µm to 416 0.5 µm (e.g. Tsutusmi, 1999, Fig. 2). These curves were obtained from the solidified 417 melt with long displacement experiments (hundreds of meters) and the change of the 418 slope was suggested to be the effect of partial melting of the survivor grains. 419 Therefore, compared to our results obtained from the short-displacement experimental 420 melt (< 3 m), the deviation between measured density data and fitted curves in size <

421 1 μm, particularly significant in States A and B, may suggest that the ultra-fine quartz 422 grains derived from the surface comminution are trapped in the melts. Instead, the 423 PSD of the clasts from State C is described by a larger value for D. The reduced 424 thickness of the slipping zone during frictional melting is one of the factors 425 controlling the size reduction of the clasts in the coarse-grained range and, as a 426 consequence, the fractal dimension of their distribution (Tsutsumi, 1999). In addition, 427 thickness of the melt zone might strongly depend on the viscosity and thus on the 428 temperature of the melt layer. This may suggest the high value of D at State C might 429 be controlled by the melt thickness (the thinnest among the slip-stepping experiments). 430 But here we cannot evaluate how the size reduction and melt viscosity influence the D431 value due to their complicated correlation and more comprehensive studies are 432 required.

433

## 434 4. Discussion

## 435 4.1 Frictional evolution during fast fault slip at low normal stress

436 We focus on the complicated frictional evolution of granitic melts at low normal 437 stress. Chen et al. (2017b) performed frictional experiments on granitic rocks for 438 moderate velocities (0.01-0.11 m/s) to investigate the complicated frictional evolution 439 with slip at the transition between powder lubrication and melt lubrication. They 440 suggested that the evolution of the frictional behavior can be divided into three stages 441 based on different physical mechanisms: the initial weakening stage (Stage 1: powder 442 lubrication), the initial strengthening stage (Stage 2: viscous melt patches), and the 443 final weakening stage (Stage 3: melt lubrication) (Fig. 10a). They concluded that this 444 complicated frictional evolution is the result of thermally-activated deformation 445 processes. In the discussion below, we exploit the conceptual model proposed by 446 Chen et al. (2017b) to our experimental dataset. At slip initiation, during Stage 1, we 447 propose that flash heating and melting of the asperity contacts is the dominant 448 weakening mechanism once a threshold sliding velocity of > 0.1 m/s (case for 449 silicate-built rocks) is achieved (Rice, 2006; Di Toro et al., 2006b; Hirose and 450 Shimamoto, 2005; Niemeijer et al., 2011; Goldsby and Tullis, 2011; Proctor et al., 451 2014; Violay et al., 2014a; Fig. 10b-e). Then, we divide the Stage 2 of Chen et al. 452 (2017b) (dynamic fault strengthening in the presence of powders and melt patches) 453 into States A (first peak), B, and C (second peak) and keep Stage 3 of Chen et al. 454 2017b (weakening due to the presence of a continuous melt layer) as our State D (see 455 also discussion in Hirose and Shimamoto, 2005; Di Toro et al., 2006b; Niemeijer et al., 456 2011; Violay et al., 2014b; Proctor et al., 2014) (Fig. 10b-e). Below we focus on the 457 complex frictional evolution during Stage 2. 458

## 459 4.2 Initial strengthening in Stage 2 (State A)

460 At slip initiation, when flash heating occurs, temperatures significantly rise at the 461 asperity scale and melt the rock contacts (Rice, 2006). In addition, rubbing of the 462 asperity contacts induces tribo-chemical reactions and triggers different mechanisms. 463 Therefore, tribo-chemical reactions may occur at lower activation energies (and thus 464 their kinetics is more efficient at a given temperature) than thermo-chemical reactions 465 (Steinike and Tkácová, 2000). It is reasonable to assume that flash heating of the 466 granitic gneiss may result in the preferential fusion of the low melting point minerals 467 (i.e., muscovite,  $T_{\rm m} = 650^{\circ}\text{C}-900^{\circ}\text{C}$  at 1 atm; Spray, 2010, see discussion below) but 468 also in the formation of silica-rich melt droplets from fusion of high melting point 469 minerals as quartz (main mineral of the gneiss,  $T_{\rm m} = 1720^{\circ}$ C at 1 atm; Spray, 2010). 470 The melt patches in State A were derived from the accumulation of the melt droplets 471 produced by flash heating and melting at the asperity contacts, resulting in the 472 formation of relatively silica-rich melts.

In addition, the PSD data of State A shows the presence of ultra-fine grains, suggesting an additional source of grains into the melt. When frictional melts occurred and covered the entire slip surface, the survivor grains were mainly derived from thermal cracking and frictional melting of the wall rocks and sticking out from the wall rocks into the melt. Based on this, compared to the PSD of State D, we conclude that the additive grains in State A were the result of mixtures of rock clasts (by rock fragmentation) and melt patches during initial sliding.

480 The temperature measured with the infrared camera at State A is  $\sim$ 500°C, and it 481 is likely an underestimated because of (1) averaging with the slipping zone and the 482 wall-rocks and (2) slightly unfocused measuring (outside of the slipping zone). 483 Integrated with the mineralogical observation (Fig. 4e), the temperature at State A 484 should achieve the breakdown point of muscovite (at least > 650°C) and was 485 relatively lower than the other states (with shorter slip and less frictional work input; 486 see the details in section 4.3). Therefore, the estimated temperature and corresponding 487 viscosity (Fig. 8 and Table 1) indicate that the silica-rich melt patches of State A were 488 highly viscous and presumably contributed to the resistance of shearing, resulting in 489 the initial strengthening in Stage 2, similar to the observation reported by Chen et al. 490 (2017b).

491

#### 492 **4.3 Re-strengthening in Stage 2 (states B to C)**

With progressive sliding, the melt patches accumulated into a donut-shaped melt layer and the two rock specimen were partly separated from the melt layer (States A to B; Fig. 4b). Here, we simply neglect the contribution from solid-solid friction in the central part of the slip surface because the torque and thus the shear stress increase 497 with sample radius (e.g., Eq. 2), where the frictional contribution from the inner part 498 of the cylinder is negligible (Hirose and Shimamoto et al., 2005). In addition, because 499 the microstructural observation shows that solidified melts were thicker than the 500 surface roughness (Fig. 4), the solid-solid contacts at the outer melt regime were 501 unlikely to have occurred. We suggest that donut-shape melt layer is the main 502 contribution for shear resistance during the experiment. As a consequence, melt 503 viscosity and shear strain rate are the dominant parameters that control the shear 504 resistance of the simulated fault during the frictional melting regime (States B to D).

505 Melt viscosity is controlled by three parameters: temperature, presence of water, 506 and chemical composition (Lejeune et al., 1999; Fluegel et al., 2004). According to 507 the mechanically-constraint viscosity model (section 3.3), the temperatures achieved 508 in States A to D are about 1370°C, 1300°C, 1456°C, and 1265°C, respectively (Fig. 8). 509 The estimated temperature of State D (melt lubrication regime) is in good agreement 510 with previous rock friction experiments conducted at seismic slip rates (Hirose & 511 Shimamoto, 2005; Di Toro et al., 2006; Niemeijer et al., 2011, 2012; Nielsen et al., 512 2008). The estimated temperature of State D (1265°C) can be set as the upper bound 513 for the temperature achieved in the slip-stepping experiments. In fact, the temperature 514 achieved in States A to C should be lower than the one achieved during State D 515 because, under the same conditions, the shorter slip also imply less work input by the 516 motor. Therefore, according to the evaluation of temperature at State D, the estimated 517 temperature of State C (1456°C) is overestimated.

518 On the basis of the mineralogy of the survivor clasts (e.g., muscovite was melted 519 through all the slip-stepping experiments; Fig. 4e), we can take the melting point of 520 muscovite into consideration to estimate the temperature of the friction melt. The 521 melting point  $(T_m)$  of muscovite during static heating at a low heating rate of 1°C/min is 650°C (Spray, 2010) and T<sub>m</sub> increases to 900 °C at a high heating rate of 200 °C/min 522 523 (Kuo et al., 2011). Because of the difficulty of evaluating on one side the contribution 524 of grinding processes during frictional sliding that may further lower the  $T_{\rm m}$  of 525 muscovite and, on the other side the heating rate which is much higher than the 526 aforementioned heating rates tested in the laboratory (e.g., Kuo et al., 2011), we 527 assume that the lower bound of the temperature achieved during States B and C is 528 ~900°C.

In experiment LHVR1097 we measured the temperature evolution from States A to D (Fig. 3e). The temperature measured by the infrared camera is affected by (1) absorption (i) of emitted radiance by gases produced during frictional sliding (SO<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub>) and (ii) of the powders suspended in the air, (2) variance of emissivity ( $\varepsilon$ ), and (3) the thickness of the slipping zone which is thinner than the area investigated by the infrared camera (i.e., the camera measures the temperature over an area that 535 includes both the hot slipping zone and the cold wall rocks). As a consequence, the 536 temperature measured with the infrared camera is commonly considered as an 537 underestimated value. Abtahi et al. (2002) reported that broadband emissivity 538 systematically rises as the lava cools, from 0.55 at 1050°C to 0.85 at <500°C. In our 539 case, the widespread preferential melting of muscovite demonstrates that temperatures 540 were higher than 650°C in the slipping zones of States A to D. As a consequence, it 541 seems that a low value of emissivity ( $\varepsilon = 0.55$ ) is more appropriate for the temperature 542 measurements with the infrared camera, and thus, the associated estimated 543 temperature is ~850°C, as the lower bound of temperature of States B and C. Taken 544 together, the achieved temperatures for States A to C should be in the range of 850°C 545 to 1265°C (the pink area in Fig. 8).

In the temperature range of  $850^{\circ}$ C to  $1265^{\circ}$ C, the viscosity of the melt in State C is higher than the one achieved in the other states  $(10^{8.0}-10^{3.1} \text{ Pa s}, 10^{7.7}-10^{2.7} \text{ Pa s}, 10^{8.2} -10^{3.3} \text{ Pa s}$  for States A to C, respectively). In addition, the slope of the Non-Arrhenian temperature dependence of the viscosity curves become less steep. This suggests that even if the temperature of the melt increases with slip between States B and C (i.e. temperature at State C is slightly higher than the one at State B; Fig. 3e), the estimated viscosity at State C can be higher than the one at State B.

553 To summarize, because of frictional heating, the bulk temperature in the slipping 554 zone increases with slip (Fig. 3e) and likely reduces the melt viscosity, resulting in 555 melt lubrication (Di Toro et al., 2006). However, by itself, the gradual temperature 556 increase, which would result in less viscous friction melts, is at odds with the 557 re-strengthening behavior (i.e.,  $\mu_{p2}$  occurred at an higher temperature than  $\mu_{p1}$ ; Figs. 2 558 and 3e). On the basis of the temperature-viscosity model (Fig. 8), the evolution of the 559 chemical composition of the frictional melt with slip might be the most plausible 560 explanation for re-strengthening from State B to C. Though water was present at the 561 initiation of slip (the experiments were performed under room-humidity conditions) 562 and released by the breakdown of muscovite, because of the not-confined conditions, 563 gases could escape from the frictional melt. This resulted in the formation of vesicles 564 and in the negligible water content of the pseudotachylyte matrix (Raman 565 spectroscopy data in Fig. 7). As a consequence, water cannot be responsible for the 566 variation of the viscosity of the melt in any of the States. On the other hand, the 567 experiment s1286 under vacuum conditions (Table 1) may further indicate the 568 re-strengthening is irrelevant to the humidity and/or oxidation. Taken together, the 569 increase in  $SiO_2$  in the frictional melt might be the most plausible contributor for the 570 re-strengthening from State B to State C (Table 1).

571 The presence of clasts made of quartz and, to a less extent, feldspar and the 572 absence of muscovite in the solidified frictional melt (Figs. 4e, 5) suggest that 573 selective melting of low-melting-point minerals (i.e., muscovite) took place (Shand, 574 1916; Sibson, 1975; Spray, 1987). However, the chemical composition of the glass 575 matrix at States B and C is higher in SiO<sub>2</sub> and lower in CaO (Table 1) compared to the 576 composition of the glass matrix at State D which was produced at higher temperatures 577 (Fig. 3e). This chemical variation suggests that partial melting of quartz was more 578 efficient at States B and C than State D while partial melting of feldspar was 579 comparatively efficient at State D. In fact, during State D the sliding surfaces were 580 entirely separated by the frictional melt (Fig. 4d) and grain fragmentation due to 581 solid-solid contact of the asperities was less efficient than during States A to C. 582 Therefore, the presence of rock powders (mostly made by quartz and feldspar) in the 583 frictional melts likely drove further melting of the small grains by quasi-equilibrium 584 melting (Lee et al., 2017) or by the Gibbs-Thomson effect due to the high 585 surface-to-area ratio (Hirose and Shimamoto, 2003). These particular melting 586 processes resulted in the increase of Ca (although not obvious in our result) and 587 especially Si content in the frictional melt and, as a consequence, in the increase of 588 the melt viscosity from State B to State C (Table 1). Without the addition of 589 fragmentation products, the chemical composition of the glass matrix at State D was 590 then mainly derived from the melting of feldspar and muscovite.

591 We propose that the enrichment in Si in the frictional melt associated to 592 grain-by-grain fragmentation at the initiation of simulated seismic slip may explain 593 the observed fault re-strengthening observed in previous experiments performed at 594 seismic rates (Di Toro et al., 2006b; Hirose and Shimamoto, 2005; Niemeijer et al., 595 2011) (Fig. 10). Moreover, this process of chemical evolution of melt composition 596 with slip also support the model proposed by Fialko and Khazan (2005) according to 597 which thermal-activated slip strengthening is likely to occur in quartz-rich rocks at 598 shallow depths.

599

## 600 4.4 Final weakening during Stage 3 (State C to D)

601 The final fault weakening is observed in all the experiments, but of course not in 602 the short slip-stepping experiments (Fig. 2b), and is associated to the formation of a 603 continuous frictional melt layer (Fig. 4d). In fact, frictional melts with high 604 temperatures and low silica content (Table 1) are an efficient lubricant, consistently 605 with previous experimental and theoretical studies (Hirose & Shimamoto, 2005; Di 606 Toro et al., 2006a, b; Niemeijer et al., 2011, 2012; Nielsen et al., 2008). In addition, 607 our mechanical data are comparable to those of previous experiments on tonalite 608 (similar mineral assemblages without foliation) where both data show low effective 609 friction coefficient (0.13 and 0.05, respectively) at the steady-state melting (Di Toro et 610 al., 2006a, b). This suggests that the mineralogical heterogeneity of the specimens due

to the metamorphic foliation makes only a very minor influence on the obtainedvalues of shear stress.

613

#### 614 4.5 Rock friction experiments and Re-strengthening behavior

615 The mechanical data shows a complicated evolution of the friction coefficient 616 (flash weakening, first strengthening, slight weakening, second strengthening, and 617 eventually weakening) with slip at low normal stresses (< 10 MPa) (Fig. 2a). This 618 complicated evolution almost disappears and approximates an exponential decay at 619 larger normal stresses (e.g., 30 MPa) (Fig. 2a). The most significant change with increasing normal stress is the reduction of the first peak value  $\mu_{\rm pl}$  and the 620 621 disappearance of the second peak  $\mu_{p2}$  (Fig. 2a). This is the result of the more 622 continuous transition from flash weakening to melt lubrication with increasing normal 623 stress, but also with increasing slip rate and slip acceleration rate (5 m/s and 6.5  $m/s^2$ , 624 respectively, for the experiments performed with SHIVA). In fact, the higher heat 625 production rate, proportional to the product of the normal stress per slip rate, 626 dissipated on the slipping zone results in the fast generation of the frictional melt and 627 rapid reduction of the friction coefficient. Therefore, at larger normal stresses and slip 628 rates, the effect of surface comminution becomes negligible during the onset of slip.

629 In the case of the experiments performed with SHIVA, sample preparation 630 (pre-grounding process) can influence the frictional evolution with slip. Niemeijer et 631 al. (2011) pre-grounded the samples with SHIVA to achieve perfect alignment and 632 obtained a complicated friction evolution afterward. It seems that, by doing this, 633 ultra-fine powders were likely generated and potentially stuck on the asperities of the 634 slip surface, even though the powder was cleaned with an air brush before the 635 high-speed experiment. Nielsen et al. (2012) applied a new method to align the 636 SHIVA samples without pre-grinding the slip surfaces and obtained a simpler 637 evolution (which approximated an exponential decay) of the friction coefficient at slip 638 initiation (Violay et al., 2014a,b; Violay et al., 2019). Therefore, both SHIVA and 639 LHVR results demonstrate the presence of a correlation between the presence of 640 fragments in the slipping zone and the second re-strengthening behavior.

641

## 642 **4.6 Implications for natural Hoping pseudotachylyte**

We apply our experimental results to the natural observations of the Hoping pseudotachylyte. Ferré et al. (2016) proposed to use a novel linear structure decorating the slip surfaces of pseudotachylytes, described as "brushlines", to identify the coseismic slip direction. The brushlines were suggested to form through viscous brushing between protruding asperities of the host rocks and the viscous frictional melt (solid-liquid interaction) during rapid cooling at the end of slip. The chemical 649 compositions of the Hoping pseudotachylyte (average 61% silica content, Kuo, 2016) 650 and of the experimental pseudotachylytes from the same rocks generated in State C 651 (Table 1) are very similar. It implies that the Hoping pseudotachylyte likely formed 652 during small seismic slip events or at the initial fault sliding (corresponding to State B 653 to C) and contained remnant grains from fault comminution. If so, our finding about 654 the chemically induced increased viscosity of the frictional melt might be another 655 explanation for the process of viscous brushing.

656

#### 657 **5.** Conclusions

658 In this study, we present the mechanical results of slip-stepping experiments 659 performed on solid cylinders of granitic gneisses sheared at high slip velocities using 660 rotary shear machines (SHIVA and LHVR) coupled with high-resolution 661 microstructural (scanning electron microscope, Transmission X-ray microscopy, 662 image analysis, etc.) and mineralogical (synchrotron X-ray diffraction analysis, 663 micro-Raman spectroscopy, etc.) investigations of the experimental fault products. In 664 the case of the experiments performed at seismic slip rates (1.3 to 5 m/s) but low normal 665 stress (< 10 MPa), the mechanical data show a complicated frictional evolution with 666 slip which includes: initial weakening, first strengthening followed by a slight 667 decrease and then by a second strengthening and a final exponential decay towards a 668 steady state value (Figs. 2-3). This complex frictional evolution becomes less 669 pronounced in the experiments performed at larger normal stresses and is associated 670 with the evolution of the viscosity of the frictional melts (chemical composition, 671 temperature, etc.) (Figs. 4-9 and Table 1). We thus modified the model of Chen et al. 672 (2017b) for the various friction behaviors (stages 1 to 3) associated with different 673 physical mechanisms (Fig. 10). We propose that the initial fault weakening is due to the 674 grain fragmentation and flash heating and weakening on bare rock surfaces. The first 675 fault strengthening is due to braking from the formation of a highly viscous (also 676 because of the low temperature) frictional melt. The second fault re-strengthening is 677 associated with the increased viscosity mainly because of the SiO<sub>2</sub> enrichment of the 678 frictional melt. The increase in  $SiO_2$  content is probably due by a combination of 679 quasi-equilibrium melting and the Gibbs-Thomson effect on the ultra-fine quartz 680 grains. The final weakening is due to melt lubrication because of the formation of a 681 continuous melt layer covering the entire slip surface together with the reduction of the 682 viscosity because of the increased temperature of the melt.

683 Considering the experimental conditions at which the chemically-induced high 684 viscosity frictional melt forms, we suggest that the surging of frictional melts 685 generated behind the rupture front likely hampers seismic slip during initial 686 earthquake propagation at shallow depths, even though melt lubrication takes place afterward. As a natural example, although the Hoping pseudotachylytes were
generated at depths > 4km, our result suggests that the chemically-induced high
viscosity frictional melt formed in the Hoping area may therefore play a braking role
during fault rupture into shallow depths.

691

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- 937

## 938 Captions

939 Figure 1. (a) Geological setting of the Hoping area with complex tectonic structures. 940 The inset box shows the location of the Hoping area in northeast Taiwan. The study 941 area consists mainly of marble, granitic gneiss, and schist. The Hoping 942 pseudotachylyte (red star) and its surface structure "brushlines" (green box; the 943 surface was coated by Pt for SEM analysis) and the Hoping borehole are located 944 alongside the Hoping River and are hosted in granitic gneiss. The attitude of foliation 945 is mostly parallel or sub-parallel to the direction of fold axis. (b, c) Specimen 946 assembly for LHVR and SHIVA. Note that the specimen size for LHVR is 25 mm in 947 diameter and for SHIVA, the outer diameter is 50 mm and the inner diameter is 30 948 mm. Specimens were bound with iron wires and jacketed with aluminum rings for 949 LHVR and SHIVA, respectively.

950

Figure 2. Mechanical and compiled data from experiments performed with LHVR andSHIVA machines. (a) Apparent friction coefficient as a function of displacement

953 under normal stresses from 3 to 30 MPa and slip velocity at 1.3 m/s (LHVR) and 5 954 m/s (SHIVA) under room humidity (LHVR) and vacuum (SHIVA) conditions. The 955 static friction, initial weakening, first peak and second peak are indicated by the 956 arrows. (b) Shear stress of the first peak (red circle), second peak (blue diamond), and 957 steady-state value (black square) varied with normal stress. Note that the dynamic 958 shear strength for peak states and steady state is well below Byerlee's friction values 959 ( $0.6 < \mu < 0.85$ ).

960

961 Figure 3. Evolution of the apparent friction coefficient, slip velocity, axial 962 displacement and temperature with displacement of five slip-stepping experiments. (A) 963 State A (LHVR0487) stopped at the peak value representing the first 964 slip-strengthening behavior. (B) State B (LHVR0534) was stopped at the second 965 slip-weakening behavior. (C) State C (LHVR0452) stopped at the second 966 slip-strengthening behavior. (D) State D (LHVR0438) was stopped at the steady-state 967 value. (E) The experiment reproducing the condition of LHVR0438 with outcrop 968 sample coupled with the infrared thermal-sensing camera. The temperature increases 969 with slip from 150°C (minimum detected value of the infrared camera) at the slip of 970 ~0.4 m to ~850°C ( $\varepsilon = 0.55$ ) and ~600°C ( $\varepsilon = 0.9$ ) at the slip of ~5.3m.

971

972 Figure 4. Experimental results of four states with corresponding schematic sketch of 973 the solidified frictional melt distribution on the slip surface. The petrographic sections 974 were prepared to be perpendicular to the slip surface and analyzed from half-way 975 from the center to the edge. (a-d) Open Nicol observation of the experimental 976 products. The frictional melt distribution evolves from the melt patches in State A, to 977 a donut-shape melt layer in State B (5x intensity for compare) and C, to a fully 978 covered melt layer in State D, respectively. (e) In situ synchrotron XRD analysis of 979 the host rock and the experimental products from the simulated fault surface. The 980 disappearance of the diffraction peak of mica and reduction of the diffraction intensity 981 of feldspar and quartz are reflected in the XRD spectra and show no variation between 982 the four states. The different colors between the melt spectra are indicated by the 983 calculated bulk temperature where the red line refers to a higher melting temperature 984 than the pink lines.

985

Figure 5. BSE images of the frictional melts for four states. (a) In State A, quartz grains (dark grey) with sizes ranging from 2 to 10  $\mu$ m are abundant within the melt matrix (light grey). For (b), State B, the number of quartz grains with a size lower than 5  $\mu$ m decreased. For (c), State C, smaller quartz grains (< 1  $\mu$ m) appeared while larger quartz grains (> 10  $\mu$ m) disappeared. For (d), State D, quartz grains with sizes larger than 10 µm are dominant within the melt. The dark area with spherical toellipsoidal occurrence are vesicles, which are particularly larger in State D.

993

994 Figure 6. TXM images of the solidified frictional melts for the four states. Internal 995 microstructures all show substantial ultrafine quartz grains with sub-angular and 996 angular-to-spherical, and ellipsoidal surrounded by a melt matrix. (a) The enlarged 997 image highlights the appearance of the fractured grains. (b) The enlarged image 998 highlights the overlapping occurrence of ultrafine spherical quartz. (c) The enlarged 999 image shows no distinct boundary of ultrafine quartz with a sub-angular to the 1000 spherical shape. (d) The enlarged image shows a distinct occurrence of ultrafine 1001 spherical to ellipsoidal quartz.

1002

Figure 7. Representative Raman spectra of the glass from four states, quartz, andwater. The signal for the presence of water is barely observed for each state.

1005

1006 Figure 8. Non-Arrhenian temperature dependence of viscosity for four states. The 1007 viscosity curves were derived using the GRD viscosity model of the Giordano et al. 1008 (2008) based on the chemical composition of the glass matrix. The apparent viscosity 1009 corrected with clast content ( $\eta_c$ ) was derived from the mechanical data. Intersection 1010 points are the estimates of temperature of chemo-mechanical data for the four states. 1011 The temperatures achieved for states A to C were estimated between the achieved 1012 temperature of State D and the melting point of muscovite obtained by slow heating 1013 (650°C) and fast heating rate (900°C) experiments. See the main text for further 1014 discussion. Two temperatures obtained by infrared measurement from emissivity ( $\varepsilon$ ) of 0.55 and 0.9 were plotted. 1015

1016

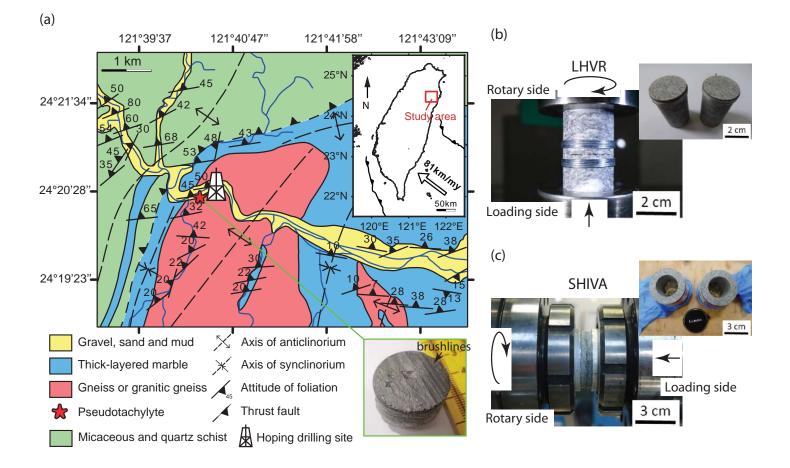
1017 Figure 9. Particle size distribution of surviving grains within the melt matrix for four 1018 states. Cumulative particle density as a function of mean diameter. The particle 1019 density, N(r) representing the cumulative number of particles per  $\mu$ m<sup>2</sup>, following a 1020 function of particle mean diameter (*r*) in micrometers with regression lines (solid lines) 1021 in a form of power law  $N(r) = N'(1 + r/r')^{-D}$ . The obtained particle density from TXM 1022 analyses is relatively higher than the interpolated regression line (dashed line) for the 1023 four states.

1024

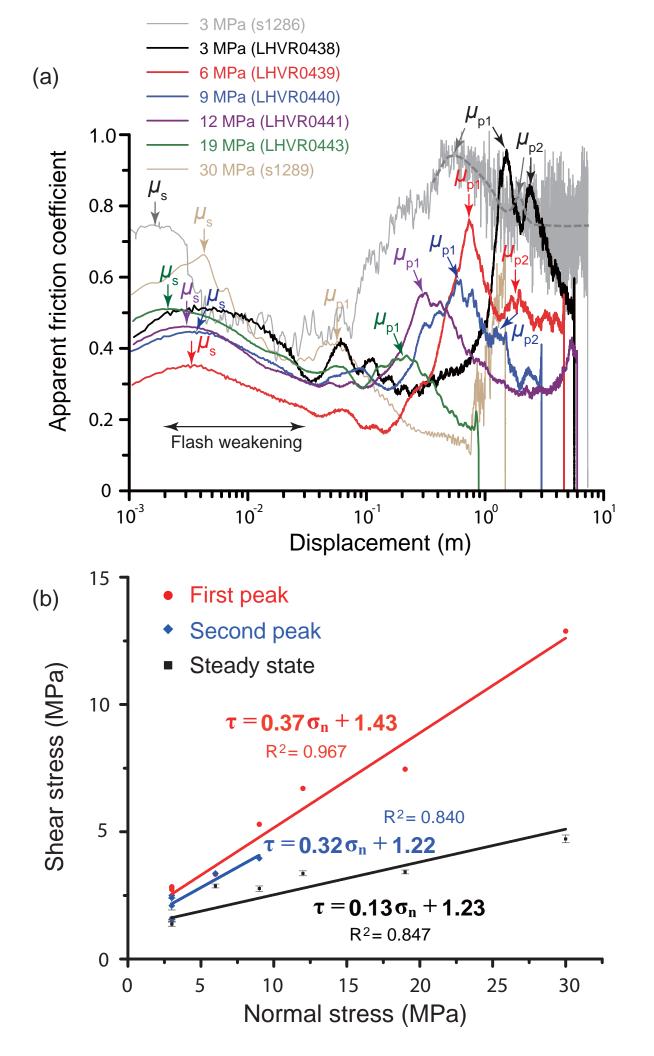
Figure 10. Frictional evolution with displacement for crystalline silicate rocks. (a)
Schematic sketch of the frictional evolution of Sierra white granite sheared at a slip
velocity of 0.048 m/s under a normal stress of 1.22 MPa (after Chen et al., 2017b).
The associated slipping mechanisms corresponding to each stage were shown. (b)

Indian Gabbro sheared at a slip velocity of 0.85 m/s under a normal stress of 1.5 MPa 1029 1030 (after Hirose and Shimamoto, 2005). (c) South Africa gabbro sheared at a slip velocity 1031 of 3 m/s under a normal stress of 20 MPa (after Niemeijer et al., 2012). (d) Tonalite 1032 sheared at a slip velocity of 1.3 m/s under a normal stress of 20 MPa (after Di Toro et 1033 al., 2006b). (e) Schematic sketch of the frictional evolution of granitic gneiss sheared 1034 at a slip velocity of 1.3 m/s under a normal stress of 3 MPa. Similar to Chen et al. 1035 (2017a), the evolution of powder, melt growth, and temperature with displacement is 1036 further added into our conceptual model and Stage 2 is further delimited into two 1037 regimes by the grey line. The four schematic diagrams of the slipping zone summarize 1038 how the PSD, particle volume fraction, melt distribution, microstructures and clast 1039 packing varied with frictional evolution with slip for the reference experiment 1040 LHVR0438.

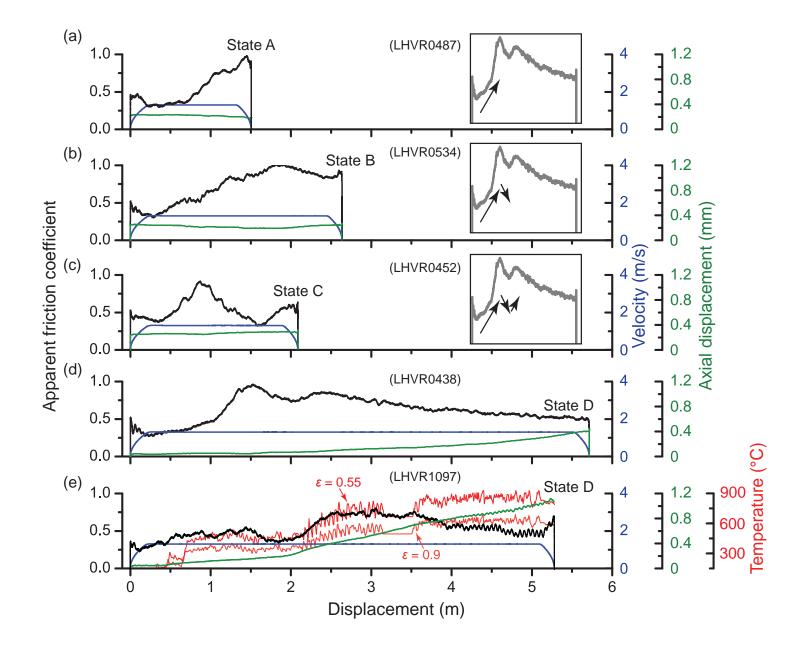
Figure\_01.



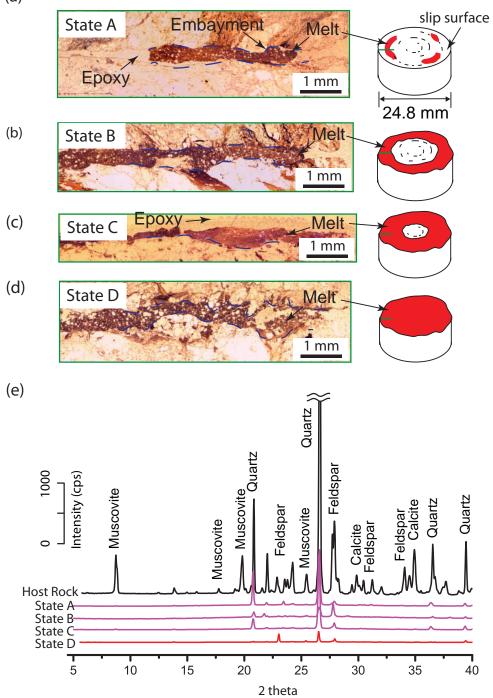
Figure\_02.



Figure\_03.

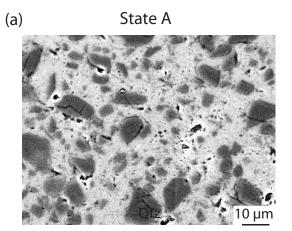


Figure\_04.

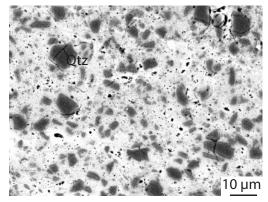


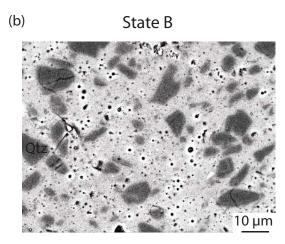
(a)

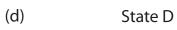
Figure\_05.

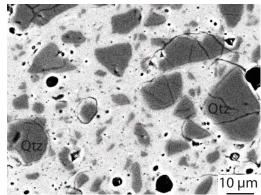


(c) State C

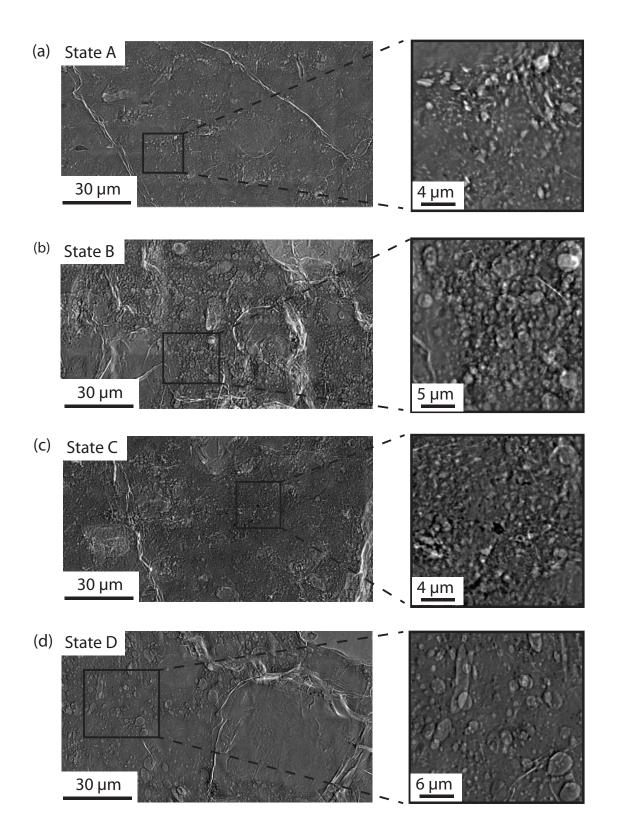




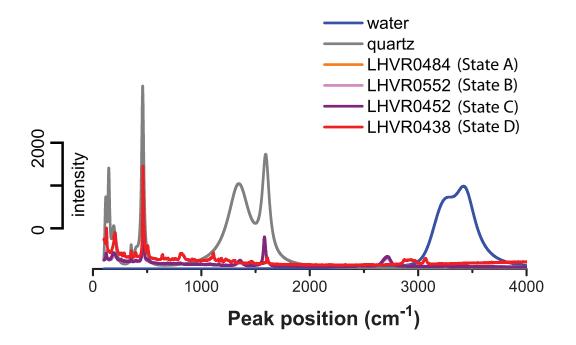




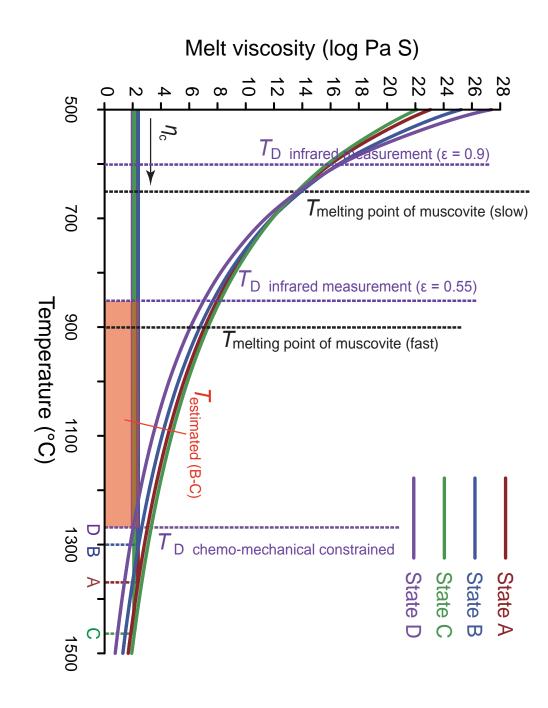
Figure\_06.



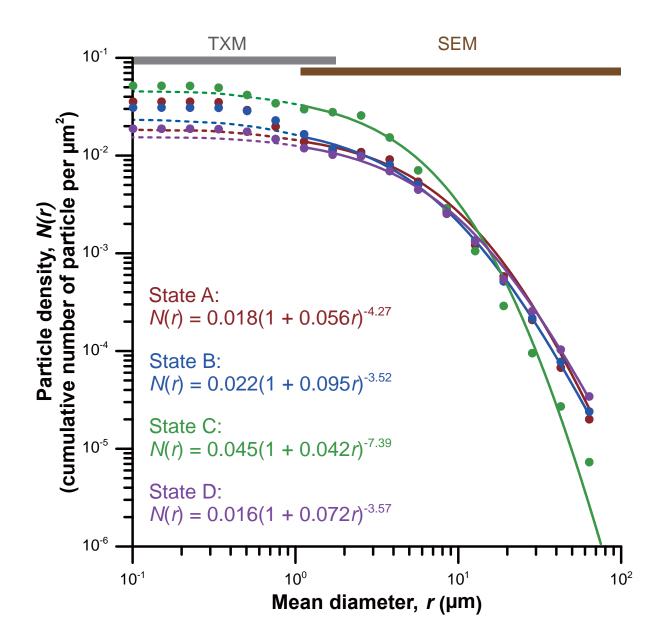
Figure\_07.



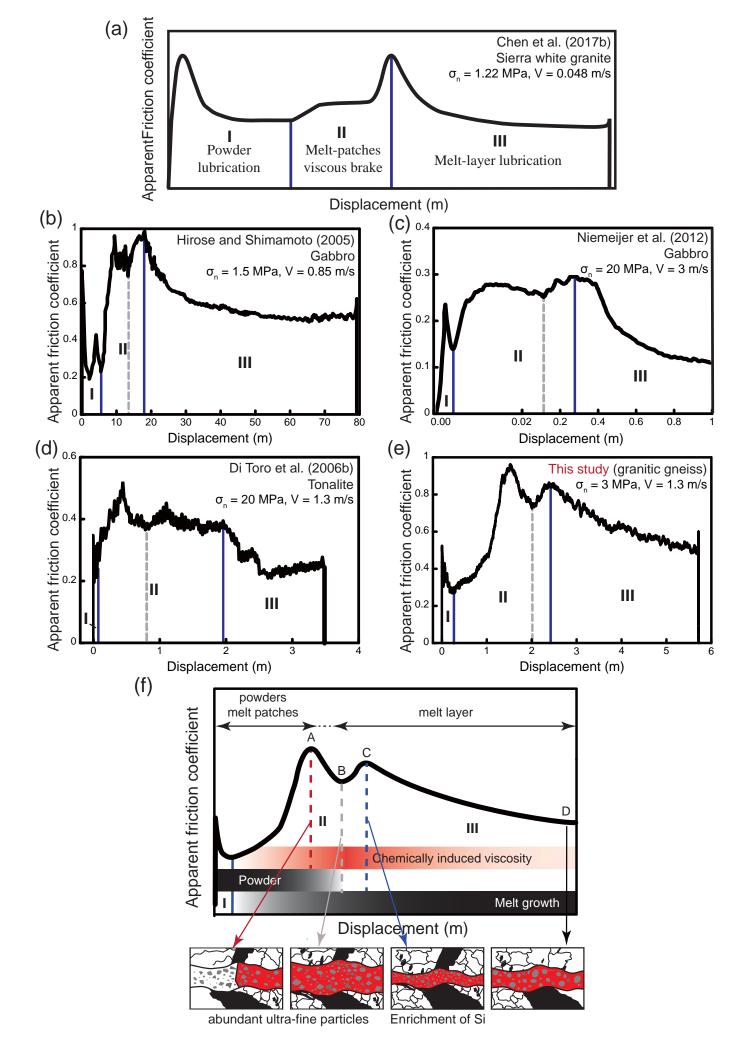
Figure\_08.



Figure\_09.



Figure\_01.



	n	Si	Al	Mg	Ca	Na	K	Ti	Fe	Mn	Р	Total	Estimated melt viscosity range (Pa s)
Granitic gneiss (Kuo, 2016)	4	65.6 ± 2.42	$16.2 \pm 0.67$	$1.8 \pm 0.13$	$2.8\pm0.44$	$2.7\pm0.05$	$3.8 \pm 0.28$	$1.0\pm0.14$	$5.8 \pm 1.21$	$0.1 \pm 0.00$	$0.2 \pm 0.05$	100.00	-
State A (LHVR0484)	39	$58.3\pm5.87$	$18.6\pm2.64$	$2.1\pm0.39$	$4.2\pm0.77$	$2.9\pm0.51$	$6.2\pm0.82$	$0.6\pm0.63$	$7.2\pm1.25$	_	-	100.00	10 <sup>8.0-3.1</sup>
State B (LHVR0552)	40	$53.0\pm3.47$	$20.4\pm1.35$	$2.6\pm0.46$	$4.9 \pm 1.12$	$2.7\pm0.85$	$7.1\pm0.63$	$1.0\pm0.59$	$8.4 \pm 1.48$	_	-	100.00	10 <sup>7.7–2.7</sup>
State C (LHVR0452)	39	$61.3\pm5.91$	$17.5\pm2.63$	$2.2\pm0.42$	4.4 ± 1.22	$3.0\pm0.52$	$6.1\pm0.92$	$0.2\pm0.39$	$5.3\pm1.20$	-	-	100.00	10 <sup>8.2–3.3</sup>
State D (LHVR0438)	40	$50.1\pm3.02$	$17.2 \pm 1.54$	$2.5\pm0.3$	$13.5\pm1.92$	$2.8\pm0.31$	$6.3\pm0.95$	$0.6\pm1.34$	$7.0\pm1.62$	_	_	100.00	10 <sup>7.1–2.7</sup>

Table 1. Normalized average chemical composition of the granitic gneiss, glass matrix at four states, and chemically-based melt viscosity. Noted that the bulk composition of the granitic gneiss is cited from Kuo (2016) which using XRF to analyze four rock samples.

n, number of analyses.

Experiment	σ <sub>n</sub> (MPa)	First Peak $ au_{p1}$ (MPa)	Second peak $ au_{p2}$ (MPa)	Average steady state $ au_{ss}$ (MPa)	Slip velocity V (m/s)	Total slip D (m)	Acceleration/ Deceleration (m/s <sup>2</sup> )	Melt thickness w (mm)	Apparent viscosity $\eta_{app}$ (Pa S)	Clast content $\phi$ (%)	Apparent viscosity corrected with clasts $\eta_c$ (Pa S)	Ambient condition
LHVR0484 (State A)	3	4.7	_	_	1.3	1.6	3.5	0.27	613	20	229	RH
LHVR0487 (State A)	3	2.9	_	_	1.3	1.5	3.5	_	_	_	_	RH
LHVR0534 (State B)	3	3	_	_	1.3	2.6	3.5	_	_	_	_	RH
LHVR0552 (State B)	3	3	_	_	1.3	2.7	3.5	0.33	548	18	232	RH
LHVR0452 (State C)	3	2.5	1.55	_	1.3	2.1	3.5	0.17	203	11	125	RH
LHVR0438 (State D)	3	2.8	2.5	1.5	1.3	5.7	3.5	0.23	265	20	99	RH
LHVR1097 (State D)	3	1.6	2.1	1.4	1.3	5.3	3.5	_	-	—	-	RH
LHVR0439	6	4.6	3.4	2.8	1.3	4.7	3.5	_	_	_	_	RH
LHVR0440	9	5.3	4	2.8	1.3	3	3.5	_	_	_	_	RH
LHVR0441	12	6.7	-	3.3	1.3	6	3.5	_	_	_	_	RH
LHVR0443	19	7.5	_	3.4	1.3	1.9	3.5	_	_	_	_	RH
s1286	3	3	2.4	1.6	5	7.5	6.5	_	_	_	_	Vacuum
s1289	30	12.7	_	4.7	5	1.5	6.5	_	_	_	_	RH

Table 2. List of experiments, experimental conditions, mechanical data, and mechanically-constrained apparent viscosity.

RH, room humidity