**Quasi-equilibrium melting of quartzite upon extreme friction**

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**S1 Text**

**S1. Fault-slip-weakening mechanisms/ Friction experiments involving quartz rocks using the rotary shear apparatus:**

Fault-slip-weakening (i.e., decreases in the dynamic friction coefficient of faults plane) during accelerated slip can account for large coseismic displacements and accompany earthquakes of high magnitude (Mw > 7–8). Several fault-weakening mechanisms including the formation of melt layers have been proposed based on pioneering frictional shear experiments [See 1-3 for a detailed review]. Pioneering studies on the frictional properties of quartz rock have been performed using the rotary shear apparatus4,5. These friction experiments performed at slower slip rates—up to ~0.1 m/s reported the formation of amorphous silica particles4,5. The friction experiment for the quartzite during rapid frictional sliding has been challenging because of a breakdown of the quartz-bearing rocks in less than a few seconds without the formation of melt layers without confining pressure medium around the specimen during frictional melting6. Nevertheless, frictional melting of quartz-dominant sedimentary rock, novaculite (consisting of much finer silica particles) at a slip rate of 1.3 m/s showed the dynamic fault weakening7 (see S12 below).
S2. The melting temperature depression in SiO$_2$-H$_2$O binary and SiO$_2$-K$_2$O-Al$_2$O$_3$ ternary oxides:

Phase relationships in the SiO$_2$-H$_2$O binary are well understood. The liquidus in the system decreases with increasing pressure (from 1726 °C at 1 atm to 1450 °C at 40 MPa). Therefore, the observed extensive melting of quartzite (as suggested from the SiO$_2$-rich nature of the natural pseudotachylytes from muscovite quartzite) in the natural fault plane may be due to the combined effects of presence of water and an increased pressure on the melting temperature depression. While the pressure could reach high pressure locally at the asperity contact at an early stage of frictional slip, the estimated pressure condition during the frictional melting experiment is ~13 MPa where the melting temperature could be ~1600 °C, which is higher than the estimated formation temperature of the melts in the current experiment. The melting relationships in the SiO$_2$-K$_2$O-Al$_2$O$_3$ ternary have also been well understood, where the addition of K$_2$O-Al$_2$O$_3$ components into single component SiO$_2$ system leads to a systematic decrease in melting temperature.

S3. Estimation of thickness of the melt layer:

The melt layer’s thickness evolves with varying amounts of fault displacement. The thickness of melt layer (~0.7 mm) was estimated from the recovered rock cylinders (run 3849), where an amorphous layer developed (Fig.S1.e). In addition, it was estimated from run 2547 where the thin-section image of the recovered specimen shows the presence of a glass layer with an estimated thickness of ~0.67 mm (Fig.S1.f), which is similar to that of run 3849. Because the melt layer’s thickness evolves during slip, it is necessary to determine whether the glass layer’s thickness that we measured for the recovered sample represents the melt layer’s thickness during frictional sliding. The maximum temperature was attained nearly at the end of the frictional slip (Fig.S2). While the axial displacement could affect the thickness, noticeable change in axial displacement has not been observed. The change in the axial displacement (or shortening) between 0.767 s and 0.770 s, when the slip ended, was estimated to be 0.009 mm, indicating that any change in the thickness of a melt layer is likely to be negligible. After the experiment ended, additional shortening of 0.053 mm occurred, perhaps because of the ejection of melts and/or the cooling of the rock specimen that expanded thermally during frictional heating. Based on these observations and the fact that the volume of melts is larger than that of solids, the melt layer during slip is expected to be close to (or larger than) ~0.7 mm.
Temperature of slip surfaces during frictional heating:

Temperature of melt during rotary shear experiment vs. its melting temperature. The temperature estimated from the melts within the simulated fault represents the temperature of liquids (not “solids”), although it is certainly not the maximum temperature (see section below). The peak temperature that we measured with a radiation thermometer is the temperature of the melt during frictional heating ($T_r$). The actual melting temperature ($T_{ma}$) of the particles during frictional slip is thus expected to be lower than $T_r$ (i.e., $T_r \geq T_{ma}$). Because the measured $T_r$ is lower than the equilibrium melting temperature of SiO$_2$ ($T_m$) (i.e., $T_m > T_r \geq T_{ma}$). Therefore, the current result, with an unambiguous melting of quartzite during rapid frictional sliding, shows that the estimated temperature of liquids (~1400-1500 °C) is still much lower than the equilibrium melting temperature (1726 °C) (Fig. S2).

Temperature difference between the melt at the outer rim and the melt on the inner slip surface. The melt temperature at the outer rim is expected to be lower than that at the inner part of the slip surface partly because of heat loss$^{11,12}$. While the difference in temperature between the melt at the outer rim and the melt on the inner slip surface is difficult to measure, the maximum velocity (and, thus, the maximum temperature) is expected to be attained at the outermost part of the sample. However, because of the heat loss as the melts come into contact with the air, the temperature of the inner part could be somewhat higher. For example, previous experimental study, together with numerical simulations that considered cooling at the outer surfaces, showed a temperature drop at the specimen’s surface of ~50 K (ref. 13). Earlier temperature estimation of felsic rocks during rapid frictional melting under rotary shear has shown that, based on numerical simulations, the temperature at the outer rim is approximately 60 K lower than that at the inner part of the sample$^{14}$. These previous experiments were performed with a longer fault displacement during rotary shear, which resulted in a more gradual heating; however more heat loss is expected for the longer displacement experiment. In the current study with the duration of < 1 s under extreme frictional sliding, the heat loss through air is expected to be relatively minor, reducing the temperature drop. In addition, the current temperature measurement may not be of the ‘extruded melt’ but represents the peak temperature of the melts exposed to air during friction. Therefore, although the possibility of a moderate temperature drop may not be fully disregarded, the temperature difference between the inner part of the specimen and its margin is likely to be only several tens of degree K. This indicates that the melt temperature even at the inner part of the specimen is still much lower than the equilibrium melting temperature of bulk silica.

Estimation of the temperature at the slip surface using heat flow equation. Given the difficulty and uncertainty involving other methods of temperature estimation, current temperature measured with a radiation thermometer brings the most robust and reliable
estimations of the temperature of a melt layer. Additionally, simple analytical
calculations based on heat flow equation using thermal properties, density of materials,
and mechanical data (e.g., shear stress) during frictional sliding can also be useful. For
example, based on theory of heat conduction, the temperature of slip surfaces of the
quartz rock can be calculated. Considering the average shear stress (~6.7 MPa, \(\tau\)),
average slip rate (~1.3 m/s, \(V\)) and duration of slip (~0.8 s, \(t\)), temperature rise in the
experiments can be roughly estimated by the following equation\(^{15}\):

\[
\Delta T = \left(\frac{\tau V}{\rho c}\right)\sqrt{\frac{t}{\pi a_{th}}}
\]

where \(\rho c\) is volumetric specific heat (742 J kg\(^{-1}\)K\(^{-1}\) at 20 °C and 1151 J kg\(^{-1}\)K\(^{-1}\) at 750 °C,
respectively) and \(a_{th} = K/\rho c\) is thermal diffusivity (\(K\) is thermal conductivity, \(3.3 \times 10^{-6}\)
m\(^2\)s\(^{-1}\) at 20 °C and \(6.9 \times 10^{-7}\) m\(^2\)s\(^{-1}\) at 750 °C, respectively). As the estimated value is heavily
dependent on the effect of temperature on the thermal properties, we used both low-
and high-temperature (20 °C and 750 °C, respectively) properties for the temperature
calculation. The temperature rise estimated for runs 2547 (with a fault displacement of
~1.0 m) and 3849 (with a fault displacement of ~0.8 m) are 1115–1644 °C and 1028–1516
°C, respectively (see S12 below). While the result may be roughly consistent with the
estimated temperature in the current study, the method based on Eq. S1 is subject to a
larger degree of uncertainty in temperature estimation. Therefore, despite limitations,
the estimated temperature with a radiation thermometry provides rigorous constraints
on the melting temperature of the quartzite.

**Melting hysteresis.** The equilibrium melting temperature of particles may depend on
kinetic paths. For instance, it has been shown that super-heating of nanoparticles is
expected during rapid heating\(^{16}\). Therefore, the actual melting of particles could occur at
a temperature higher than bulk \(T_m\), which is in contrast to the observed trend in the
current study. Therefore, the observation of melting of quartzite at a lower temperature
upon heating indicates that the melting temperature of rock during rapid frictional
sliding is indeed lower than the equilibrium melting temperature of SiO\(_2\).

**S5. Characterization of starting material and friction-produced materials:**

Figure S3 shows the optical microscope images of the Brazilian quartzite that
consists mainly of quartz and a minor fraction of muscovite. The grain size of the quartz
varies from ~10 μm to 300 μm. Muscovite crystals (~3-15 μm) are observed at the quartz
grain boundaries and inside the quartz grain. The latter may be captured during
recrystallization. An accessory mineral containing Fe and Ti (red arrow) was also
observed in the muscovite. The Raman spectra for Brazilian quartzite showed the
features due to hydroxyl group in muscovite (not shown here). While minor presence of
these groups may not be fully disregarded, features attributable to hydroxyl groups are
not clearly present in their expected frequency ranges (from 3000 cm\(^{-1}\) to 3700 cm\(^{-1}\)) in the spectrum for the quenched glasses in frictionally melted quartzite.

Figure S4 shows the BSE image of the friction-induced materials. The whitish streak in Figure 1E (in the main text) is ultra-fine Mg-bearing quenched crystals that may have formed from the melting of muscovite in the Brazilian quartzite. They also contain less than 1% FeO and TiO\(_2\). While the exact nature of the quenched crystals (in Fig. S4) remains to be explored, based on its chemical composition, they may consist of multiple mineral phases, such as quartz and K-feldspar with other oxide inclusions\(^{17}\). The figure also shows that melting occurs at the interfaces between minerals, indicating heterogeneous melting. In regard to this heterogeneity, regardless of the attainment of equilibrium conditions upon frictional melting, the image shows the heterogeneous distribution of the glasses and quenched crystals. Therefore, a full homogenization of the melts has not occurred during frictional melting, consistent with an earlier suggestion of un-mixing between melts with two distinct compositions\(^{17}\).

**S6. \(^{29}\)Si MAS NMR peak assignment:**

The NMR peak assignments of the silica polymorphs are somewhat complicated because of low signal intensity (caused by long relaxation time), peak overlap with the quenched glass, and a potential increase in the degree of topological disorder in the crystal upon friction. The observed Si peaks in the current \(^{29}\)Si MAS NMR spectra could be assigned on the basis of the previous \(^{29}\)Si NMR studies on diverse silica polymorphs and silicate glasses, together with inputs from the current characterization of the friction-induced products using the Raman spectroscopy/XRD/TEM. Based on the known \(^{29}\)Si NMR chemical shift ranges for silica polymorphs alone, the -109 ppm peak may be attributed to either cristobalite and/or mixture of cristobalite and tridymite\(^{18}\); previous NMR studies have shown that the multiple peaks at ~-109--114 ppm in the silica may correspond to mixed (and disordered) cristobalite and tridymite\(^{18-21}\). However, as the presence of major fractions of cristobalite and tridymite was not observed in the current diffraction (XRD/TEM) and Raman analyses, the larger fraction of the -109 ppm peak should not be due to those polymorphs. Rather, the presence of \(\beta\)-quartz is clearly revealed in the current experimental studies. Because of difficulty in synthesis, only an in-situ \(^{29}\)Si static NMR study of \(\beta\)-quartz single crystal is available\(^{19}\), and its \(^{29}\)Si MAS NMR spectrum for the powdered crystal is not currently available. While further experimental and theoretical confirmation is certainly necessary, based on the clear presence of \(\beta\)-quartz, the peak can be assigned to \(\beta\)-quartz.

The peak at -80 – -90 ppm can also be assigned to Q\(_4\)(2Al) (four coordinated \(^{14}\)Si species with 2 aluminum cations as the next nearest neighbors) and/or Q\(_3\)(1OH) (\(^{14}\)Si species with 1 hydroxyl group as the next nearest neighbor)\(^{22}\). The preliminary \(^{1}\)H MAS NMR spectra and the \(^{1}\)H-\(^{29}\)Si cross polarization MAS NMR spectra for the recovered
sample does not reveal the clear evidence of hydroxyl group density. Thus the feature corresponds to Q1 (2Al).

S7. Melting of quartz: insights from the chemical composition of glass matrix:

The chemical composition of glass matrix in experimentally produced pseudotachylyte provides insights into the nature of melting during frictional heating. Based on the disequilibrium melting model, the SiO2 content in the glass matrix can be directly predicted from the temperature condition during frictional heating and the melting temperatures of the minerals that make up wall rock. When frictional heating of quartzite occurs below the equilibrium melting temperature of silica (1726 °C), the disequilibrium model predicts the preferential melting of minerals with the lower Tm (i.e., muscovite), so at the estimated temperature condition (~1350–1500 °C) only muscovite (~1% of quartzite) is expected to melt. The SiO2-rich nature of the “glass matrix” in the natural pseudotachylyte indicates that fractions of quartz undergo melting.

S8. Batch melting of amorphous silica nanoparticles:

While systematic and quantitative studies have not yet been performed (as such study is beyond the scope of the current manuscript), in our preliminary study of amorphous silica nanoparticles with diameter of 10-20 nm (through batch melting in the tube furnace), we observe the melt formation at ~1600 °C, indicating that the silica nanoparticles melt at a much lower temperature than its equilibrium melting temperature. The Raman spectra for silica glass and cristobalite presented in Figure 2C in the main text were obtained from the melting of the amorphous silica nanoparticles at ~1600 °C. We note that the detailed structure of amorphous silica nanoparticles (not the crystal but all-amorphous, without periodic lattice) with varying diameter, have been studied extensively using TEM and NMR (e.g., 23 and references therein), revealing the effect of particle size on the nature of silica nanoparticles.

S9. Effect of hydration on melting properties of quartzite:

It has been shown that the hydration of nanoparticles is affected by the humidity, leading to a formation of hydrogen bonds between water and the silica surfaces. Therefore, the humidity can affect the melting behavior, as the melting temperature of silica decreases with increasing water content at an elevated pressure conditions. Because of extensive dehydration of the fluid component prior to and/or during melting, the effect on the melting temperature is expected to be less significant at low pressure conditions studied here. In the current study, the presence of OH species in the
wall rock is evident from the spectroscopic results. However, these features are absent in the friction-induced glasses, indicating the extensive devolatilization (removal of hydrous components) during frictional sliding.

Whether frictional melting during co-seismic slip occurs in the absence or presence of fluids has been discussed previously (e.g., 24 and references therein). Whereas the water in muscovite and nanoparticles can be removed during frictional heating, thermal pressurization is expected in the natural fault plane at an elevated pressure conditions, which could further decrease the friction coefficient and limit the temperature rise during melting. In contrast, it could also decrease the melting temperature, leading to melting of the rock. Indeed, it has been shown that natural pseudotachylyte can be formed from the muscovite-bearing quartzite, suggesting that frictional melting can occur at the presence of fluids in the fault plane17.

S10. Equilibrium, quasi-equilibrium, and disequilibrium melting:

We summarize the characteristics of the frictional melting models here. The conventional disequilibrium model postulates the preferential melting of minerals with low melting points, so the model is suitable for explaining the melting behavior of a rock with more than two minerals with distinct melting temperatures. The disequilibrium melting model assumes the melting of individual phases, rather than melting at the interface between minerals. The model is phenomenologically correct as it is often consistent with observations in natural pseudotachylyte, but is certainly oversimplified. The model may not fully account for the melting of a rock that consists of a single mineral. In contrast, the melting temperature of a hypothetical rock consisting of a single mineral can be described based on the quasi-equilibrium model presented here: a decrease in the particle size results in the melting temperature depression. We note that the melting temperature depression due to particle size reduction is based on equilibrium thermodynamics, but it has not yet been adapted to account for the melting of rocks. The presence of metastable phases and their melting during friction are also taken into consideration, so the model can be more relevant to the melting of natural rocks with metastable assemblages. As an example of the SiO2-H2O binary, the conventional equilibrium melting has been well established based on phase equilibria. The quasi-equilibrium model can be used to account for the melting temperature depression in the binary, as the melting temperature of β-quartz is lower than that of cristobalite. In the SiO2-muscovite pseudo-binary, the rigorous melting temperature of muscovite is difficult to constrain because of its thermal decomposition. Nevertheless, thermal decomposition of muscovite and/or the formation of aluminosilicate melts derived from muscovite are expected to be accompanied by the melting of β-quartz. Equilibrium melting can account for the melting temperature depression, as the composition of the melts shows non-negligible fractions of K2O and Al2O3. Therefore, we
believe that the equilibrium melting should be taken into consideration during the frictional melting. All of the conventional disequilibrium and equilibrium models are combined in the quasi-equilibrium melting model. Taking into consideration the effect on melting temperature depression of particle size reduction and the formation of metastable phases during rapid frictional sliding, the quasi-equilibrium melting in the current study extends the scope of the previous melting models, so it accounts for the diverse melting process that involves both simple and complex multi-component systems during friction.

S11. Metastable melting vs. Disequilibrium melting:

At 1 atm, equilibrium phase relations for crystalline SiO$_2$ are well-established. β-quartz becomes thermodynamically stable over α-quartz above 573 °C, before transforming to tridymite and cristobalite at 870 and 1470 °C, respectively. Cristobalite then melts at 1726 °C. A previous experimental study showed that formation of β-quartz is preferred over cristobalite, even at a slower increase in temperature than the current heating rate (> ~1500 K/s) [ref.25 and references therein]. Therefore, unless a sufficiently slow ramp rate is used, preferential formation of β-quartz over cristobalite is expected during rapid frictional heating. These previous experimental study and thermodynamic calculations confirmed that the metastable melting of β-quartz is expected to occur at approximately 1400 °C$^{25-27}$. We note that cristobalite can be quenched to room temperature (instead of forming β-quartz upon quenching). Therefore, the observed absence of cristobalite in the quenched pseudotachylyte indicates that formation of cristobalite has not been occurred during frictional heating. We note that the metastable melting of β-quartz is distinct from the previous disequilibrium melting, where minerals with lower melting temperatures preferentially melt upon frictional heating. In this regime, melting of the potential metastable phases was not taken into consideration. In contrast, the current study postulates that the melting of metastable phases contributes to the observed melting-temperature depression.

S12. Frictional behaviors of quartzite during frictional melting:

Figure S7 shows the frictional behaviors of quartzite during rapid sliding with varying slip rate. It is evident that slip weakening during frictional melting of quartzite is observed after longer displacement (Fig S7.a). The observed amount of weakening is modest compared with the weakening reported for other crustal rocks. However, because the slip weakening is more rapid and dramatic at higher normal stresses (e.g.,$^{11}$), we expect that, if seismic slip occurs at depth in nature, more efficient fault-slip weakening will occur. Figure 7S.b shows the evolution of velocity and temperature during rapid frictional sliding. The difference in the friction behavior at the initial stage
of slip is due to the difference in the initial surface condition. For instance, run 294-1 was conducted after a long experiment at ~mm/s. In that case, initial friction was low because of the lubricating effect of silica nanoparticles (or hydrated silica) that are formed during the initial smoothing in the slip surfaces. The presence of the silica nanoparticles may suppress an abrupt increase in friction at the initial stage of the run. After the initial stage of slip, the friction behaviors of the three runs are similar. The frictional behavior of quartzite is similar to that of novaculite during rapid frictional sliding\(^7\). While the origin remains to be explored, the large temperature decrease during the displacement from ~0.1 m to 0.2 m may result from the large drop in friction. Such a positive correlation between frictional drop and temperature decrease has been observed previously for other rotary shear experiments; in particular, the friction drop (or weakening) may be partly accompanied by phase transition and/or dehydration in constituent minerals\(^28\). The temperature rise during rotary shear is largely inhibited because of an endothermic dehydration reaction\(^29\). Therefore, the temperature decrease may be partly explained by the phase transitions involving quartz (\(\alpha\)-\(\beta\) transition) because the transition is also known to be endothermic.

In our preliminary study of the effect of fault displacement on friction behaviors at a normal stress of 13 MPa and a slip rate of 1.3 m/s, we found no evidence of macroscopic melting of quartzite with a fault displacement of ~0.62 m. Instead, a layer of silica powder was observed in the slip zone. The particle size varies from tens of nanometers to micrometers. In contrast, a melt layer was formed in the run with the fault displacement of ~0.84 m. While more quantitative estimation remains to be done, the result shows that macroscopic frictional melting may occur at a displacement between 0.62 m and 0.84 m.
Figure S1 | Friction-produced materials during rotary shear experiments of quartzite.

a. Snapshot showing the escape of hot material from the slip zone (run 3849). 
b. Solidified melt fallen to the bottom of the sample room (run 294-1). 
c. Recovered rock cylinder showing solidified melt (pale blue) and powder layer at the central portion of the slip zone (white) (run 2547). 
d. Close-up of glassy solidified melt layer observed in a fragment of the rock specimen (run 2547). 
e. Recovered rock cylinders with solidified melt layers (run 3849). 
f. Optical micrographs of a melt layer in the recovered specimen (run 2547) with plane-polarized light.
Figure S2 | Estimation of temperature during frictional heating of quartzite. a. Snapshot of the frictional melting experiment on quartzite (run 3849). b. Snapshot of the temperature distribution recorded by an infrared thermal imaging camera. An emissivity of 1.00 is assumed. c. Temperature profiles across the melt layer (0.7 mm in thickness). Emissivities of 0.87, 0.90, and 1.00 are used for the red, blue, and black profiles, respectively.\textsuperscript{30}
Figure S3 | Optical micrographs of Brazilian quartzite with plane-polarized light (A, x40; C, x200) and cross-polarized light (B, x40; D, x200). qtz and ms refer to quartz and muscovite, respectively. The red arrows show the Fe- and Ti-rich parts in the muscovite crystals.
Figure S4 | BSE image of the recovered products from frictional heating. a. Pseudotachylyte with melt network within quartz clasts (identical to figure 1-E). b. Close-up view of the whitish streak (open square with label “b”) in fig. a. c. Pseudotachylyte with quartz wall rock. d. Close-up view of part of the white area (open square with label “d”) e. EDS pattern of the area in figure S4.b (with label “e”). f. EDS pattern of the area in figure S4.d (with label “f”). Enlarged EDS patterns are added as insets.
Figure S5 | XRD patterns for quartzite and frictionally melted quartzite (FMQ) with varying scattering angles between 15° and 40°. Red, purple, green, and blue vertical lines correspond to the peak positions and intensities of α-quartz (JCPDS file nos. 01-078-1258, 01-085-0795, 01-070-3755, 01-083-0539), β-quartz (JCPDS file no. 00-047-1144), tridymite (JCPDS file no. 00-001-0387), and α-cristobalite (JCPDS file no. 01-082-1409), respectively.
Figure S6 | TEM analysis of recovered amorphous silica matrix. a. High-resolution TEM images of glass matrix quenched from friction-induced melts. b. Electron diffraction patterns. c. EDXS of the frictionally generated silica glass. Cu in the EDXS is due to the Cu-grid used in the measurement.
Figure S7 | Frictional behaviors of quartzite during frictional melting a. Frictional behaviors of quartzite at a slip rate of 1.3 m/s. b. A close-up view of the part of a that shows the change in friction, slip rate, and temperature over the first 1 m displacement of the fault. ε denotes thermal emissivity. Two ε values of quartz (i.e., 0.87 and 1) were used for temperature estimation. The temperature in the fault slip zone was measured every 1/30 s. The temperature axis (red) is shown on the right side of the figure. The pale red square refers to the estimated temperature range calculated from Eq. S1. c. Comparison of frictional behavior between quartzite (this study) and novaculite7 under similar sliding conditions.
Table S1. Average chemical compositions (wt%) of quartz and muscovite in the wall rock and glass (EPMA analysis).

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<th>Quartz (n = 34)</th>
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<th>Glass (n = 42)</th>
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<td>Average</td>
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Table S2 | Raman modes for quartz in the Brazilian quartzite and other reference silica.

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<td>$\alpha$-quartz</td>
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<td>696</td>
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