Effect of normal stress on the frictional behavior of brucite: 1 Application to slow earthquakes at the subduction plate 2 interface in the mantle wedge 3

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16

17 Abstract

18	We report the results of friction experiments on brucite under both dry and wet conditions under
19	various normal stresses (10-60 MPa). The final friction coefficients of brucite were determined to be 0.40
20	and 0.26 for the dry and wet cases, respectively, independent of the normal stress. Under dry conditions,
21	velocity-weakening behavior was observed in all experiments at various normal stresses. Under wet
22	conditions, velocity weakening was observed at low normal stress (10 and 20 MPa), whereas velocity
23	strengthening was determined at a higher applied normal stress. Microstructural observations on recovered
24	experimental samples indicate localized deformation within a narrow shear band, implying that a small
25	volume of brucite can control the bulk frictional strength in an ultramafic setting. Among serpentinite-
26	related minerals, weak and unstable frictional behavior of brucite under the hydrated mantle wedge
27	conditions may play a role in slow earthquakes at the subduction plate interface in the mantle wedge.

30 1. Introduction

31 Serpentinite is generated by the hydration of ultramafic rocks and has various mineral 32 compositions depending on temperature-pressure conditions of the MgO-SiO₂-H₂O system (Evans et al., 33 2013). As serpentinite has been observed in various important tectonic settings and is considered to 34 contribute to the weakness of serpentinite-dominant areas, the frictional properties of serpentinite have been 35 investigated for several decades (see Guillot et al., 2015; Hirth and Guillot, 2013 for a review). A large 36 volume of serpentinite is located in the mantle wedge in which olivine-rich rock of the upper mantle is 37 hydrated by slab-derived water and composes the subduction plate interface as suggested by geological and 38 seismological studies (Bostock et al., 2002; Christensen, 2004; Guillot and Hattori, 2013; Hyndman and 39 Peacock, 2003; Kawahara et al., 2016; Kawakatsu and Watada, 2007; Mizukami et al., 2014; Peacock and 40 Hyndman, 1999; Reynard, 2013). Because of the mechanical weakness of serpentinite, the relationship 41 between the presence of serpentinite and the aseismic behavior below the downdip limit of seismogenic 42 zones has been argued (Hyndman and Peacock, 2003; Oleskevich et al., 1999). However, many recent 43 observations indicated that slow earthquakes, such as Episodic Tremor and Slip (ETS), Slow Slip Events 44 (SSE), or Low-Frequency Earthquakes (LFE), occur at a depth of the mantle wedge in various subduction 45 zones (Audet and Kim, 2016; Obara, 2002; Obara and Kato, 2016; Rogers and Dragert, 2003; Shelly et al., 46 2006). As those slow earthquakes can trigger or be triggered by huge megathrust earthquakes (Obara and 47 Kato, 2016), the nucleation processes of slow earthquakes are important for understanding seismic activities 48 at subduction zones.

49 Recent seismological and geological studies expect that several hundreds of meters to kilometers 50 wide layer is serpentinized and foliated along the subduction plate interface, and the deformation of this 51 serpentinite layer is likely to relate to slow earthquakes at a depth of the mantle wedge (Bostock et al., 52 2002; Calvert et al., 2020; DeShon and Schwartz, 2004; Dorbath et al., 2008; Kawakatsu and Watada, 2007; 53 Nakajima et al., 2009; Ramachandran and Hyndman, 2012; Tarling et al., 2019). Within this foliated

54 serpentinite layer, both meta-ultramafic and meta-sedimentary blocks are present and metasomatic 55 reactions occur at the boundary between these blocks and the serpentinite matrix (Guillot et al., 2015; 56 Tarling et al., 2019). Such a block-in-matrix structure exhibits complex rheological behavior such that shear 57 stress is controlled by both ductile and brittle deformations of block and matrix depending on the strain rate (Fagereng and den Hartog, 2017; den Hartog and Spiers, 2014; Niemeijer and Spiers, 2007; Tarling et al., 58 59 2019). Thus, understanding the deformation properties of both block and matrix is essential to constrain 60 how the subduction plate interface behaves and generates slow earthquakes. This was also underlined by a 61 geological study on the Livingstone Fault, New Zealand (Tarling et al., 2019), which found that the 62 cataclastic slip surface was coated by scaly serpentinite, suggesting that the deformation process of 63 serpentinite is important for brittle deformation as well as widespread ductile deformation at the subduction 64 plate interface.

65 In addition, near lithostatic pore pressure conditions, which lead to low effective normal stress conditions, have been inferred based on seismic velocity structures at the plate interfaces of several 66 67 subduction zones where slow earthquakes coincidently occur in the regions such as Cascadia, SW Japan, 68 Central Mexico, and Hikurangi (Audet et al., 2009; Audet and Kim, 2016; Eberhart-Phillips and Reyners, 69 2012; Matsubara et al., 2009; Shelly et al., 2006; Song and Kim, 2012). This low effective normal stress 70 condition may be correlated to slow earthquakes because frictional deformation becomes dominant, rather 71 than viscous deformation, in terms of shear strength (French and Condit, 2019; Gao and Wang, 2017). 72 Furthermore, the low effective normal stress condition seems favorable for the nucleation of slow 73 earthquakes (Liu and Rice, 2007, 2009; Rubin, 2008; Segall et al., 2010) and is also consistent with smaller 74 stress drops than regular earthquakes (Ide et al., 2007; Rubinstein et al., 2007, 2008; Schmidt and Gao, 75 2010). Thus, frictional properties of serpentinite under the low effective normal stress condition likely play 76 an important role in slow earthquakes at the subduction plate interface near the mantle wedge.

77	Serpentinite in the mantle wedge is mainly composed of an antigorite-olivine assemblage in
78	warm subduction zones like Cascadia, whereas a brucite-antigorite assemblage dominates in the case of
79	cold subduction zones such as that in NE Japan (Peacock and Hyndman, 1999). Because fluids from
80	subducting slabs have a high SiO2 content, talc is stable in the vicinity of the slab-mantle boundaries
81	(Hirauchi et al., 2013; Peacock and Hyndman, 1999). Serpentinite is made up of serpentinite-related
82	minerals, such as antigorite, brucite, and talc, and as those minerals show different frictional behavior, the
83	frictional properties of each mineral should be understood to interpret the mechanical behavior of bulk
84	serpentinite. Many previous experimental studies investigated the frictional properties of antigorite and talc
85	(Hirauchi et al., 2013; Moore et al., 1997; Moore and Lockner, 2007, 2008; Okazaki and Katayama, 2015;
86	Reinen et al., 1994; Sánchez-Roa et al., 2017; Takahashi et al., 2007; Tesei et al., 2018). However, brucite
87	has rarely been considered in previous studies, as it is challenging to detect brucite under natural conditions
88	because of its fine-grained nature (Hostetler et al., 1966). Brucite is not thermodynamically stable when the
89	slab-derived water contains high SiO2 content, and the mantle wedge may undergo the silica metamorphism
90	(Manning, 1997; Peacock and Hyndman, 1999). However, geological works on the exhumed mantle wedge
91	regions suggested that the silica metamorphism has not occurred widely within the shallow mantle wedge
92	because talc zones or metasomatic reactions are often limited in the narrow part near the meta-sedimentary
93	rocks (Angiboust and Agard, 2010; D'Antonio and Kristensen, 2004; French and Condit, 2019; Guillot et
94	al., 2009; Kawahara et al., 2016; Mizukami et al., 2014; Nagaya et al., 2020; Reynard, 2013; Tarling et al.,
95	2019). These observations indicate that the serpentinite layer at the subduction plate interface may contain
96	brucite because of low silica metamorphism as brucite itself has sometimes been found (Kawahara et al.,
97	2016; Mizukami et al., 2014). Hydrothermal experiments also support the finding that the SiO_2 is effectively
98	consumed and brucite can stably exist with antigorite (Oyanagi et al., 2015, 2020). Although deformation
99	may localize at the metasomatic region (Hirauchi et al., 2013; Tarling et al., 2019), the foliated structure of
100	serpentinite matrix implies that the serpentinite layer still accompanies some portion of deformation at the

subduction plate interface. Furthermore, as brucite is a sheet-structure mineral, which often shows a low
friction coefficient due to weak interlayer bonding, its frictional behaviors may play a role in earthquakes
at the serpentinite layer (Moore et al., 2001; Moore and Lockner, 2004).

104 Only a few previous experimental studies under high normal stress conditions of 100 or 150 MPa 105 have been conducted on the frictional properties of brucite. It was shown that brucite has friction 106 coefficients of 0.40–0.46 (dry) or 0.28 (wet), which are lower than those of antigorite (Moore and Lockner, 107 2004, 2007; Morrow et al., 2000). Regarding the velocity dependence, significant stick-slip behavior has 108 been observed for dry brucite at both room and high temperature, implying velocity-weakening behavior. 109 Conversely, wet brucite shows velocity-strengthening behavior at room temperature, which gradually 110 changes to velocity weakening with increasing temperature (Moore et al., 2001; Moore and Lockner, 2007). 111 The friction coefficient of a serpentinite gouge can be lowered by approximately $\sim 10-15$ % due to the 112 presence of brucite (Moore et al., 2001). The weakness and velocity-weakening behavior of brucite under 113 certain conditions might affect nucleation processes of slow earthquakes at the subduction plate interface 114 in the mantle wedges because velocity-weakening behavior is likely to relate to slow earthquakes as 115 proposed in previous studies. The dilatancy hardening in the velocity-weakening system (Rubin, 2008; 116 Segall et al., 2010), the slip-weakening (Ikari et al., 2013), the transition from the velocity-weakening to 117 velocity-strengthening system at a cut-off velocity (den Hartog et al., 2012; Matsuzawa et al., 2010; 118 Shibazaki and Iio, 2003), or the slow stick-slip (Leeman et al., 2016, 2018; Okazaki and Katayama, 2015) 119 are proposed as mechanisms that generate slow earthquakes. Most of them require the velocity-weakening 120 system to nucleate earthquakes, especially for seismologically detected events like LFEs; therefore, the 121 velocity-weakening behavior of brucite can be suggestive of slow earthquakes at the subduction plate 122 interface.

However, the frictional behavior of brucite at low effective normal stress has not been studied in spite of its potential relationship to slow earthquakes. In this study, we experimentally investigated the frictional behavior of brucite at various effective normal stresses ranging from 10 to 60 MPa to understand
the effect of brucite on the seismic activities at the subduction plate interface in hydrated mantle wedges.

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128 **2. Methods**

- 129 **2.1. Friction experiment**
- 130 2.1.1. Sample preparation

131 Brucite nanoparticles with a grain size of 70 nm chemically synthesized by FUJIFILM Wako 132 Pure Chemical Corporation were used for the friction experiments to simulate its fine-grained nature (Fig. 1). The synthetic samples had a purity of 99.9 % (data from FUJIFILM Wako Pure Chemical Corporation). 133 134 A biaxial testing machine at Hiroshima University, Japan, was used for all friction experiments 135 in this study (Noda and Shimamoto, 2009). There are two gouge layers between three gabbro blocks (Fig. 136 1). The surfaces in contact with gouges were roughened before the experiments using Carborundum (grit 137 80) to prevent slip between the blocks and sample. All brucite samples were dried in the vacuum oven 138 overnight under 120 °C before the experiments. This temperature was selected to remove adsorbed water 139 and prevent the dehydroxylation of brucite into periclase (MgO). For the dry experiments, the brucite 140 powder was quickly sandwiched between the blocks to form the gouge after removing it from the vacuum 141 oven, and the blocks with samples were then put in the testing machine. For the wet experiments, dried 142 brucite was mixed with distilled water before placing it in the gouges and then sandwiched between blocks.

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144 **2.1.2. Experimental procedures**

Normal stress was horizontally applied on the side blocks, and shear stress was applied vertically
by pushing the center block downward (Fig. 1). Before applying shear stress, the desired normal stress was

147 applied to the blocks for 1 h to prevent an effect of the compaction of the gouge during shear deformation 148 (nominally precompaction). For the wet experiments, the blocks and gouges were placed in the tank filled 149 with distilled water for 1 h under a normal stress of 250 kPa before the precompaction with the desired 150 normal stress such that water-wet conditions were achieved. Note that as we did not have any mechanism 151 to prevent the gouge from squeezing out for the wet experiment; therefore, the gouge thickness for wet 152 experiments becomes narrower than that for dry ones. After the precompaction, shear stress was applied 153 with a constant load point velocity of 3 μ m s⁻¹. Velocity step tests were repeatedly conducted after the shear 154 displacement reached 10 mm by abruptly increasing the load point velocity to 33 µm s⁻¹ and decreasing it 155 to 3 µm s⁻¹ after a shear displacement of 1 mm (Fig. 2). The normal stress conditions of 10, 20, 40, and 60 156 MPa were tested for both the dry and wet cases to study the influence of effective normal stress. In addition, 157 several experiments were conducted with different total shear displacements to investigate the evolution of 158 the gouge microstructure in both the dry and wet experiments (Table 1).

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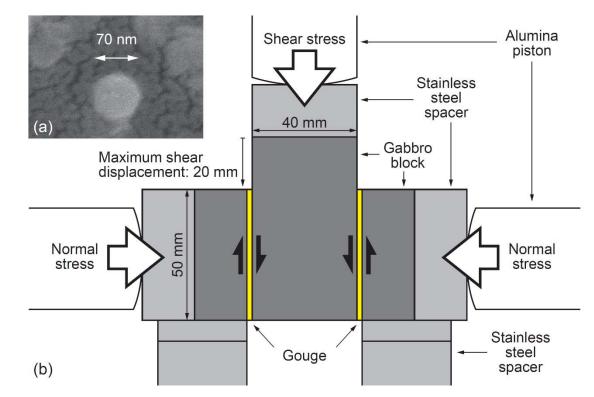


Figure 1: (a) SEM micrograph of synthetic brucite used in this study. (b) Schematic view of the biaxial testing
machine used in this study.

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165 2.2. Data analysis

166 2.2.1. Mechanical data

167 The friction coefficient μ was calculated from the ratio of the shear stress to the normal stress. 168 Note that cohesion stresses were 0.36 and 0.47 MPa for dry and wet cases, respectively, calculated by linear 169 regression of shear stress and normal stress of all the experiments. Because the obtained cohesion stresses 170 were too small to affect the friction coefficients, the cohesion stress was not considered in this study. The 171 shear displacement was corrected using the stiffness of the testing machine $(4.4 \times 10^8 \text{ N m}^{-1})$. The velocity 172 step tests were analyzed using the rate- and state-dependent friction (RSF) law (Dieterich, 1979; Ruina, 173 1983). Before conducting the following analyses, the friction coefficient versus the displacement curve was 174 detrended for the slip-weakening trend, which was obtained from the friction data in the second half of each 175 velocity step of 500 µm shear displacement. Detrended data were fitted to the following the RSF law:

176
$$\mu = \mu_0 + a \ln\left(\frac{V}{V_0}\right) + b_1 \ln\left(\frac{V_0 \theta_1}{d_{c1}}\right) + b_2 \ln\left(\frac{V_0 \theta_2}{d_{c2}}\right), \tag{1}$$

where a, b_1 and b_2 are nondimensional parameters, μ_0 is the steady-state friction coefficient before the velocity step, V_0 and V are the sliding velocities before and after the velocity step, d_{c1} and d_{c2} are the characteristic slip distances, and θ_1 and θ_2 are the state variables. We estimated the effect of elastic interaction due to the machine stiffness on V using the following relationship:

181
$$\frac{d\mu}{dt} = k (V_{\rm lp} - V), \tag{2}$$

182 where V_{lp} is the load point velocity, which was abruptly changed, and k is the system stiffness, which was 183 treated as an unknown parameter (in μ m⁻¹). The Dieterich (aging) law (Dieterich, 1979; Marone, 1998; 184 Ruina, 1983) was used for the state variable in this study.

185
$$\frac{d\theta_i}{dt} = 1 - \frac{V\theta_i}{d_{ci}}, i = 1, 2$$
(3)

186 A MATLAB code, RSFit3000, developed to fit the velocity step and slide hold slide tests (Skarbek and 187 Savage, 2019) was used for the analyses of velocity step tests. Second variables b_2 , θ_2 , and d_{c2} (Blanpied 188 et al., 1998) were only introduced when the experimental data were poorly fitted (upsteps of HTB575 and 189 HTB598; Fig. 4); otherwise, b_2 and θ_2 were treated as 0. The value of a - b $(a - b_1 - b_2)$, or $a - b_1$ 190 was then calculated for each step, which describes the instability of the simulated fault: the state of fault is 191 defined as velocity strengthening and stable when a - b is positive, whereas it is defined as velocity 192 weakening and potentially unstable when a - b is negative. Note that d_c values for the velocity steps whose velocities decreased from 33 to 3 µm s⁻¹ (downsteps) are larger than those for the velocity steps 193 194 whose velocities increased from 3 to 33 μ m s⁻¹ (upsteps). Because we chose to use the Dieterich (aging) 195 law to fit the RSF law, d_c reflects the diameter of the contact area between grains (Dieterich, 1979; Ruina, 196 1983). When the load point velocity is 3 µm s⁻¹, the lifetime of one contact area is longer than that with a 197 load point velocity of 33 μ m s⁻¹. Therefore, the contact diameter, d_c , for the load point velocity of 3 μ m 198 sec⁻¹ (downsteps) is larger than that for 33 μ m s⁻¹ (upsteps). In addition, d_c is also considered to reflect the 199 shear localization (Marone and Kilgore, 1993); when the shear localizes, d_c decreases. Hence, the 200 difference in d_c has qualitative information on the shear localization within the gouge. Although there are 201 still debates on the choice of constitutive laws (Bhattacharya et al., 2015, 2017; Marone, 1998), as all 202 constitutive laws give the same result on a - b, we calculated the value of a - b by using separately 203 obtained a and b with the aging law. The focus of this study will be the a - b value because it plays an 204 essential role in the nucleation process of earthquakes. However, other parameters like d_c and stiffness are 205 also important to the nucleation process, and therefore, those parameters should be assessed in future studies. 206 When the system is velocity weakening, that is, a - b is negative, it starts to vibrate 207 automatically (stick-slip) when the system stiffness is lower than a critical stiffness, whereas conditionally 208 stable sliding is achieved when the system stiffness is higher than a critical stiffness. The critical stiffness 209 k_c can be described as follows when quasi-static stick-slip behavior is assumed:

210
$$k_{\rm c} = \frac{N(b-a)}{d_{\rm c}},\tag{4}$$

where *N* is the applied normal force (Ruina, 1983). Thus, as the normal force *N* applied to the velocityweakening system increases, the system starts to show stick-slip behavior. In other words, the occurrence of stick-slip represents that the system is velocity-weakening. We determined the a - b value for dry experiments with normal stresses of 40 and 60 MPa by simply comparing the averaged friction coefficients during the stick-slip behavior for two velocities based on the following relationship:

216
$$a - b = \frac{\Delta \mu_{\rm ss}}{\Delta \ln V},\tag{5}$$

where $\Delta \mu_{ss}$ and $\Delta \ln V$ are variations in the steady-state friction coefficient and the sliding velocity in the log scale, respectively. In this case, neither *a*, *b*, nor *d*_c can be determined.

219

220 **2.2.2. Microstructure**

In the case of sheet-structure minerals, the friction between basal planes of the crystals [(0001) plane for brucite] is thought to be significant due to their weak bonding. The shear surfaces of the samples recovered from friction experiments using sheet-structure minerals often show smooth surfaces composed of platy particles aligned parallel to the sliding direction (Moore and Lockner, 2004). Further, according to the experiments with natural samples, the aligned platy particles of interconnected talc were reported to contribute to the low friction coefficient of low angle normal faults (Collettini et al., 2009). Because these experiments indicate that the crystal orientation within the gouge has a significant

228 effect on the friction coefficients of sheet-structure minerals, observations of thin sections of recovered

229 samples were conducted after the experiments (Table 1) to investigate the effects of the deformation 230 structures and crystal orientation within the gouges on the frictional behavior. After the experiment, we 231 impregnated the gouge and the blocks with epoxy resin to keep the deformation structures within the gouge. 232 Thin sections parallel to the shear direction and normal to the gouges with a thickness of 30 µm were 233 prepared from the impregnated samples. The scanning electron microscope (SEM, JEOL JXA-8900 at the 234 Atmosphere and Ocean Research Institute, University of Tokyo, Japan) was used to observe the 235 microstructures of the gouges. An accelerating voltage of 15 kV and a beam current of 10.0 nA were used 236 for all backscattered electron (BSE) observations. The crystal orientation was determined with a polarizing 237 microscope at the University of Tokyo, Japan.

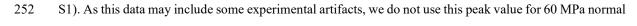
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239 **3. Results**

240 **3.1. Mechanical behaviors**

241 **3.1.1. Friction coefficients**

242 In general, both dry and wet experiments show high friction coefficients at a shear displacement 243 of 1.5-2 mm (hereafter peak friction coefficients) followed by slip-weakening trends with a shear 244 displacement of about 10 mm towards steady state (Figs. 2 and S1). The final friction coefficients at a shear 245 displacement of ~ 20 mm for dry and wet conditions under all normal stress conditions were 0.40 ± 0.04 and 246 0.26±0.03, respectively (Table 1). These final friction coefficients are mostly independent of the applied 247 normal stress (Fig. 3) and consistent with previous experimental results, that is, 0.38-0.46 and 0.28 for dry 248 and wet brucite at an applied normal stress of 100 MPa at room temperature, respectively (Moore and 249 Lockner, 2004, 2007). The friction coefficient for dry experiments is also close to the theoretical value of 250 0.30±0.03 (Okuda et al., 2019). Note that the peak friction coefficient of wet brucite at an effective normal 251 stress of 60 MPa is high because of sudden stress drops in the initial stage of the shear displacement (Fig.



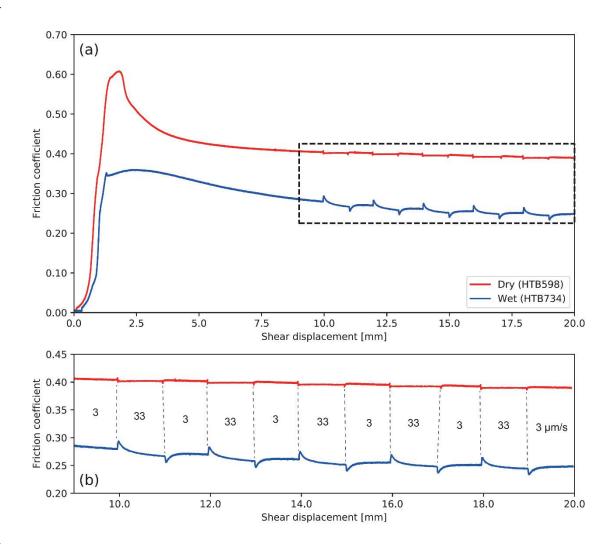
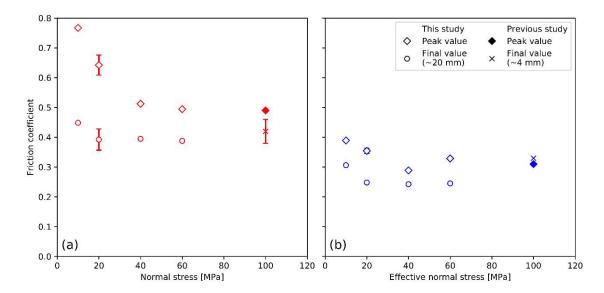


Figure 2: (a) Friction coefficients for dry (HTB598) and wet (HTB734) experiments at a normal stress of 20 MPa.
Slip-weakening behavior was observed after the peak under both dry and wet conditions. (b) Enhanced view of
velocity step sequences as indicated by the dotted square in (a). The velocities at given shear displacements are
displayed between two lines.



261

262 Figure 3: Relationship between normal stress and the peak or final friction coefficients for the dry (a) and wet 263 (b) experiments. Data at a normal stress of 100 MPa were obtained from previous experiments (Moore et al., 264 2001; Moore and Lockner, 2004, 2007; Morrow et al., 2000). The final friction coefficients do not show a clear 265 trend with normal stress. For this study, the error bar represents the one-sigma standard deviation among 266 multiple data. For 100 MPa dry data, the final friction coefficient and the error bar denote the averaged value 267 of stick-slip behavior and its amplitude, respectively. Note that the peak friction coefficient of wet brucite at an 268 effective normal stress of 60 MPa is high because of sudden stress drops in the initial stage of the shear 269 displacement (Fig. S1). As this data may include some experimental artifacts, we do not use this peak value in 270 this study.

271

272 **3.1.2.** Velocity dependencies

For wet experiments, negative a - b values were observed at low normal stresses of 10 and 20 MPa (Figs. 4a and b). However, the a - b values became almost neutral at 40 MPa and positive at 60 MPa. A positive a - b value was consistent with the previous experiments on wet brucite at an effective normal stress of 100 MPa (Moore et al., 2001; Moore and Lockner, 2007). The a - b values obtained for the upsteps and downsteps insignificantly differ (Figs. 4a and b). In the experiments with normal stress conditions of 20, 40, and 60 MPa, the constitutive parameter a is almost constant with 0.0054 for both upsteps and downsteps, whereas b decreases from 0.0064 to 0.0042 and from 0.0076 to 0.0040 for upsteps

and downsteps, respectively, as the normal stress increases (Figs. 4e and f). Accordingly, we concluded

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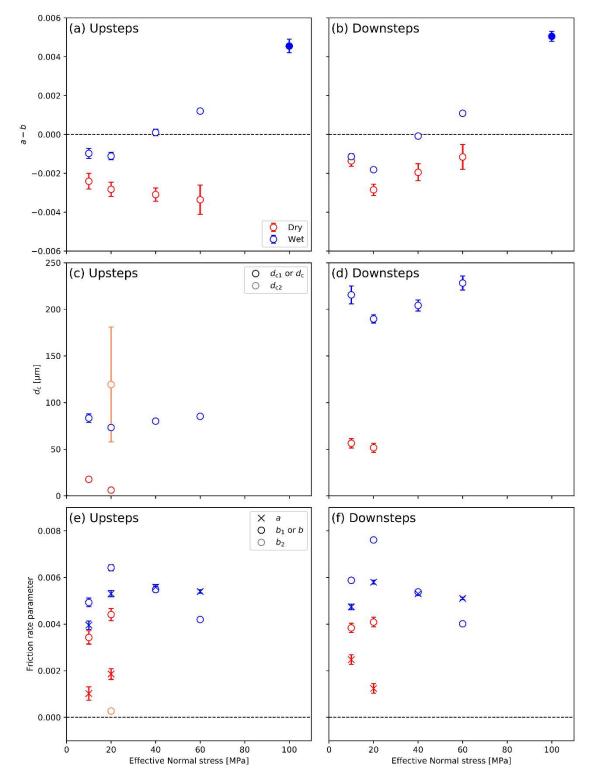
that the decrease in b induces the transition from negative to positive a - b. The d_c values at different

282 effective normal stresses insignificantly differ (Figs. 4c and d).

283 For dry experiments, negative a - b values were obtained at all normal stress conditions (Figs. 284 4a and b). When the normal stress was higher than 40 MPa, stick-slip behavior was observed. This unstable 285 stick-slip behavior was also reported in the case of the dry experiment at a higher normal stress of 100 MPa 286 (Moore and Lockner, 2004; Morrow et al., 2000). No information on a, b, and d_c values for 40 and 60 287 MPa experiments was obtained because of the stick-slip behavior. As shown in the wet conditions, larger 288 $d_{\rm c}$ values were observed for the downsteps (Figs. 4c and d). Note that the second variables b_2 and $d_{\rm c2}$ 289 were introduced in two experiments (HTB575 and HTB598). However, their effects on the earthquake 290 nucleation process, that is, a - b value, are small because the b_2 values are much smaller than b_1 , 291 although d_{c2} value is much larger than d_{c1} (Fig. 4e and Table S1).

The constitutive parameters a and b and critical slip distance d_c of the dry and wet experiments significantly differ. The a, b, and d_c values of the wet experiments are larger than those of the dry experiments (Fig. 4). The critical slip distances d_c of the upsteps and downsteps under wet conditions were 5–15 times and 3–4 times larger than those under dry conditions, respectively.

296



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Figure 4: Results of the velocity step tests. Values of a - b for upsteps (a) and downsteps (b), d_c for upsteps (c), and downsteps (d), a and b for upsteps (e), and downsteps (f). The errors represent the one-sigma standard deviations of all upsteps or downsteps under each experimental condition, including the errors of the nonlinear least-square fitting processes. The a - b values at a normal stress of 100 MPa were obtained from a previous

- study (solid symbol; Moore et al., 2001). Because stick-slip behavior was observed in the dry experiments at normal stresses of 40 and 60 MPa, a - b values were calculated by eq. 5 (Sect. 2.2.1). Second variables b_2 and d_{c2} were introduced for upsteps of the dry experiments at a normal stress of 20 MPa.
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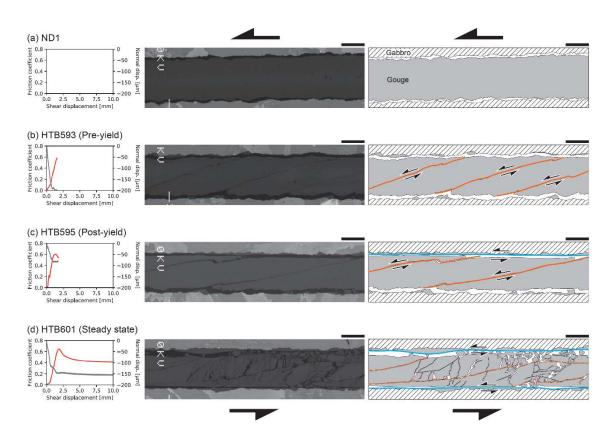
307 **3.2. Microstructure**

308 **3.2.1. Evolution of deformation structures**

309 As all samples (both dry and wet) showed a peak value followed by a transition into the steady 310 state, we chose shear displacements before the peak friction coefficient (pre-yield), after the peak friction 311 coefficient (post-yield), and in the steady state (10 mm) to study the evolution of the deformation structures. 312 Note that the steady state may not be achieved at a shear displacement of 10 mm, but as the final friction 313 coefficients were similar to the friction coefficients at 10 mm shear displacement, here we used the term 314 "steady state" and considered that the microstructure at 10 mm shear displacement might be consistent with 315 the steady state. We followed the description of the microstructure of a sheared gouge by Logan et al. 316 (1979). The results for the dry and wet experiments are shown in Figs. 5 and 6, respectively.

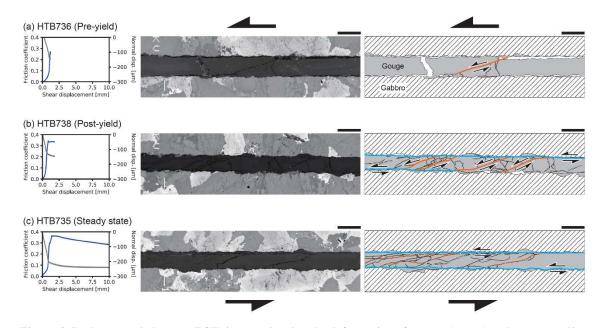
317 Before the shear loading, no shear structure was observed (Fig. 5a). When the shear force was 318 loaded, the Riedel shear propagated in the pre-yield regime, and the gouge thickness decreased rapidly at 319 first (Figs. 5b and 6a). Subsequently, the boundary shear started to develop in the post-yield (Figs. 5c and 320 6b). In the steady state, the boundary shear was created, and the Riedel shear tilted subparallel to the 321 boundary shear (Figs. 5d and 6c). The surfaces of the gabbro blocks were filled with brucite, and the 322 boundary shear was much smoother than the original block surface. These observations are consistent with 323 previous studies (Haines et al., 2013; Kenigsberg et al., 2019, 2020; Logan et al., 1992; Marone, 1998), 324 although clear Y shear and P foliation were not observed in this study. The gouge thickness remained almost 325 constant after the post-yield and steady state, suggesting that the deformation may localize parallel to the 326 shear deformation, i.e., parallel to the boundary shear. The thickness of the entire gouge in the steady state was 400 and 150 µm in the dry and wet cases, respectively (Figs. 5d and 6c). The narrow thickness of the 327

328 gouge in the wet case may result from the leakage of the sample during the experiment, but we did not have 329 any mechanism to prevent the gouge from leaking out. The difference in the entire gouge thickness may 330 not affect the overall frictional characteristics because both dry and wet cases showed the Riedel shear 331 development at first, followed by the boundary shear development. Observation of grain contact is needed 332 for clarification, but it was not possible in this study because the grains were very small (70 nm in diameter). 333



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Figure 5: Backscattered electron (BSE) images showing the deformation of gouges (center) and corresponding interpretive sketches (right) of the dry experiments. The friction coefficients and normal displacement are shown in the left panels using colored and gray lines. The orange lines, blue lines, gray area, hatched area, and white area in the sketches correspond to the Riedel shear, boundary shear, brucite gouge, gabbro block, and epoxy resin, respectively. The orange dot lines are the Riedel shears, which may not be active. The arrows represent the slip directions. The scale bars represent 200 µm.



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Figure 6: Backscattered electron (BSE) images showing the deformation of gouges (center) and corresponding
 interpretive sketches (right) of the wet experiments. See Fig. 5 for descriptions.

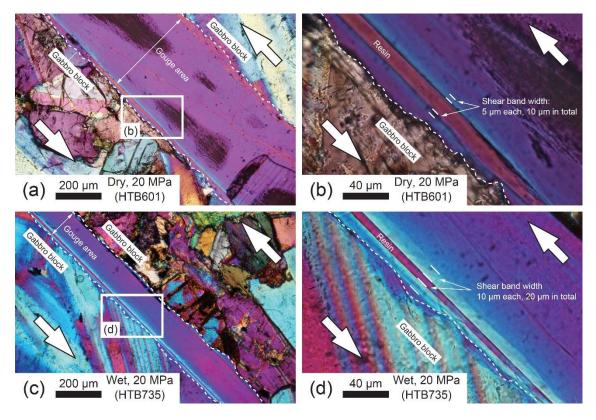
345

346 **3.2.2.** Crystal orientation

Because brucite has a negative elongation (Berman, 1932) and its birefringence is 0.014-0.020(Deer et al., 2013), the interference color of brucite under crossed nicols with the sensitive color plate inserted becomes second-order blue or first-order yellow when the *c* axis of brucite is normal or parallel to the X'-direction of the sensitive color plate, respectively.

In the dry sample (HTB601; Figs. 7a and 7b), a second-order blue line can be observed parallel 351 to the smooth boundary shear, implying that the basal (0001) plane of the brucite particles is aligned along 352 353 the boundary shear parallel to the shear direction. We did not observe any alignment along the Riedel shears, 354 suggesting that deformation along the Riedel shears cannot be dominant at the steady state. Based on the 355 magnified view, the brucite particles are oriented within 10 µm around the boundary shear (Fig. 7b). 356 Because the purple area indicates that the brucite particles are randomly oriented, the shear strain can be 357 localized within a thickness of 10 µm. Hereafter, we call this oriented area "shear band." In the wet samples, 358 the crystals are also oriented along the boundary shear (Figs. 7c and 7d). The thickness of the shear band is 20 μ m (Fig. 7d), which is a little wider than that for the dry experiments. This observation is consistent with the relationship between the shear localization and d_c value (Marone and Kilgore, 1993): the degree of shear localization for the dry sample is higher than that for the wet sample, and d_c for dry sample was smaller than for wet sample (Fig. 4). Note that detailed transmission electron microscopy is required in future studies to confirm the crystal orientation and shear band thickness, as shown in previous studies (Verberne et al., 2014a; Viti, 2011).





366

Figure 7: Observation of the crystal orientation using the polarizing microscope under crossed nicols with the sensitive color plate. The arrows indicate the shear direction. The X'-direction of the sensitive color plate is parallel to the shear direction. (a) Dry experiment with 20 MPa normal stress (HTB601). (c) Wet experiment with 20 MPa normal stress (HTB735). (b and d) Magnified views of (a) and (c), respectively. The shear band thicknesses are indicated in the figures. The white dashed line represents the boundary between the gabbro block and the brucite gouge.

374 4. Discussion

375 4.1. Mechanical weakness of a small amount of brucite

Based on the microstructural observations in Sect. 3.2, the boundary shear is smooth, filling the rough surface of the gabbro block as a "fault mirror" (Siman-Tov et al., 2013). The brucite particles are aligned along the boundary shear, suggesting that the deformation within the narrow shear band is responsible for most of the deformation of the gouge during the steady state. In addition, the constant gouge thickness during the steady state suggests that the gouge deformation occurs parallel to the shear direction, consistent with shear deformation localized within the shear band.

382 Because previous studies showed that a smooth slip surface reduces the friction coefficient 383 compared to a roughened slip surface (Anthony and Marone, 2005), the development of the smooth 384 boundary shear observed in this study would reduce the friction coefficient with increasing shear 385 displacement (Haines et al., 2013). In addition, the slip between the basal planes of sheet-structure minerals 386 also plays an important role for weak friction because the friction between single crystals of sheet-structure 387 minerals has a lower friction coefficient than that of powdered samples (Horn and Deere, 1962; Kawai et 388 al., 2015; Niemeijer, 2018; Okamoto et al., 2019). Based on the observed alignment of the basal plane of 389 brucite within the shear band, the friction between the basal planes of brucite crystals might enhance the 390 weak friction of brucite. Because the preferred planes of nanoparticles tend to be aligned even when the 391 velocity is low (Verberne et al., 2013, 2014b), nanoparticles could contribute to the slip-weakening 392 behavior. Based on these phenomena, we conclude that the mechanical weakness of brucite observed in 393 this study is likely derived from the smooth boundary shear of fine brucite particles and alignment of the 394 basal plane of brucite parallel to the boundary shear.

The results of several previous experimental studies showed that the friction coefficient of a mixture of strong and weak materials inversely correlates with the volume of the weak materials (Giorgetti et al., 2015; Logan and Rauenzahn, 1987; Moore and Lockner, 2011; Niemeijer and Spiers, 2007; 398 Shimamoto and Logan, 1981; Takahashi et al., 2007; Tembe et al., 2010). Based on the maximum amount 399 of brucite in serpentinite, that is ~20 vol. % (Kawahara et al., 2016; Moore et al., 2001), the expected 400 friction coefficient of the antigorite-brucite mixture is 0.53, assuming a simple linear mixing law between 401 the wet friction coefficients of 0.6 for antigorite and 0.26 for brucite. This value is not small, but the bulk 402 friction coefficient of the mixture will decrease if weak brucite crystals are interconnected with each other. 403 The microstructural observations showed that the shear band is less than 50 µm wide (Sect. 3.2.2; Fig. 7); 404 therefore, a narrow network of brucite can decrease the bulk strength. The results of a recent petrographic 405 study of a hydrated paleo-mantle wedge revealed brucite thin films parallel to antigorite particles, 406 suggesting the significant role of brucite in the development of the sheared structure of the antigorite-407 brucite assemblage in the hydrated mantle wedge (Mizukami et al., 2014). Because the maximum thickness 408 of the brucite film in the antigorite-brucite assemblage is several hundred micrometers (Kawahara et al., 409 2016; Mizukami et al., 2014), that is, larger than 50 µm, brucite has the potential to weaken the bulk strength 410 of serpentinite drastically.

411

412 **4.2.** Application to the mantle wedge conditions

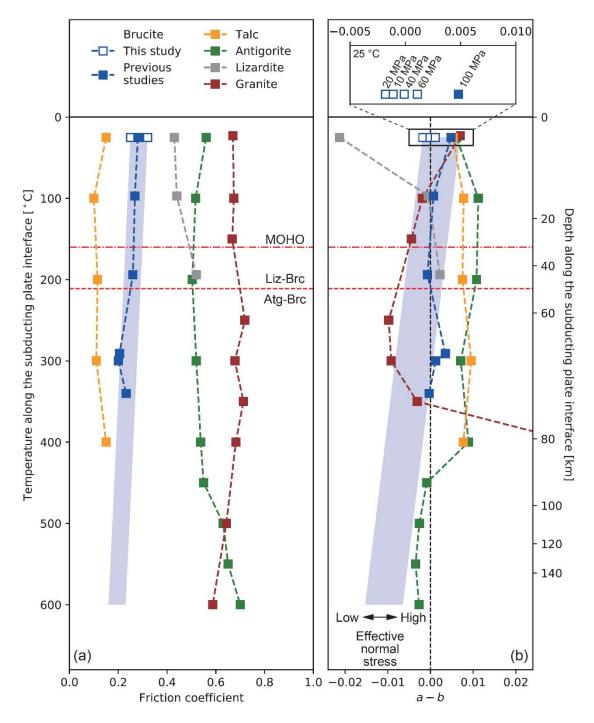
413 When we consider the effect of brucite on the seismic activities in the mantle wedge, the effect 414 of temperature should be taken into account because all our experiments were conducted under room-415 temperature conditions. According to previous experiments on brucite under hydrothermal conditions in 416 which the temperature was varied, the friction coefficient and the a - b values decrease with increasing 417 temperature (Moore et al., 2001; Moore and Lockner, 2007). Because a nearly neutral a - b value was 418 observed at an effective normal stress of 150 MPa and a temperature of 340 °C (Moore et al., 2001), brucite 419 shows unstable behavior under a wide range of temperature-pressure conditions, especially at low effective 420 normal stress. Based on the estimated frictional properties of brucite under the mantle wedge condition, we 421 compared brucite to other mineral phases to interpret the earthquake processes within the mantle wedge422 (Fig. 8).

423 In the mantle wedge, ultramafic minerals, such as olivine, transform into serpentine minerals, 424 such as antigorite, tale, and brucite, due to hydration. In cold subduction zones, such as beneath NE Japan, 425 likely containing brucite under the temperature-pressure conditions of the mantle wedge, the 426 thermodynamically stable mineral assemblages are lizardite-brucite (Liz-Brc) at depths shallower than 50 427 km or antigorite-brucite (Atg-Brc) assemblage under deeper and warmer conditions (Peacock and 428 Hyndman, 1999). Previous experimental studies on antigorite suggested potential seismic activities due to 429 the unstable frictional behavior of antigorite at high temperatures above 450 °C (Okazaki and Katayama, 430 2015; Takahashi et al., 2011) at which crustal (granitic) rock shows stable friction (Fig. 8), whose friction 431 coefficient (0.5–0.7) is not as low as that of brucite (Fig. 8). Although lizardite, which thermodynamically 432 destabilizes at ~200 °C, potentially shows unstable frictional behavior at low temperature (Moore et al., 433 1997), its friction coefficient is 0.4–0.5, lower than that of antigorite but higher than that of brucite (Fig. 8). 434 Therefore, antigorite and lizardite are not preferably deformed if other weaker minerals, such as brucite, 435 are present in continuous fault strands in which the deformation localizes.

436 Another candidate for such a weak mineral stable under mantle wedge conditions is talc. Talc 437 has a low friction coefficient of 0.1–0.2 at low to high temperatures (Fig. 8); therefore, it might contribute 438 to the creep behavior of the San Andreas fault (Moore and Lockner, 2008) or weaken the slab-mantle 439 interface (Hirauchi et al., 2013; Hyndman and Peacock, 2003). However, because talc has a stable frictional 440 behavior at any temperature, leading to aseismic creep (Moore and Lockner, 2008; Sánchez-Roa et al., 441 2017), it cannot nucleate earthquakes. When we consider talc in the mantle wedge, talc is 442 thermodynamically stable at high Si concentrations and temperature. Whereas, the mineral assemblage 443 consists of brucite and antigorite when the Si content and temperature are low (Peacock and Hyndman, 444 1999). Talc was not widely observed in the paleo-mantle wedge exposed in the Shiraga body, central 445 Shikoku, Japan, at the temperature-pressure condition where the antigorite-brucite system is 446 thermodynamically stable (Kawahara et al., 2016; Mizukami et al., 2014). Although only the antigorite exists stably in the antigorite-brucite stability field when the Si content is high, brucite is widely distributed 447 448 in the Shiraga body (~10-15 %), suggesting low Si metasomatism in the shallow hydrated mantle wedge 449 (Kawahara et al., 2016). Hence, brucite can stably exist within the mantle wedge rather than talc. Although 450 talc is still significantly important for the deformation at the subduction plate interface (Hirauchi et al., 451 2013), the possible occurrence of brucite and its weak and unstable frictional characteristics implies that 452 brucite may be a possible control for the seismic activities at the subduction plate interface in the shallow 453 hydrated mantle wedge.

454 The results of recent seismological studies showed that plate interfaces in the shallow mantle 455 wedge have a nearly lithostatic pore pressure due to slab-derived water at various subduction zones such as 456 SE Japan, Cascadia, Central Mexico, and Hikurangi (Audet et al., 2009; Audet and Kim, 2016; Eberhart-457 Phillips and Reyners, 2012; Matsubara et al., 2009; Shelly et al., 2006; Song and Kim, 2012). Such low 458 effective normal stress conditions are conducive for brittle deformation rather than ductile behavior (French 459 and Condit, 2019; Gao and Wang, 2017). Slow earthquakes in the mantle wedge of various subduction 460 zones (Audet and Kim, 2016; Obara and Kato, 2016) might be induced by the low effective normal stress 461 because the low effective normal stress conditions are conducive for the nucleation of slow earthquakes (Liu and Rice, 2007, 2009; Rubin, 2008; Segall et al., 2010). As the a - b value of brucite decreases with 462 463 decreasing effective normal stress, brucite at low effective normal stress possibly causes the nucleation of 464 slow earthquakes in the mantle wedge. Notably, the possible presence of talc or brucite-free antigorite due 465 to high Si content in the vicinity of the slab-mantle interface (Hirauchi et al., 2013; Peacock and Hyndman, 466 1999) might affect the partitioning of deformation (French and Condit, 2019) and the contribution of brucite 467 to the deformation. The mechanisms of nucleation of slow earthquakes are still debated from both 468 theoretical and experimental studies, for example, the dilatancy hardening, the transition from negative to

469	positive $a - b$ value, the slip-weakening, or the slow stick-slip are all considered possible mechanisms
470	(den Hartog et al., 2012; Ikari et al., 2013; Leeman et al., 2016, 2018; Matsuzawa et al., 2010; Okazaki and
471	Katayama, 2015; Rubin, 2008; Segall et al., 2010; Shibazaki and Iio, 2003). As other serpentinite-related
472	minerals show stable frictional behavior, i.e., positive $a - b$, friction experiments with mixtures of brucite
473	and other minerals like talc or antigorite may provide further information on the generation of slow
474	earthquakes. In addition, the linkage between high pore fluid pressure and the effective normal stress is still
475	debated (Hirth and Beeler, 2015; Noda and Takahashi, 2016); therefore, experiments under hydrothermal
476	conditions with high confining pressure and high pore fluid pressure must be conducted in the future.
477	



479

Figure 8: Friction coefficients (a) and velocity dependences (b) of brucite (this study; Moore et al., 2001), talc (Moore and Lockner, 2008), antigorite (Okazaki and Katayama, 2015; Takahashi et al., 2011), lizardite (Moore et al., 1997), and granite (Blanpied et al., 1998). The vertical axes are identical to the temperature gradient along the subduction plate interface in NE Japan (Peacock and Wang, 1999). The red chain horizontal line represents the typical depth of the MOHO. The red dotted horizontal line represents the phase boundary between lizardite– brucite (Liz–Brc) and antigorite–brucite (Atg–Brc; Peacock and Hyndman, 1999). The blue shaded areas are

the estimated frictional characteristics extrapolated from experimental results. With the decrease in the effective normal stress, the a - b value decreases, as indicated by the arrow. This trend was confirmed at room temperature, as shown in the inset at the top of (b) and Fig. 4.

489

490 **5.** Conclusions

491 In this study, the influence of effective normal stress on the frictional characteristics of brucite 492 was experimentally determined under both dry and wet conditions at room temperature. The final friction 493 coefficients of brucite are 0.40 and 0.26 in the dry and wet cases, respectively, independently of the applied 494 normal stress, while the peak friction coefficients are inversely correlated with the applied normal stress. 495 In all dry experiments, velocity-weakening or stick-slip behavior was observed at every normal stress. In 496 the wet experiments, velocity-weakening, -neutral, and -strengthening behaviors were observed at normal 497 stresses of 10 and 20, 40, and 60 MPa, respectively. Combining with the previously reported temperature 498 effect, this result suggests that brucite is weak and unstable under a wide range of temperature-pressure 499 conditions. The microstructural observations reveal that the low friction coefficient and slip weakening 500 from the peak to steady-state friction coefficient are due to the smooth boundary shear and basal plane 501 orientation parallel to the boundary shear. Because the deformation is concentrated within a narrow shear 502 band with a thickness less than 50 μ m, a small amount of brucite can weaken the bulk strength of the 503 antigorite-brucite assemblage. Compared to other serpentinite minerals, brucite is the only mineral that 504 shows both low friction coefficient and velocity-weakening behavior. Hence, we conclude that weak, 505 unstable brucite contributes to the nucleation of slow earthquakes in the shallow hydrated mantle wedge.

506

507 Table 1. Summary of the experimental conditions and results.

Friction coefficient

a - b

Experime nt	Conditio n	Norm al	Final shear displaceme	Pea k	Stead y	Stead y	Upsteps ^a	Downsteps ^a
IIt	11	stress	nt	valu	y state	y state		
		54055	IIt	e	(10	(20		
				-	mm)	(=° mm)		
HTB550	Dry	20	18 mm	0.67	,	0.35 ^b	N/A	N/A
	5	MPa						
HTB575	Dry	20	20 mm	0.68	0.46	0.44	-	-
		MPa					0.0047 ± 0.000	0.0048 ± 0.00
							3	02
HTB580	Dry	10	20 mm	0.77	0.49	0.45	-	-
		MPa					0.0024 ± 0.000	0.0014 ± 0.00
							4	03
ND1	Dry	20	0 mm	N/A	N/A	N/A	N/A	N/A
		MPa						
HTB593	Dry	20	1.5 mm ^c	N/A	N/A	N/A	N/A	N/A
		MPa						
HTB595	Dry	20	$2.0 \ mm^{d}$	0.60	N/A	N/A	N/A	N/A
		MPa						
HTB598	Dry	20	20 mm	0.61	0.41	0.39	-	-
		MPa					0.0010 ± 0.000	0.0009 ± 0.00
							2	02
HTB601	Dry	20	10 mm	0.65	0.42	N/A	N/A	N/A
		MPa						
HTB641	Dry	40	20 mm	0.51	0.41	0.395	-	-
		MPa					0.0031 ± 0.000	
							3°	04 ^e
HTB642	Dry	60	20 mm	0.50	0.40	0.39	-	-
		MPa					0.0034 ± 0.000	0.0012 ± 0.00
							8 ^e	06 ^e
HTB734	Wet	20	20 mm	0.35	0.28	0.25	-	-
		MPa					0.0011±0.000	0.0018±0.00
	TT 7 /	20	10	0.25	0.00	37/1	2	01
HTB735	Wet	20	10 mm	0.37	0.29	N/A	N/A	N/A
		MPa						

HTB736	Wet	20	1.2 mm ^c	N/A	N/A	N/A	N/A	N/A
		MPa						
HTB737	Wet	10	20 mm	0.39	0.32	0.31	-	-
		MPa					0.0010 ± 0.000	0.0011 ± 0.00
							3	02
HTB738	Wet	20	1.8 mm ^d	0.34	N/A	N/A	N/A	N/A
		MPa						
HTB739	Wet	40	20 mm	0.29	0.26	0.24	$0.0001 {\pm} 0.000$	-
		MPa					2	0.0001 ± 0.00
								01
HTB741	Wet	60	20 mm	0.33	0.25	0.25	$0.0012{\pm}0.000$	0.0011 ± 0.00
		MPa					1	01

Note. ^aAll parameters (a, b, and d_c) used for the velocity step tests are listed in Table S1. ^bValue at a shear displacement of 18 mm. ^cShear loading was stopped before the peak friction coefficient was reached. ^dShear loading was stopped shortly after the peak friction coefficient was reached. ^eStick-slip behavior was observed and a - b value was determined by eq. 5.

508

509 Data availability

510 The results of all experimental data are available in the Supporting Information.

511

512 Author contributions

513 H.O. conceptualized this study. H.O. and I.K. conducted the experiments. H.O. and H.S.

514 conducted analyses before experiments. H.O. carried out formal analyses and microstructural analyses. H.O.

515 prepared the original manuscript, which was reviewed and edited by all coauthors. K.K. was the supervisor.

516 I.K., H.S., and K.K. designed the research project.

518 Competing interests

519 The authors declare that they have no conflict of interest.

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527	

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