Electrical conductivity in texturally equilibrated fluid-bearing forsterite aggregates at 800 $^{\circ}$ C and 1 GPa: implications for the high electrical conductivity anomalies in mantle wedges

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Abstract

Aqueous fluids are one of the principal agents of chemical transport in Earth's interior. The precise determination of fluid fractions is essential to understand bulk physical properties, such as rheology and permeability, and the geophysical state of the mantle. Laboratory-based electrical conductivity measurements are an effective method for estimating the fluid distribution and fraction in a fluid-bearing rock. In this study, the electrical conductivity of texturally equilibrated fluid-bearing forsterite aggregates was measured for the first time with various fluid fractions at a constant salinity of 5.0 wt.% NaCl at 1 GPa and 800 °C. We found that the electrical conductivity nonlinearly increases with increasing fluid fraction, and the data can be well reproduced by the modified Archie's law. The three-dimensional (3-D) microstructure of the interstitial pores visualized by the high-resolution synchrotron X-ray computed micro-tomography (CT) shows a change in fluid distribution from isolated pockets at a fluid fraction of 0.51 vol.% to interconnected networks at fluid fractions of 2.14 vol.% and above due to grain anisotropy and grain size differences, accounting for the nonlinear increase in electrical conductivity. The rapid increase in conductivity indicates that there is a threshold fluid fraction between 0.51 and 2.14 vol.% for forming interconnected fluid networks, which is consistent with the 3-D images. Our results provide direct evidence that the presence of > 1.0 vol.% aqueous fluid with 5.0 wt.% NaCl is required to explain the high conductivity anomalies above 0.01 S/m detected in deep fore-arc mantle wedges.

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22	Key points							
23 24 25	1. Electrical conductivity in texturally equilibrated brine-bearing forsterite aggregates was measured at 800 °C and 1 GPa by a novel setup.							
26 27 28	2. 3-D images were obtained by synchrotron X-ray CT to precisely determine the fluid fractions and to visualize the fluid distribution.							
29 30 31	3. Above 1.0 vol.% fluid with 5 wt.% NaCl is required to explain the high conductivity anomalies above 0.01 S/m in deep fore-arc mantles.							
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36 Abstract

37 Aqueous fluids are one of the principal agents of chemical transport in Earth's interior. The precise determination of fluid fractions is essential to understand bulk physical properties, 38 39 such as rheology and permeability, and the geophysical state of the mantle. Laboratory-based 40 electrical conductivity measurements are an effective method for estimating the fluid 41 distribution and fraction in a fluid-bearing rock. In this study, the electrical conductivity of 42 texturally equilibrated fluid-bearing forsterite aggregates was measured for the first time with 43 various fluid fractions at a constant salinity of 5.0 wt.% NaCl at 1 GPa and 800 °C. We found 44 that the electrical conductivity nonlinearly increases with increasing fluid fraction, and the 45 data can be well reproduced by the modified Archie's law. The three-dimensional (3-D) 46 microstructure of the interstitial pores visualized by the high-resolution synchrotron X-ray 47 computed micro-tomography (CT) shows a change in fluid distribution from isolated pockets 48 at a fluid fraction of 0.51 vol.% to interconnected networks at fluid fractions of 2.14 vol.% 49 and above due to grain anisotropy and grain size differences, accounting for the nonlinear 50 increase in electrical conductivity. The rapid increase in conductivity indicates that there is a 51 threshold fluid fraction between 0.51 and 2.14 vol.% for forming interconnected fluid 52 networks, which is consistent with the 3-D images. Our results provide direct evidence that 53 the presence of > 1.0 vol.% aqueous fluid with 5.0 wt.% NaCl is required to explain the high 54 conductivity anomalies above 0.01 S/m detected in deep fore-arc mantle wedges.

55 Plain Language Summary

56 Aqueous fluids are one of the principal agents for transporting material in the Earth's 57 interior. Electrical conductivity measurements can be used to reveal the distribution of water 58 in the Earth's mantle because the presence of a water-rich fluid in a rock can significantly 59 enhance the bulk conductivity. To better constrain the fluid distribution and fraction in 60 mantle wedges, we measured the electrical conductivity of olivine containing minor amounts 61 of a salt-bearing aqueous fluid at high pressure and temperature. The three-dimensional 62 microstructure of the interstitial pores visualized by the high-resolution synchrotron X-ray 63 computed micro-tomography shows that the fluid distribution changes from isolated pockets 64 to interconnected networks with increasing fluid fraction, accounting for the observed nonlinear increase in electrical conductivity. Our results provide direct evidence that the 65 66 presence of > 1.0 vol.% aqueous fluid with 5.0 wt.% NaCl is required to explain the high 67 conductivity anomalies detected in deep fore-arc regions.

68 Key words: Electrical conductivity; Brine-bearing forsterite aggregates; Textural
69 equilibrium; X-ray CT; Mantle wedge; Subduction zones.

70

71 **1. Introduction**

72 In subduction zones, aqueous fluids released from the subducting slab percolate 73 through the mantle wedge, controlling arc magma genesis and element recycling (e.g., 74 Hermann et al., 2006; Iwamori, 1998; van Keken et al., 2011; Tatsumi, 1986). Because the 75 presence of fluid significantly enhances the bulk electrical conductivity of rocks (e.g., Guo et 76 al., 2015; Shimojuku et al., 2012, 2014; Sun et al., 2020), the distribution of fluids in 77 subduction zones may be imaged by magnetotelluric (MT) observations. Recently, a growing 78 number of MT observations have revealed high electrical conductivity anomalies (0.001-1 79 S/m) in the deep fore-arc crusts and mantle wedges in some subduction zones (e.g., Araya 80 Vargas et al., 2019; Pommier & Evans, 2017; Worzewski et al., 2011). Because the 81 temperature is relatively low in fore-arc regions (<700-800 °C, Syracuse et al., 2010), 82 aqueous fluid rather than silicate melt has been assumed to be the highly conductive phase 83 present in the lower crust and mantle wedge, supplied by the subducting slab through a 84 permeable window in the fore-arc mantle wedge (Huang et al., 2019). Although conductive 85 minerals such as sulfide (Ducea & Park, 2000), graphite (Frost et al., 1989; Glover, 1996), 86 and magnetite (Manthilake et al., 2016) have also been suggested to be the cause of the 87 observed high electrical conductivity, their volume fraction may not be sufficiently high to 88 form interconnected networks in rocks.

89 Fluid fraction is a key parameter that controls not only the electrical conductivity, but also other physical properties of fluid-bearing rocks, such as rheology, permeability, and 90 91 seismic wave velocities (Bloch et al., 2018; Rippe et al., 2013; von Bargen & Waff, 1986; 92 Worzewski et al., 2011). The accurate quantitative estimation of the fluid fraction, therefore, 93 is fundamentally important to better interpret geophysical anomalies and to understand the 94 dynamics of material cycling in subduction zones. A number of models with homogeneous 95 and simple fluid geometries have been proposed to describe the fluid fraction dependence of 96 electrical conductivity in fluid-rock mixtures (Archie's law, Archie, 1942; modified Archie's 97 law, Glover et al., 2000; Hashin-Shtrikman upper and lower bound model (HS model), 98 Hashin & Shtrikman, 1962; tube model, Schmeling, 1986; cube model, Waff, 1974). In the

99 cube and HS upper bound models, all grain boundaries are assumed to be completely wetted 100 by a fluid phase. In the tube model, the fluid forms an interconnected tube network at grain 101 edges, and Archie's law is empirical and has been utilized to describe fluid distribution in 102 porous water-saturated sandstone, allowing change in fluid connectivity with increasing fluid 103 fraction (e.g., ten Grotenhuis et al., 2005).

104 At high pressure (P)-temperature (T) conditions, the equilibrium fluid distribution is 105 primarily controlled by the dihedral angle (i.e., wetting angle) between the rock matrix and 106 aqueous fluid. In an ideal system with isotropic interfacial energy and uniform grain size, the 107 system has one true dihedral angle accompanied by curved solid-fluid interfaces with 108 constant mean curvature, and the fluid distribution is homogenous. The fluid forms an 109 interconnected network at all grain edges, as in the tube model, for a dihedral angle below the 110 critical value of 60°. For higher dihedral angles, the fluid fills isolated pockets and a 111 relatively high fluid fraction is required to form interconnected fluid networks (Watson & 112 Brenan, 1987; Holness, 1992, 1993). In reality, however, minerals have anisotropic 113 interfacial energies; thus, the system has a range of true dihedral angles associated with 114 crystallographically controlled faceted interfaces. Pores surrounded by facetted planes can 115 result isolated fluid pockets at grain corners even at dihedral angles smaller than 60°(e.g., 116 Price et al., 2006), affecting the bulk fluid connectivity of the system. In addition, based on 117 the grain size distribution in real rocks, fluid may concentrate in the domains of smaller grains (Wark & Watson, 2000). Therefore, the fluid distribution in a fluid-bearing, deep-118 119 seated rock may deviate from the ideal systems, preventing a precise evaluation of the fluid 120 fraction from MT data using simple theoretical models. Experimental measurements of the 121 electrical conductivity in texturally equilibrated fluid-bearing rocks are necessary to constrain 122 the actual relationship between electrical conductivity and fluid fraction.

123 In the last decade, many laboratory-based electrical conductivity measurements have 124 been conducted in fluid-bearing systems (e.g., Guo & Keppler, 2019; Guo et al., 2015; 125 Shimojuku et al., 2012, 2014; Sun et al., 2020). For example, Shimojuku et al. (2012, 2014) 126 measured the electrical conductivity of a fluid-bearing quartzite as a function of fluid fraction 127 and fluid salinity, and suggested that a high fluid fraction above 32.0 vol.% or a high salinity 128 above 10.0 wt.% is required to account for high conductivity anomalies in the lower crust. 129 However, these measurements were performed with short run durations at constant P–T; thus, the attainment of textural equilibrium was not guaranteed, similar to other previous 130

131 experiments. Olivine is the dominant mineral in mantle wedges, while previous studies 132 utilized quartz (Shimojuku et al., 2012 and 2014), albite (Guo et al., 2015), and 133 clinopyroxene (Sun et al., 2020) as the matrix mineral, which may affect the dihedral angle 134 (i.e., fluid geometry) and fluid chemistry (i.e., charge carriers in the fluid) in the experimental 135 systems. In addition, the lack of the three-dimensional (3-D) analyses of the fluid fraction and 136 pore structures in previous experiments hampers the accurate interpretation of MT data due to 137 an insufficient physical understanding of the fluid fraction dependence of electrical 138 conductivity.

In this study, we measured the electrical conductivity in texturally equilibrated H_2O – NaCl-bearing forsterite (Mg-endmember of olivine) aggregates with various fluid fractions at a NaCl concentration of 5.0 wt.% at 800 °C and 1 GPa. Combined with 3-D images of the fluid distribution determined by synchrotron X-ray computed micro-tomography (CT), the results are used to discuss the fluid fraction dependence of electrical conductivity and estimate the fluid fraction in mantle wedges.

145 **2. Experimental procedures**

146 **2.1. Sample preparation**

147 To avoid the difficulties associated with electrical conductivity measurements in ironbearing systems, synthetic forsterite was used as the starting material. Polycrystalline 148 149 forsterite aggregates were synthesized from iron-free gel powder at 1200 °C and 1.8 GPa in a 150 piston cylinder apparatus with 1/2-inch diameter talc-pyrex assemblies at Bayerisches 151 Geoinstitut, University of Bayreuth. Details of the sol-gel preparation are described in 152 Ohuchi and Nakamura (2006). The gel powders were placed in an oven at 300 °C for 12 h to 153 remove moisture, and were then loaded in a platinum capsule (outer diameter: 5.0 mm) that 154 was sealed by arc welding. The piston cylinder apparatus was first pressurized up to 90% of 155 the target pressure; then, the remaining pressure was applied after heating to the target 156 temperature. During the run, the temperature was monitored and controlled by an S-type 157 thermocouple (Pt–Pt₉₀Rh₁₀). The experiment duration was 24 h at the target temperature. The 158 experiments were quenched to room temperature within 1 min, and the pressure was released 159 slowly. The run products were extracted from the platinum capsules and checked under an 160 optical microscope. The synthesized forsterite grains were then ground in an agate mortar and 161 were sieved to obtain size fractions of 38-53 µm. Moreover, reagent-grade NaCl (Chem.

Pure, 99.999% purity) was dissolved in deionized and distilled water under ambientconditions to obtain a solution with a salinity of 5.0 wt.%.

164 **2.2. Electrical conductivity measurements**

165 Electrical conductivity measurements were conducted at 800 °C and 1.0 GPa in a 166 piston cylinder apparatus with 3/4-diameter talc-pyrex assemblies at Bayerisches Geoinstitut, 167 University of Bayreuth. The configuration of the cell assemblies was as described by Guo 168 and Keppler (2019). A 1-mm diameter platinum rod and a Pt₉₅Rh₅ capsule with an outer 169 diameter of 5.0 mm and inner diameter of 4.4 mm were employed as the inner and outer electrodes, respectively (Figure 1). A ceramic disk with 1 mm thickness was placed at the 170 171 bottom of the Pt₉₅Rh₅ capsule. The forsterite powder and saline fluid of each experiment (initial fluid fractions: 0.5 wt.%, 2.5 wt.%, 5.0 wt.% and 10.0 wt.%) were loaded into the 172 173 capsule between two alumina disks with 1-mm-diameter holes containing the platinum rod 174 inner electrode. A boron nitride disk at the end of the capsule provided the seal. A thin 175 ceramic Al₂O₃ tube tightly fitted around the Pt rod and the central hole of the Pt₉₅Rh₅ lid was 176 served as an insulator. Two Pt wires with a diameter of 0.35 mm were welded to the inner 177 and outer electrodes and connected to a Solartron 1260 impedance analyzer for measuring the 178 conductivity. The whole capsule was surrounded by another shell of boron nitride to improve 179 sealing. The procedures for increasing temperature and pressure were the same as described 180 above. The experiments were conducted at 800 °C and 1 GPa for 8 days, and the impedance 181 spectra were measured every day using an alternating current voltage of 500 mV sweeping 182 from 10 MHz to 1 Hz.





184 Figure 1. Sample characterization. (a) Capsule recovered after an electrical conductivity measurement with a 185 final fluid fraction of 15.05 vol.%. (b) Representative backscattered electron (BSE) image of the cross section of 186 the recovered capsule with a final fluid fraction of 15.05 vol.%. (c) BSE image of the recovered sample with a 187 final fluid fraction of 2.14 vol.%. (d) BSE image of the recovered sample with a final fluid fraction of 15.05 188 vol.%. The fluid pores were filled with epoxy resin during sample preparation. The orange marks represent the 189 forsterite-forsterite triple junctions with intersection angles of $\sim 120^{\circ}$, suggesting the attainment of 190 textural equilibrium. (e) Secondary electron (SE) image of the recovered sample with a final fluid fraction of 191 15.05 vol.%, showing small dihedral angles between forsterite grains and fluids. (f) Representative histogram of 192 the grain size distribution, normalized by the mean grain size of the recovered sample with a final fluid fraction 193 of 15.05 vol.%. The grain size is concentrated with a peak around the mean grain size. Abbreviation: Fo =194 forsterite.

195 The bulk conductivity (σ_b) of the whole cell was calculated from the resistance and 196 the dimensions of the cell, using the equation for the cell constant in cylindrical geometry:

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$$\sigma_b = \frac{\ln(r_o/r_{i)}}{2\pi l R},$$
 (1)

198 where the cylindrical electrodes have an outer radius r_o , and inner radius r_i , l is the length of 199 the sample, and the measured electrical resistance is R. The electrical conductivities 200 measured in this study were usually very high so that the background conductivity of 201 assemblies was negligible for all investigated systems (Guo & Keppler, 2019; Ni et al., 2011). 202 The two-electrode method employed in this study requires a correction for the resistance of 203 the electrode wires sample if the measured resistance of the sample is very low (Pommier et 204 al., 2010). We utilized the short-circuit method introduced by Ni et al. (2011) to correct our 205 measured data.

206 **2.3. Scanning electron microscope (SEM) observations**

207 The experiments were immediately quenched to room temperature at the end of 208 experimental run within 1 min by turning off the heating power after the last electrical 209 conductivity measurement. The recovered capsules were cut open using a diamond wire saw 210 to expose the run products. The run products were then impregnated with epoxy resin in a 211 vacuum box and polished down to 1.0 µm surface roughness with alumina powder and 212 subsequently to 0.06 µm by colloidal silica suspension. The polished cross sections were 213 observed using a field emission-type SEM (FE-SEM; JEOL JSM-7100F) with an accelerating 214 voltage of 15 keV. The dimensions of the samples (i.e., outer radius r_o , inner radius r_i , and 215 length *l*) were carefully measured in backscattered electron (BSE) images.

216 **2.4. Synchrotron X-ray microtomography**

In order to determine the fluid distribution and fraction, the recovered samples were prepared for X-ray CT analysis with synchrotron radiation. Microtomography of run products was conducted at the undulator beam line BL20XU at the Spring-8 facility, Hyogo, Japan. The cross-section of the monochromatic X-ray beam was approximately $2 \text{ mm} \times 1 \text{ mm}$ at 240 m from the light source (around the sample position). The highly collimated undulator radiation from the low emittance storage ring used in this study is very suitable for high spatial resolution tomography. The undulator gap (K-value) was properly tuned at each energy to obtain a relatively large field of view in the vertical direction; that is, the selected energy was slightly lower than the odd-order peaks of the undulator radiation. Therefore, the flux density of the monochromatic beam was approximately 5×10^{12} photons s⁻¹ mm⁻² (Uesugi et al., 2001) at the sample position. The contrast resolution of the X-ray CT system in SPring-8 has been reported by Uesugi et al. (1999). For each recovered sample, around 1,800 CT images were taken by X-ray CT, and a considerable sub-volume of the run products was extracted from these images, avoiding any cracks, to precisely evaluate the fluid fraction.

231 **2.5. Determination of water content**

232 The water content in the forsterite aggregate of one recovered sample (Sample No. 233 ECM-3) was measured using a Thermo Scientific Nicolet iN10 FT-IR Microscope (FTIR) at 234 Tohoku University. A doubly polished section of the sample, with a thickness of ~300 µm, 235 was prepared for the FT-IR measurement. The unpolarized infrared spectra of the sample 236 were obtained from 17 sampling points. At several points measurements were carried out 237 with either increasing aperture size (from 50×50 to 400×400 µm) or scanning time (from 3 238 to 25 s), to evaluate the influence of aperture size or scanning time on measured content. 239 Then 10 sampling points were measured with a constant aperture size of $200 \times 200 \ \mu m$ and scanning time of 12 s. The water content was calculated from the average of the measured 240 241 absorbance, using the integrated extinction coefficients of Bell et al. (2003) and an 242 orientation factor of 1/3.

3. Results

244 **3.1.** Micro-textures and fluid fractions of the recovered sample

245 The run products are composed of sintered aggregates of forsterite and interstitial pores, which were occupied by saline aqueous fluid (Figure 1). Textural maturation was well 246 247 established within the run duration of 8 days. The presence of many forsterite triple junctions 248 with angles of ~120° (Figure 1-d) indicates the attainment of balance of interfacial tensions at triple junctions (Huang et al., 2020; Liu et al., 2018). The grain size distribution determined 249 250 from BSE images by Image-J shows a narrow grain size distribution with a peak around the 251 mean grain size (Figure 1-f), in agreement with textural equilibrium (Faul, 1997; ten 252 Grotenhuis et al., 2005). A thin reaction layer with a thickness of less than 100 µm was 253 observed between the forsterite and the Al₂O₃ disk. Although this layer contains interstitial

pyroxene along with relict olivine, its influence on the bulk electrical conductivity should be
negligible, as all these phases are Fe-free and therefore poor conductors (e.g., Dai & Karato,
2009).

257 The fluid fractions of the experiments were precisely determined using the 3-D 258 images that were obtained from synchrotron X-ray CT of the run products. The fluid fractions 259 of the samples before and after electrical conductivity measurement are shown in the Table 1. 260 Representative 3-D images of the sub-volumes of the recovered samples are shown in the 261 Supporting Information Figure S1. The fluid fraction in the recovered samples is always 262 smaller than the initial fluid fraction, with the difference being up to 12.37 vol.%. Although 263 the initial fluid fraction was precisely controlled, the fluid loss at high P-T conditions was 264 inevitable because of the initially imperfect sealing of the experimental setup, especially 265 during the first day. After this period, the fluid fraction likely remained constant, as discussed 266 below, based on the time-series electrical conductivity data (Figure 2). The fluid fractions 267 estimated from the recovered samples represent the fluid fractions during the last 268 measurements of electrical conductivity.

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Table 1. Summary of experimental conditions and results.

	Run No.	T (°C)	P (GPa)	wt.% of fluid (before EC)	vol.% of fluid (before EC)	vol.% of fluid (after EC)	Salinity (wt.%)	σ (s/m)	logσ (S/m)
	ECM-6	800	1	0.50	1.7(0.1)	0.51(0.05)	5.00	1.65E-03	-2.78
	ECM-4	800	1	2.50	8.0(0.1)	2.14(0.05)	5.00	3.56E-02	-1.45
	ECM-3	800	1	5.00	15.2(0.1)	8.14(0.05)	5.00	2.41E-01	-0.62
270	ECM-5	800	1	10.00	27.4(0.1)	15.05(0.05)	5.00	4.80E-01	-0.32

Note: The volume fractions of fluid after the experiments were determined from the recovered samples by usingsynchrotron X-ray CT.

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Figure 2. Conductivity variation with run time in the forsterite $-H_2O$ -NaCl system at 800 °C and 1 GPa. The fluid fractions determined in the recovered samples are also shown in each panel. Note that the vertical scale is different for the sub-figures **a**, **b** and **c**, **d**.

278 **3.2. Electrical conductivity variation with time**

279 Figure 2 shows the variation in electrical conductivity during each experiment. 280 Experiments were designed to run for 8 days to guarantee textural equilibration, and the 281 electrical conductivity of the samples were measured each day during the run. The 282 experiment with the lowest fluid fraction of 0.51 vol.%, however, was merely run for only 5 283 days because the thermocouple failed on the fifth day. In all experiments, the conductivity 284 strongly decreased during the first day and then gradually stabilized over time. Most likely, 285 the strong change in conductivity at the beginning of the experiment is related to fluid loss, 286 while the more gradual change after the first day may be related to textural changes.

The conductivity in all systems was essentially stable after 72 h, which indicates that both fluid fraction and fluid geometry remained constant (e.g., Figure 1-d). Therefore, we consider the electrical conductivity measured just before quenching as the electrical conductivity at textural equilibration.

3.3. Electrical conductivity under textural equilibrium conditions

The impedance spectra obtained in the last electrical conductivity measurements of 292 293 each experiment are shown in Figure 3. For the experiments with fluid fractions of 0.51, 2.14, 294 and 8.14 vol.%, the electrical impedance spectra contain well-developed and complete semi-295 circle arcs at high frequencies. In these cases, the cell resistance can be obtained from the diameter of the semicircle. For the highest fluid fraction of 15.05 vol.%, the semi-arc 296 297 disappeared completely and was replaced by a segment under the real axis (Z'), which was most likely caused by inductance of the Pt wires. In this case, the cell resistance was directly 298 299 obtained from the intercept with the real axis (Z''=0).



301 Figure 3. Impedance spectra of samples with textural equilibrium at different fluid fractions in the forsterite-302 H₂O-NaCl system at 1 GPa and 800 °C. (a) At the lowest fluid fraction of 0.51 vol.%, the sample conductivity 303 is governed by the interior of the forsterite grains, and the resistance may be obtained either from arc fitting or 304 from the intercept with the real axis. (b and c) At higher fluid fractions, the sample conductivity is governed 305 mostly by the fluid-bearing grain edges, and the resistance may be obtained either from arc fitting or from the 306 intercept with the real axis. (d) At the highest fluid fraction, the resistance may be determined from the Z' 307 intercept of the spectrum (where Z''=0). (e) Comparison of the electrical resistances in all investigated fluid 308 fraction systems. The electrical resistance of different fluid fractions shows different magnitudes.

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310 It is expected that the conductivity circuits in forsterite aggregates depend on the geometrical distribution of the present fluid (Roberts & Tyburczy, 1999; Shimojuku et al., 311 312 2012). The impedance spectra of the systems in this study are governed by a combination of 313 resistor and capacitor circuits. From a combination of the measured impedance spectra 314 (Figure 3) and visualized 3-D fluid distributions (Figure 4), the conduction circuits in this 315 study can be determined. In low fluid fraction systems with isolated fluid pockets, the sample 316 conductivity is mainly governed by the interior of the forsterite grains, and the electrical 317 circuit is composed of the response of the grain and fluid in a series circuit. In high fluid 318 fraction systems with an interconnected fluid network, the sample conductivity is mostly 319 governed by the fluid tube, and the electrical circuit is composed of the response of the grain 320 interior and fluid-filled grain edges in a parallel circuit.



322 323 324 Figure 4. Three-dimensional (3-D) images of fluid-bearing forsterite aggregates after electrical conductivity measurements. (a-d) 3-D images of sub-volumes of recovered samples. (a1-d1) 3-D images of the fluid distributions extracted from a-d. Blue areas represent the forsterite aggregates; red areas represent the interfaces 325 326 between fluids and forsterite crystals; black areas represent the interior of fluid channels. The fluid distribution is relatively localized on smaller scales, and homogenous on larger scales for all samples. The spaces between 327 the fluid are occupied by olivine crystals. The fluid in a1 occurs mostly as isolated fluid pockets. The fluid in b1 328 is locally interconnected. The fluids in c1 and d1 are well interconnected. In c1 and d1, there is an obvious 329 bottleneck effect, i.e. the interconnected networks are composed of coarse channels (white double-headed 330 arrows) and fine channels (white arrows). The fluid fraction for each sample is also shown. The cube size of all 331 3-D images is 100 μ m \times 100 μ m \times 100 μ m.

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333 **3.4. 3-D images of fluid distribution**

334 The fluid distribution in the recovered samples was visualized using the software ParaView for the sub-volume 3-D CT images (Figure 4). The fluid distribution is relatively 335 336 localized on smaller scales, and homogenous on larger scales for all samples. For the system with the lowest fluid fraction of 0.51 vol.%, the aqueous fluid occurred mostly as isolated 337 338 fluid pockets (Figure 4-a1). In contrast, the aqueous fluid was locally interconnected via 339 tubular pathways in the system with a fluid fraction of 2.14 vol.% (Figure 4-b1). For the higher fluid fractions of 8.14 and 15.05 vol.%, the interstitial fluid was well interconnected 340 341 (Figure 4-c1, d1), with the diameter of interconnected tubes increasing with increasing fluid 342 fraction.

343 **3.5. Water content in forsterite**

The infrared absorption spectra in the range of from 3100 to 3800 cm^{-1} collected on 344 10 sampling points with the same aperture size of $200 \times 200 \ \mu m$ and scanning time of 12 s 345 346 are shown in Figure 5. The spectra show sharp absorption peaks derived from structurally bound water in forsterite were observed between 3500 and 3750 cm⁻¹. Moreover, a band at 347 3300 cm^{-1} and a broad absorption background at $3100-3800 \text{ cm}^{-1}$ are also visible and most 348 likely represent free water in fluid inclusions in the forsterite crystals and water in some 349 350 microcracks, grain boundaries, and dislocations of the forsterite aggregate, respectively. The entire absorption range of $3100-3800 \text{ cm}^{-1}$ was integrated to calculate the bulk water content 351 352 of the forsterite aggregate (Figure 5-a, e.g., Ohuchi & Nakamura, 2007). In contrast, to calculate the water content in the forsterite lattice, a linear background was subtracted from 353 the sharp bands at $3500-3750 \text{ cm}^{-1}$ (Figure 5-a) and the remaining absorption was integrated, 354 355 as shown in in Figure 5-b.



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Figure 5. Unpolarized FTIR absorption spectra of the recovered sample ECM-3. The spectra were collected from 10 sampling points with the same aperture size of $200 \times 200 \ \mu\text{m}$ and scanning time of 12 seconds. The sample thickness is ~300 μm . (a) Absorbance spectra in the range of 3100–3800 cm⁻¹. The total area of all absorption peaks between 3100 to 3800 cm⁻¹ was integrated to calculate the water content of the forsterite aggregates. The characteristic peaks of the absorption spectra were divided into three types: spa–1, spa–2, and spa–3. Spa–1 at 3300 cm⁻¹ is the absorption peak for aqueous fluids in the fluid inclusions of the forsterite

363 crystals; spa–2 at 3500–3750 cm⁻¹ shows the absorption peaks for OH in the forsterite lattice; and spa–3 shows a 364 broad absorption background at 3100–3800 cm⁻¹, which can most likely be attributed to water in some 365 microcracks, grain boundaries, and dislocations, rather than structurally bound water. (b) Absorption spectra 366 between 3500 to 3750 cm⁻¹ extracted after baseline correction. The area of these absorption peaks was 367 integrated to calculate the water contents in the forsterite lattice.

368 To evaluate the effect of the aperture size or scanning time on the bulk water content and forsterite lattice water content, up to 17 infrared absorption spectra were collected in the 369 range of 3100 to 3800 cm⁻¹ with various aperture sizes or scanning times. Their respective 370 influence on the water content is shown in Figure S2 and S3 of the Supporting Information. 371 372 For the 10 sampling points with the same aperture size of $200 \times 200 \,\mu\text{m}$ and scanning time of 373 12 s, the bulk water content in the forsterite aggregates ranges from 415 to 512 ppm H_2O 374 with an average of 461 ppm, while the water content in the forsterite lattice ranges from 137 375 to 163 ppm H₂O with an average of 149 ppm. The bulk water content showed a wider range 376 compared to the water content in the forsterite lattice because of inhomogeneity of the sample.

377 **4. Discussion**

4.1. Fluid fraction dependence of electrical conductivity

379 In Figure 6-a, the electrical conductivities measured in this study are plotted as a function of the fluid fraction as estimated from the CT data. The electrical conductivity 380 steeply increased from 1.65×10^{-3} S/m at 0.51 vol.% to 3.56×10^{-2} S/m at 2.14 vol.%, but 381 increased more slowly with increasing fluid fractions above 2.14 vol.%. At fluid fractions 382 above 8.14 vol.%, the electrical conductivity increased gently. This fluid fraction dependence 383 384 of the electrical conductivity may reflect the changes in the fluid distribution in the forsterite 385 matrix. The 3-D images showed that the fluid was distributed as isolated pockets at 0.51 386 vol.% (Figure 4-a1), while it formed an interconnected network at fluid fractions above 2.14 387 vol.% (Figure 4-b1). This indicates that there is a threshold fluid fraction between 0.51 and 388 2.14 vol.%. Once the fluid fraction exceeds this critical value, the fluid establishes an 389 interconnected network, shifting the predominant conduction pathway from the forsterite 390 interior to the saline aqueous fluid network. The drastic change in fluid connectivity, therefore, may exert a primary control on the fluid fraction dependence of electrical 391 conductivity. At fluid fractions above 8.14 vol.%, the further increase in conductivity may be 392 393 related to the enlargement of the average diameter in the fluid network.



395 Figure 6. Fluid fraction dependence of the bulk conductivity. (a) Electrical conductivity with textural 396 equilibrium as a function of fluid fraction in the forsterite-H₂O-NaCl system at 800 °C and 1 GPa. The 397 electrical conductivities for single-crystal dry olivine (C92, Constable et al., 1992), dry forsterite aggregate (Y17, 398 Yoshino et al., 2017), 100–200 ppm H₂O-bearing olivine aggregate (W06, Wang et al., 2006), and saline 399 aqueous fluids (GK19, Guo & Keppler, 2019) are shown along with our data. The fluid fraction dependence of 400 several electrical conductivity models (modified Archie's law, Glover et al., 2000; HS upper and lower model, 401 Hashin & Shtrikman, 1962; tube model, Schmeling, 1986) are shown in the diagram. The hydrogen-bearing 402 forsterite conductivity in ECM-6 of this study was used as the solid phase conductivity in all models. (b) Effect 403 of iron on the bulk electrical conductivity in the different models. We used the iron-bearing hydrous olivine 404 conductivity (Wang et al., 2006) and hydrogen-bearing forsterite conductivity in ECM-6 of this study for the 405 models. The ranges of the high electrical conductivity anomalies in the mantle wedge are shown as light green 406 areas.

407

408 **4.2. Charge carriers in the aqueous fluid**

409 Because the conductivity in our study was primarily controlled by the interconnected 410 aqueous fluids, considering the charge carriers in the fluid is important. For the fluids with 5 411 wt.% NaCl in our study, Na⁺ and Cl⁻ ions produced by dissociation of NaCl are the dominant 412 charge carriers. Manning (2013) reported that the dissociated fraction of NaCl in aqueous 413 fluid at 1 GPa and 800 °C is ~0.82 with a salinity lower than 5.8 wt.%, suggesting that most 414 of the NaCl in the aqueous fluid was dissociated in our experiments. Although their 415 contribution to the bulk electrical conductivity is likely limited, H⁺ and OH⁻ produced by 416 minor H_2O self-dissociation at elevated temperatures and pressures may also play a role as 417 charge carriers (Manning, 2018). Elevated solubility of forsterite in a NaCl-bearing aqueous 418 fluid was observed in previous experimental studies (Macris et al., 2020), suggesting that the solution in our study contained Mg²⁺ and Si⁴⁺-bearing species. At a low concentration and 419 420 nearly neutral pH, the predominant species of dissolved silica is monomeric orthosilicic acid 421 (Manning, 2018), which does not contribute to electrical conduction. Although there may 422 have been several ionic species in the fluid in our samples, the bulk electrical conductivity is 423 most likely controlled by the motion of the dominant Na^+ and Cl^- ions within the 424 interconnected fluid network in a saline aqueous fluid-bearing system (Guo & Keppler, 2019).

425 4.3. Comparison with the electrical conductivity of fluid-free forsterite aggregates and 426 with pure saline fluids

The bulk electrical conductivities measured in this study in aggregates of forsterite containing saline fluid should fall within the range of inherent conductivities in both endmember systems because no significant charge carriers are released into the aqueous fluid from forsterite grains, as discussed in the Section 4.2. Therefore, to verify the reliability of our measurements, we compared our results with the electrical conductivities of saline fluid
and of fluid-free forsterite aggregates measured in previous studies (Figure 6-a). For saline
fluid, the electrical conductivity of aqueous fluid with 5.8 wt.% NaCl measured at 1 GPa and
800 °C was adopted for comparison (Guo & Keppler, 2019). Its reported conductivity is 35
S/m, and significantly higher than the values measured in this study even at the highest fluid
fraction of 15.05 vol.%.

437 Because there is no data of conductivities of hydrous iron-free forsterite aggregates, we compared our data with the conductivity of dry forsterite aggregates at 800 °C and 1 GPa, 438 as calculated with the equation given by Yoshino et al. (2017). Its conductivity of 2.46×10^{-6} 439 440 S/m is almost three orders of magnitude smaller than the conductivity measured at the lowest 441 fluid fraction of 0.51 vol.% in this study. Water dissolved in forsterite is expected to reduce 442 this difference because it strongly increases the conductivity. The electrical conductivity range of hydrous iron-bearing olivine ($X_{\text{Fe}} = \text{Fe} / (\text{Mg} + \text{Fe}) = \sim 0.1$) with 100–200 ppm H₂O at 443 800 °C (Wang et al., 2006, $3.0-4.0 \times 10^{-3}$ S/m) is two to three orders of magnitude higher 444 than that of dry olivine at the same temperature (Constable et al., 1992, 8.0×10^{-6} S/m). 445 Therefore, the conductivity of hydrous forsterite aggregates with 137-163 ppm H₂O, as in 446 447 this study, may be comparable to or slightly smaller than that of the fluid-bearing hydrous 448 forsterite aggregates at the lowest fluid fraction of 0.51 vol.%. These comparisons show that 449 the conductivities measured in this study are between those in fluid-free forsterite aggregates 450 and those in saline fluids reported in previous studies, demonstrating the reliability of our 451 measurements.

452 **4.4.** Comparison with models of electrical conductivity in the fluid-bearing rocks

In this section, we compare the electrical conductivities measured in this study to those predicted using models with simple fluid geometries. The HS upper and lower bound model (Hashin & Shtrikman, 1962), the cube model (Waff, 1974), the tube model (Schmeling, 1986), and a modified Archie's law (Glover et al., 2000) have been proposed to describe the effect of the fluid fraction on bulk electrical conductivity.

The HS model (Hashin & Shtrikman, 1962) provides the upper (HS⁺) and lower bounds (HS⁻) on the bulk conductivities of fluid–rock mixtures assuming extreme cases of fluid geometry, i.e., completely interconnected fluid films along grain boundaries (dihedral 461 angle = 0°) or isolated fluid pockets (dihedral angle > 60°). The two extremes are described 462 by the following equations:

$$HS^{+}:\sigma_{b} = \sigma_{f} \left(1 - \frac{3(1-\phi)(\sigma_{f} - \sigma_{s})}{3\sigma_{f} - \phi(\sigma_{f} - \sigma_{s})} \right),$$
(2)
$$HS^{-}:\sigma_{b} = \sigma_{s} \left(1 + \frac{3\phi(\sigma_{f} - \sigma_{s})}{3\sigma_{s} + (1-\phi)(\sigma_{f} - \sigma_{s})} \right),$$
(3)

463 where $\sigma_{\rm f}$, $\sigma_{\rm s}$, and ϕ are the electrical conductivity of the fluid, the solid matrix, and the fluid 464 fraction, respectively. The cube model (Waff, 1974) assumes that cubic grains with low 465 conductivity and uniform grain size are surrounded by a high-conductivity fluid layer of 466 uniform thickness. The result of this model is similar to that of the HS⁺ model. Its 467 approximate bulk conductivity is described as follows:

$$\sigma_b = \frac{2}{3}\sigma_f \phi + \sigma_s (1 - \phi). \tag{4}$$

In the tube model (Schmeling, 1986), the fluid is assumed to be distributed as a rectangulartube network along the grain edges, but with unwetted grain boundaries:

$$\sigma_b = \frac{1}{3}\sigma_f \phi + \sigma_s (1 - \phi) \,. \tag{5}$$

470 Based on the above equations, it is obvious that the tube model gives a lower 471 conductivity than the cube model.

472 Archie's law is an empirical relation between the fluid fraction and bulk conductivity 473 (Archie, 1942), which is commonly applied to describe the electrical conductivity of a porous 474 sandstone. The conventional Archie's law is designed for a conductive phase saturating a non-conductive matrix at shallow depths and low temperatures, where the conductivity of the 475 476 matrix is negligible, whereas the conductivities of rock-forming minerals at high P-T 477 conditions may significantly contribute to the bulk conductivity (Glover et al., 2000). To 478 include the effect of solid conductivity, a modified form of the conventional Archie's law 479 (modified Archie's law) was proposed by Glover et al., (2000) as follows:

$$\sigma_b = \sigma_f \phi^m + \sigma_s (1 - \phi)^p, \tag{7}$$

$$p = \frac{\log(1 - \phi^m)}{\log(1 - \phi)},\tag{8}$$

where the exponents *m* and *p* represent the connectivity of the fluid and solid phases,respectively. This model allows a change in fluid connectivity according to the fluid fraction.

In Figure 6-a, our results are compared with those of the models. For the model 482 calculations, we assumed $\sigma_f = 35$ S/m for the conductivity of aqueous fluid with a salinity of 483 5.8 wt.% (Guo & Keppler, 2019). For σ_{s} we used the conductivity of the system with the 484 lowest fluid fraction in this study (i.e., $\sigma_s = 1.65 \times 10^{-3}$ S/m) in which the fluid was not 485 486 interconnected as revealed by 3-D images (Figure 4-a1); thus, the effect of fluid on the 487 conductivity could be minor. Our conductivities fall within the range of those of the HS 488 models and thus do not violate the physical bounds. Because the dihedral angle defined by 489 the curved olivine-fluid interfaces is smaller than 60° in the olivine + saline fluid system at 490 800 °C and 1 GPa with a NaCl concentration of 5.0 wt.% (Huang et al., 2019), the fluid in 491 our experiments is expected to be distributed along the triple junctions and result in mostly 492 the same conductivities as in the tube model. However, our conductivities are actually 493 smaller than those of the tube model (Figure 6-a). There are two plausible reasons for this 494 discrepancy. First, this discrepancy may be attributed to the interfacial anisotropy of forsterite 495 in our system, which would lead to a range of true dihedral angles having the different 496 wetting properties in different crystallographic orientations. Along with the curved forsterite-497 fluid interfaces, for example, faceted planes were observed in all the recovered samples 498 (Figure 1). At low fluid fractions, the fluid pores surrounded by faceted planes have difficulty 499 forming an interconnected network even at dihedral angles smaller than 60° (Price et al., 500 2006), while they can be interconnected at relatively high fluid fractions depending on the 501 dihedral angle. This could result in the establishment of fluid interconnections at a range of 502 fluid fractions, which is consistent with the change in fluid connectivity according to the fluid 503 fractions confirmed by our CT images. Second, the normal grain growth driven by Ostwald 504 ripening results in a heterogeneous grain size distribution ranging from 5 to 100 µm in our 505 systems (Figure 1), which plays a significant role for the fluid distribution and thus the 506 permeability (e.g., Cerpa et al., 2017; Wark & Watson, 2000). The fluid is expected to be 507 concentrated in domains with finer grain sizes, assuming the presence of curved interfacial 508 boundaries everywhere in a fluid-bearing, deep-seated rock system. Although the presence of 509 faceted interfacial boundaries may decrease the degree of fluid redistribution, the

510 permeability in domains with finer grain sizes will match or exceed that in domains with 511 coarser grain sizes (Wark & Watson, 2000). However, this fluid localization induced by the 512 above two factors, to some extent increases the tortuosity of the fluid path on the whole rock 513 scale, which might hamper the enhancement of the bulk electrical conductivity and lead to a 514 deviation of the bulk electrical conductivity obtained in the present study from an idealized 515 tube model. As revealed by the CT images, a certain volume fraction of fluid is required to 516 establish the interconnected networks in the fluid-bearing forsterite aggregates. Furthermore, 517 even for the high fluid fraction systems, in which the fluid was well interconnected, our 518 conductivities were still smaller than those of the tube model. One plausible explanation for 519 this is the variation in tube diameter along the tubes as modeled by Watanabe and Peach 520 (2002), which is consistent with the bottleneck effect observed in the 3-D images of this 521 study.

Although the modified Archie's law is derived from the empirical Archie's law, it considers both the conductivities of solid and fluid phases and describes the change in the degree of fluid connectivity with increasing fluid fraction, which could be an appropriate mixing model for determining the fluid fraction dependence of our conductivities, as discussed in the Section 4.1. Therefore, we used the modified Archie's law to fit our data with a variable cementation exponent (m), and found that the m value of 1.9 best explains our data (Figure 6-a).

529 **5. Implication: Estimation of fluid fractions in fore-arc mantle wedges**

530 An increasing number of MT studies have found the presence of high electrical 531 conductivity anomalies (0.001–1 S/m) in the deep fore-arc regions of Cascadia (Jiracek et al., 532 1989; Soyer & Unsworth, 2006; Wannamaker et al., 2014), Mexico (Jödicke et al., 2006), 533 Costa Rica (Worzewski et al., 2011), North Chile (Araya Vargas et al., 2019; Brasse, 2002; 534 Schwalenberg et al., 2002), North Honshu (Mishina, 2009), Southern Kyushu (Ichiki et al., 535 2000), Ryukyu–Philippine (Shimakawa & Honkura, 1991), and New Zealand (Wannamaker 536 et al., 2002 and 2009). These high electrical conductivity anomalies are most likely due to the 537 presence of slab-derived aqueous fluids. In hot subduction zones such as Cascadia and 538 Mexico, fluids liberated from the shallow subducting slabs at depths of 30-60 km can 539 penetrate through the highly serpentinized mantle wedge and accumulate in the deep fore-arc 540 region (Nakatani & Nakamura, 2016; Reynard et al., 2011). In intermediate and cold 541 subduction zones, in contrast, saline fluid liberated at a depth of ~80 km can penetrate all the

way through the fore-arc mantle wedge without mantle melting and serpentinization due to low dihedral angles (Huang et al., 2019). Therefore, our experimental results can be applied to MT data of intermediate and cold subduction zones where the slab-derived saline fluids can be present in the non-serpentinized fore-arc mantle. Remarkably high electrical conductivity anomalies of 0.1–1 S/m were often observed in crustal regions where the dominant minerals are not olivine. We thus focus on the high electrical conductivity anomalies of 0.001–0.1 S/m detected at mantle depths.

549 Our experimental results provide a realistic relationship between the electrical 550 conductivity and fluid fractions in the forsterite-saline fluid system at 800 °C and 1 GPa, with 551 a fluid with a NaCl concentration of 5 wt.%, under conditions of textural equilibrium. In natural mantle settings, however, olivine contains iron, which enhances the electrical 552 553 conductivity of the olivine matrix through polaron conduction (Fei et al., 2020; Yoshino & 554 Katsura, 2013). To incorporate the effect of iron into the conductivity-fluid fraction 555 relationship, we recalculated the bulk conductivity by using the conductivity of hydrous ironbearing olivine aggregate $(3.0 \times 10^{-3} \text{ S/m}, \text{ Wang et al., 2006})$ according to the modified 556 Archie's law with m = 1.9 and the tube model (Figure 6-b). The results showed that the 557 558 difference in the bulk conductivity is negligible between the iron-free and iron-bearing 559 systems when the fluid forms an interconnected network in the models (i.e., the modified 560 Archie's law at fluid fractions above ~1 vol.% and the tube model). At fluid fractions below 1 vol.%, the bulk conductivity of the modified Archie's law is slightly higher in the iron-561 562 bearing system than in the iron-free system due to the contribution of the matrix conductivity.

In Figure 6-b, the range of electrical conductivity anomalies (0.001–0.1 S/m) detected 563 564 in fore-arc mantle wedges is shown. Based on the modified Archie's law calibrated by our 565 experimental data, the presence of ~1.0-5.0 vol.% aqueous fluid with 5.0 wt.% NaCl is 566 required to cause the high electrical conductivity anomalies of 0.01-0.1 S/m in fore-arc 567 mantle wedges. This result does not depend on the iron concentration of olivine, and is 568 almost consistent with a fluid fraction of ~ 2.0 vol.% for most mantle wedges (Iwamori, 569 1998). The anomalies of 0.003–0.01 S/m may represent less interconnected fluid pores with 570 fluid fractions below 1.0 vol.% in the iron-bearing olivine matrix. The lower conductivities of 571 0.001–0.003 S/m may be attributed to iron-bearing olivine with a water content smaller than 572 100–200 ppm under fluid-unsaturated conditions. The conventional tube model would lead to lower fluid fractions of ~0-0.8 vol.% for the anomalies of 0.003-0.1 S/m. The large 573

574 difference in the estimated fluid fractions between our model (i.e., $\sim 1.0-5.0$ vol.%) and the 575 conventional tube model (i.e., $\sim 0-0.8$ vol.%) emphasizes the importance of our experimental 576 data, which describe the realistic fluid distribution with interfacial anisotropy at mantle 577 conditions.

578 The effects of temperature, pressure, and fluid compositions on the estimated 579 electrical conductivity should be considered. Salinity significantly affects the conductivity 580 because salt acts as an effective charge carrier in the form of dissociated ions (Guo & Keppler, 2019) and also increases the solubility of olivine, which increases the solute 581 582 concentration in the fluids and affects the fluid geometry by changing the dihedral angle 583 (Huang et al, 2019). The original salinity of the slab-derived fluid produced by dehydration 584 reactions has been estimated to be 0.5-2.0 wt.% (Li & Hermann, 2015), but it can be 585 enhanced during the fluid ascent in the fore-arc mantle wedge owing the preferential 586 incorporation of water into the mantle minerals (Bernini et al., 2013; Reynard, 2016; Reynard 587 et al., 2011). The salinity of 5.0 wt.% used in this study is plausible because comparable 588 values were reported in upwelling slab-derived fluids (6.6 wt.%, Kusuda et al., 2014; 589 Matsubaya et al., 1973) and fluid inclusions in a mantle xenolith (4.1–6.1 wt.%, Kawamoto et 590 al., 2013). In addition, it does not deviate much from the salinity of seawater (3.5 wt.%), 591 which is likely representative of pore fluids expelled by mechanical compaction in the 592 shallow part of subduction zones. Carbon dioxide in the fluid may decrease the olivine-fluid 593 dihedral angles at low-T and high-P conditions corresponding to fore-arc mantle wedges 594 (Huang et al., 2020). The higher P-T conditions expected at greater depths enhance the 595 conductivity of the fluid (Guo & Keppler, 2019) and the olivine solubility (Yoshino et al., 596 2007). The presence of pyroxenes may also increase the solubility of olivine (Tiraboschi et 597 al., 2018) and decrease the olivine-fluid dihedral angle (Huang et al., 2020). These factors 598 more or less increase the bulk conductivity to some extent, thereby lowering the estimated 599 fluid fractions, although further experimental constraints are needed. Therefore, our data 600 could provide the upper bound on the estimated fluid fraction in the fore-arc mantle. 601 Nevertheless, the presence of significant amounts of aqueous fluid in the mantle suggests 602 intense fluid circulation through the fore-arc mantle wedge in subduction zones.

603 **6. Conclusion**

In this study, we measured the electrical conductivity of fluid-bearing forsterite aggregates with various fluid fractions and a constant salinity of 5.0 wt.% NaCl in the fluid at 1 GPa and 800 °C. Textural equilibrium was achieved owing to the long run durations of up to 8 days applied in this study. This was verified by the concentrated grain size distribution with a peak close to the mean grain size and the attainment of balance of interfacial tension at triple junctions. Our results enable us to establish a more realistic relationship between fluid fraction and bulk electrical conductivity in a non-ideal system, and to precisely estimate the fluid fraction in mantle wedges.

612 We found that the electrical conductivity increases nonlinearly with increasing fluid fractions, and that the modified Archie's law reproduces the experimental data very well. The 613 614 3-D microstructure of the interstitial pores determined by synchrotron X-ray CT shows a 615 change in fluid distribution from isolated pockets to interconnected networks with increasing 616 fluid fraction, accounting for the nonlinear increase in the electrical conductivity. Although 617 the dihedral angle in the olivine-fluid system with a salinity of 5 wt.% is smaller than the 618 critical value of 60° at 1 GPa and 800 °C (Huang et al, 2019), the interfacial anisotropy of 619 forsterite and the difference in grain size might localize the fluid distribution and further 620 hamper the bulk electrical conductivity. These factors result in a deviation of the electrical 621 conductivity obtained in our study from the idealized tube model. Moreover, a rapid increase 622 in bulk conductivity indicates that a threshold fluid fraction exists between 0.51 and 2.14 623 vol.%, beyond which interconnected fluid networks form, which is consistent with the 624 visualized 3-D fluid distributions in the recovered samples. Our results provide direct 625 evidence that the presence of > 1.0 vol.% aqueous fluid with moderate salinity is required to 626 explain the high conductivity anomalies above 0.01 S/m detected in fore-arc mantle wedges.

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- 640 electrical conductivity measurement, 3-D CT image, grain size distribution, and FTIR data, to
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