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# Electrical conductivity of omphacite and garnet indicates limited deep water recycling by crust subduction

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3 **Electrical conductivity of omphacite and garnet indicates limited**  
4 **deep water recycling by crust subduction**

5

6 Hanyong Liu<sup>1</sup>, Kai Zhang<sup>1</sup>, Jannick Ingrin<sup>2</sup>, Xiaozhi Yang<sup>1\*</sup>

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10 <sup>1</sup> State Key Laboratory for Mineral Deposits Research, School of Earth Sciences and

11 Engineering, Nanjing University, Nanjing 210023, PR China

12 <sup>2</sup> Univ.Lille, CNRS, INRAE, ENSCL, UMR 8207, UMET, Unité Matériaux et

13 Transformations, F 59000 Lille, France

14

15

16

17 \* Corresponding Author ([xzyang@nju.edu.cn](mailto:xzyang@nju.edu.cn))

18

19 **Abstract**

20 It is usually stated that oceanic crust recycling at subduction zones introduces large  
21 quantities of water into Earth's interior. However, it remains clouded how much water  
22 is recycled to the deep mantle. This is largely due to the challenge in directly sampling,  
23 and analyzing the water content of, a deep subducting crust. The subducting crust below  
24 ~30 km depth is dominated by omphacite and garnet in eclogite-facies rocks. Here we  
25 have determined the electrical conductivity of omphacite and garnet in representative  
26 subduction-related eclogites, each with varying contents of Fe and H<sub>2</sub>O that are key in  
27 electrical conduction. Considering the measured conductivity, the eclogite chemistry by  
28 geochemical investigations and the highly resistive property of subducting crusts by  
29 geophysical surveys, we demonstrate that, at 70-120 km depths in the subducting crust,  
30 the H<sub>2</sub>O contents of omphacite and garnet are strikingly small, with the maximum value  
31 being <400 and <80 ppm in the former and latter, respectively. The very small water  
32 contents indicate extremely water-poor conditions, or very low water activity, during  
33 the eclogite-facies metamorphism and in the system. This further implies the absence  
34 of appreciated amounts of hydrous phases such as amphibole and chlorite in the matrix,  
35 because of the strong ability of omphacite and garnet in hosting water as documented  
36 in natural samples. We suggest that the recycling of water to the deep mantle by oceanic  
37 crust subduction is limited. The results are important for modeling the conductivity of  
38 subducting slabs and understanding the deep water circulation.

39

40 **1. Introduction**

41 The transport of water from the hydrosphere to the mantle plays a critical role in  
42 affecting mantle dynamics, magma genesis and Earth's habitability. Water is recycled  
43 back to the mantle by slab subduction. At very shallow depths (e.g., <20 km), the input

44 of water into the mantle is primarily controlled by pore water (Peacock, 1990; Cai et  
45 al., 2018). In a downgoing slab at greater depths, however, water chiefly occurs as OH  
46 groups bound in the structure of the constitutive minerals, both hydrous and nominally  
47 anhydrous, note that H<sub>2</sub> dissolution in minerals is significant only at very high pressure  
48 and reducing conditions (Yang et al., 2016). As a slab descends, water is gradually lost  
49 by the breakdown of hydrous minerals such as amphibole and chlorite at depths mostly  
50 <100 km, due to their instabilities at high temperature (Peacock, 1990; Liu et al., 1996;  
51 Frost, 2006; Schmidt and Poli, 2014). Driven by its low density, the released water rises,  
52 metasomatizes wallrocks and causes melting in the wedge. This causes the development  
53 and widespread distribution of subduction zone volcanism and ore deposits.

54 It is widely believed that the crust layer in the slab is important for water recycling,  
55 because most of the slab water is initially stored there (Ito et al., 1983; Peacock, 1990;  
56 Dixon et al., 2002; Van Keken et al., 2002; Schmidt and Poli, 2014). Beyond ~30 km  
57 depth, the slab crust is dominated by eclogites (via phase change), consisting mainly of  
58 nominally anhydrous omphacite and garnet that are the main water carriers in eclogites  
59 after hydrous minerals decompose (Lu and Keppeler, 1997; Bromiley and Keppeler, 2004;  
60 Schmidt and Poli, 2014). As such, the water contents of omphacite and garnet are key  
61 to understanding water recycling to the deep mantle. Omphacite and garnet in terrain  
62 eclogites, that were once subducted to mantle depths and then returned to surface, could  
63 hold up to several thousands of ppm H<sub>2</sub>O as structural OH, leading to the suggestion  
64 that the subducting crust is able to carry large quantities of water into the deep mantle  
65 beyond the stability of the hydrous phases (Katayama and Nakashima, 2003; Katayama  
66 et al., 2006; Sheng et al., 2007). The high water contents of omphacite and garnet in the  
67 eclogites may, however, have suffered from complex secondary fluid-rock interactions  
68 postdating eclogite peak-metamorphism (e.g., Sheng et al., 2007; Schmädicke and Gose,

69 2017). It is thus obscure how much water is present in omphacite and garnet in the deep  
70 subducting crust.

71 The electrical conductivity of nominally anhydrous minerals (NAMs) is sensitive  
72 to water (Karato, 1990; Dai and Karato, 2009; Yoshino et al., 2009; Yang et al., 2011,  
73 2012; Yang and McCammon, 2012; Zhang et al., 2012, 2019; Zhao and Yoshino, 2016;  
74 Liu et al., 2019). Meanwhile, electromagnetic surveys offer a direct window into Earth's  
75 interior. In this context, constraints can be provided on the water contents of omphacite  
76 and garnet in the subducting crust (and water recycling), if the conductivity structure of  
77 subducting crusts is resolved and the electrical property of subduction-related materials  
78 is determined. In a recent work, Liu et al. (2019) have quantified the effect of water on  
79 the conductivity of omphacite and garnet, and provided a preliminary constraint on the  
80 likely water content in the subducting crust. However, Liu et al. have not evaluated the  
81 effect of Fe, which along with H are crucial in electrical conduction, and the question  
82 arises how convincing their results might be applied to Earth's interior. In this report,  
83 we have by experimental work measured the conductivity of omphacite and garnet from  
84 representative eclogites, which plus the samples of Liu et al. (2019) show a wide range  
85 of contents in Fe and H<sub>2</sub>O. The data are used to infer the water contents of the minerals  
86 in the subducting crust, by combining with geophysically-based electrical structure and  
87 slab petrology. The results demonstrate clearly that a small amount of water is present  
88 in omphacite and garnet in deep subducting crusts.

## 89 **2. Experiments and Methods**

### 90 **2.1 Starting materials and sample characterization**

91 Three starting eclogites were from Bixiling (omp1 + grt1) and Shuanghe (omp3 +  
92 grt3) in China and Weißenstein (omp2 + grt2) in Germany, and were all related to slab  
93 subduction and exhumation (Franz et al., 1986; Sheng et al., 2007). The Weißenstein

94 eclogite was studied in Liu et al. (2019). These samples were chosen by considering Fe  
95 contents of constituting omphacite and garnet and quality of grains from >30 eclogites  
96 (*note*: the electrical property of silicates is independent of the origin). Natural eclogites  
97 were not used for conductivity studies directly, because of the occurrence of accessory  
98 materials (e.g., secondary hydrous phases, fluid inclusions and/or other impurities) and  
99 zoned water distribution that affect conductivity analyses. We analyzed the conductivity  
100 of omphacite and garnet separately and then modeled the bulk conductivity. Omphacite  
101 and garnet are representative in composition, in particular in Fe that almost covers the  
102 content ranges in subduction-involved eclogites (omp1 and omp3: Appendix).  
103 Optically clear omphacite/garnet grains, ~100-700  $\mu\text{m}$  in size, were handpicked under  
104 a binocular microscope. Each sample is chemically homogeneous in major-/minor-  
105 elements (Table 1), as measured by a JEOL JXA-8230 electron microprobe (15 kV  
106 accelerating voltage, 10 nA current and <5  $\mu\text{m}$  beam size). Grains were ground to  
107 powder, mostly 30-80  $\mu\text{m}$ , and H-annealed with minor distilled water in Ni capsules  
108 (ID 4.2 mm, OD 5.0 mm and length 10 mm) at 1-3.5 GPa and 650-700  $^{\circ}\text{C}$  (150-180 h  
109 duration) in a piston cylinder press. By this, water homogenization was achieved in  
110 each sample, and samples with different water contents were prepared. Dry samples  
111 were not prepared, because OH is common in natural omphacite and garnet. Recovered  
112 samples were cored into cylinders of 3 mm diameter and 1.7 mm length for subsequent  
113 conductivity runs, note that the conductivity of silicate minerals by lattice conduction  
114 is independent of grain size above ~5  $\mu\text{m}$  that is typical for minerals in the deep Earth  
115 (Yang and Heidelbach, 2012).

116 A precise analysis of water content is key for evaluating its effect on conductivity.  
117 The general principle for water quantification in silicate minerals by Fourier-transform  
118 infrared (FTIR) spectroscopy is given by the modified Beer-Lambert law:  $C_{\text{H}} = Ab_{\text{S}_{\text{total}}}/I$ ,

119 where  $C_H$  is OH content,  $Abs_{total}$  is the total OH integrated absorbance normalized to 1  
120 cm thickness, and  $I$  is the mineral-specific integral molar absorption coefficient. As  
121 such,  $C_H$  is determined by  $Abs_{total}$ , once the coefficient  $I$  is externally calibrated. For  
122 optically anisotropic minerals, the intensity of OH bands is sensitive to the orientation  
123 of the IR active dipole relative to incident beam, and  $Abs_{total}$  is the sum of thickness-  
124 normalized integrated absorbance along the three principal axes ( $a$ ,  $b$  and  $c$ ). This  
125 requires orienting samples and polarized FTIR analyses along the axes, which are  
126 difficult for small grains and are time-consuming for sample preparation. Recently,  
127 Shuai and Yang (2017) have shown that  $Abs_{total}$  can be well characterized by polarized  
128 FTIR analyses along any three mutually perpendicular directions of a given sample,  
129 without the necessity to orient the samples; in case very small grains which are unable  
130 to be prepared for polarized FTIR work, Qiu et al. (2018) have demonstrated that  $Abs_{total}$   
131 can be obtained by unpolarized FTIR analyses on randomly oriented grains.

132 Water contents were measured by a Bruker Vertex 70V FTIR spectrometer coupled  
133 with a Hyperion 2000 microscope. Analyses were performed on optically clear grains  
134 with an aperture of  $60 \times 60 \mu\text{m}$  and a resolution of  $4 \text{ cm}^{-1}$  (128 scans for each spectrum  
135 by a globar source, KBr-Ge beam splitter, MCT detector and wire-grid Ze-Se polarizer).  
136 For anisotropic omphacite, polarized spectra were acquired for relatively large grains  
137 in annealed samples before conductivity runs (Shuai and Yang, 2017), and unpolarized  
138 spectra on 15-20 randomly oriented grains were obtained for fine-grained samples after  
139 conductivity runs (Qiu et al., 2018). For cubic garnet, unpolarized spectra were recorded  
140 over 6-9 grains. Water contents were calibrated by the mineral-specific coefficients of  
141 Katayama et al. (2006) for omphacite and garnet in eclogites, and uncertainty is mostly  
142  $<10\%$ , as documented in Shuai and Yang (2017) and Qiu et al. (2018). For a comparison  
143 under the same framework, the same calibration was used to recalculate the  $\text{H}_2\text{O}$  content

144 of minerals in natural eclogites in available studies. This yields values different from  
145 some early work, e.g., the 1840 ppm OH of omphacite in Smyth et al. (1991), estimated  
146 by the OH peak linear intensity, is actually ~310 ppm H<sub>2</sub>O; in contrast, the H<sub>2</sub>O contents  
147 of omphacite and garnet in recent reports (Katayama and Nakashima, 2003; Katayama  
148 et al., 2006; Sheng et al., 2007; Schmädicke and Gose, 2017) are less affected. The  
149 general results of this work are not affected by adopting the calibration coefficients.

## 150 **2.2 Conductivity experiments at elevated conditions**

151 Slabs are rather oxidizing above ~200 km depth, with redox state similar to the Ni-  
152 NiO buffer (McCammon, 2005). Therefore, conductivity runs were carried out with Ni-  
153 NiO at 1-2.5 GPa and 250-850 °C in an end-loaded piston cylinder press. The assembly  
154 design resembles that in our previous studies (Yang et al., 2011, 2012; Yang, 2012; Yang  
155 and Heidelbach, 2012; Yang and McCammon, 2012; Li et al., 2016, 2017; Liu et al.,  
156 2019), and only a brief description is offered. In each run, a BN-Ni double capsule was  
157 adopted to maintain sample geometry and yield a relatively sealed chamber. BN causes  
158 reducing conditions only when O<sub>2</sub> is rich (e.g., in air) and run duration is long above  
159 ~1000 °C, by  $4\text{BN} + 3\text{O}_2 = 2\text{B}_2\text{O}_3 + 2\text{N}_2$ , and BN itself in a sealed system does not  
160 influence the redox state. It has been demonstrated that, for the design at <1000 °C and  
161 short duration (typical of conductivity runs), the reaction of Pt and Fe in samples is  
162 negligible and the redox state in the chamber is well buffered, as tested with pyroxenes  
163 and olivine (Yang et al., 2011, 2012; Yang, 2012; Yang and McCammon, 2012).

164 Before assembly, Al<sub>2</sub>O<sub>3</sub> parts were heated at 1000 °C to remove absorbed water,  
165 and during assembly, no cement/glue was used to immobilize the various parts to avoid  
166 volatile release at high temperature that affects the analyses. Completed assembly was  
167 heated at 136 °C overnight, and was heated again at 200 °C after loading into the press.  
168 Impedance spectra were recorded in the heating-cooling cycles, with a Solartron 1260



169 Impedance/Gain Phase analyser by frequency sweeping at  $10^6$ -1 Hz and 0.5 V applied  
170 voltage. Temperature of the analyses was up to 850 °C, to minimize water diffusion loss  
171 of samples upon heating. Duration was typically ~1 min or less per analysis (depending  
172 on temperature). After each run, recovered capsule was polished for optical, FTIR and  
173 backscattered electron examinations. Ni-NiO pairs were present, and sample distortions  
174 and grain growths were negligible, as observed in similar work (Yang et al., 2011, 2012;  
175 Yang, 2012; Yang and Heidelbach, 2012; Yang and McCammon, 2012; Li et al., 2017;  
176 Liu et al., 2019). Resistance ( $R$ ) was obtained from the impedance spectra (see below),  
177 and conductivity ( $\sigma$ ) was calculated by  $\sigma = L/SR$ , where  $L$  and  $S$  are the effective sample  
178 length and cross-section area, respectively. Uncertainty is usually <5% for conductivity  
179 and <20 °C for temperature.

### 180 **3. Results**

181 Representative FTIR spectra of annealed samples are shown in Fig. 1. The spectra  
182 show absorption peaks at 3610-3625, 3500-3520 and 3440-3460  $\text{cm}^{-1}$  in the omphacites  
183 and at 3610-3640 and 3530-3550  $\text{cm}^{-1}$  in the garnets, which are typical of OH bands in  
184 the eclogite minerals (Smyth et al., 1991; Katayama and Nakashima, 2003; Katayama  
185 et al., 2006; Sheng et al., 2007; Schmädicke and Gose, 2017). In each mineral, the peak  
186 frequency positions are similar, suggesting similar H incorporation mechanism, and the  
187 relative difference of peak intensity between different samples are due to different water  
188 contents and/or grain orientations. Sample  $\text{H}_2\text{O}$  contents are basically the same prior to  
189 and after each conductivity run (Table 2). Fe and other major-/minor-elements are more  
190 sluggish than H, and their contents are unchanged in the runs. Representative complex  
191 spectra are presented in Fig. 2. The spectra usually show a high frequency arc and a low  
192 frequency tail, although the tail is sometimes not obvious, and the spectral shapes are  
193 similar to those reported for other minerals (Dai and Karato, 2009; Yoshino et al., 2009;

194 Yang et al., 2011, 2012; Yang, 2012; Zhang et al., 2012, 2019; Zhao and Yoshino, 2016;  
195 Li et al., 2016, 2017; Liu et al., 2019). The arc and tail was by sample lattice conduction  
196 and electrode effects, respectively.

197 The conductivity of NAMs (and other Earth materials) is quantitatively described  
198 by the Arrhenius relation:

$$\sigma = \sigma_0 \cdot \exp(-\Delta H / RT) \quad (1)$$

199 where  $\sigma_0$  is a constant,  $\Delta H$  is the activation enthalpy,  $R$  is the ideal gas constant, and  $T$   
200 is temperature. Conductivities in the first heating were sometimes affected by residual  
201 moisture in the chamber, yielding data not obeying Eq. (1) as observed in similar studies  
202 (Yang et al., 2012; Yang and Heidelbach, 2012; Li et al., 2017; Liu et al., 2019), and the  
203 influenced data were excluded. Conductivity data and the fittings to Eq. (1) are shown  
204 in Fig. 3, and fit parameters are given in Table 2. Sample conductivity is much greater  
205 than the assembly background conductivity (Yang et al., 2011). The data are consistent  
206 between different heating-cooling cycles in each run (see also Appendix), suggesting  
207 the absence of system hysteresis. The conductivity is slightly larger at 2.5 than at 1 GPa,  
208 indicated by the runs with an OH-bearing omphacite (omp3). This is consistent with  
209 the previous study on an OH-bearing pyrope that shows similar conductivity  
210 enhancement by pressure (Dai and Karato, 2009). In contrast, the conductivity of dry  
211 olivine or garnet decreases slightly with increasing pressure (Xu et al., 2000; Dai and  
212 Karato, 2009). The difference is caused by the different pressure dependence between  
213 small polaron and H conduction in the minerals (see Section 4), but the effect of  
214 pressure (at a small variation) on the conductivity of silicate minerals is in general  
215 insignificant.

#### 216 **4. Fe- and H-dependent conductivity**

217 In Fe- and H-bearing NAMs, the conductivity is related to the conduction of small

218 polarons (electron-hole hopping between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ) and protons (H). Our data show  
219 that, at otherwise identical conditions, the conductivity of both omphacite and garnet  
220 increases with Fe and  $\text{H}_2\text{O}$  contents (Fig. 3), agreeing with studies on other NAMs (Dai  
221 and Karato, 2009; Yoshino et al., 2009; Yang et al., 2011, 2012; Yang and McCammon,  
222 2012; Zhang et al., 2012; Zhao and Yoshino, 2016). The data trends of our omphacites  
223 are consistent with those of synthetic omphacites at 3 GPa (Zhang et al., 2019), and the  
224 conductivity of the omphacite (omp3) with 7.92 wt.% FeO and 70 ppm  $\text{H}_2\text{O}$  is close to  
225 that of an augite with 8.31 wt.% FeO and 75 ppm  $\text{H}_2\text{O}$  at 1 GPa (Yang and McCammon,  
226 2012) (Fig. 3a). The conductivity of the omphacite (omp1) with 1.46 wt.% and 200 ppm  
227  $\text{H}_2\text{O}$  resembles that of the one (omp2) with 4.27 wt.% and 150-290 ppm  $\text{H}_2\text{O}$ , implying  
228 an insignificant effect of Fe at that content range. At a similar 155-160 ppm  $\text{H}_2\text{O}$ , the  
229 conductivity of the Fe-rich garnet (15.40 wt.% FeO) is less than that reported by Dai  
230 and Karato (2009) for a Fe-poor pyrope (7.42 wt.% FeO) at 8 GPa (Fig. 3b). Probably,  
231 this is related to the different pressures (1 vs. 8 GPa) and H-species (note that different  
232 H-defects could have different mobility, contributing differently to the conduction), e.g.,  
233 narrow OH bands at  $3700\text{-}3500\text{ cm}^{-1}$  in this study vs. broad bands at  $3800\text{-}3000\text{ cm}^{-1}$  in  
234 Dai and Karato (2009). The conductivity of our garnets is close at similar contents of  
235 FeO (26.55-26.58 wt.%) and  $\text{H}_2\text{O}$  (100-110 ppm, with similar OH band shapes, Fig. 1).  
236 The activation enthalpy is Fe-dependent in omphacite,  $\sim 87$ , 81-84 and 65 kJ/mol at 1.64,  
237 4.27 and 7.92 wt.% FeO, respectively, but is broadly Fe-independent in garnet, 88-91  
238 kJ/mol over a large range of Fe contents, 15.40-26.58 wt.% FeO (Table 1). Therefore,  
239 the effect of Fe on the conductivity of omphacite and garnet is mineral dependent.

240 For the same omphacite or garnet in eclogite samples under similar conditions, the  
241 conductivity is linearly proportional to water content below  $\sim 1000\text{ }^\circ\text{C}$  and at moderate  
242  $\text{H}_2\text{O}$  contents (Liu et al., 2019; Zhang et al., 2019). This has also been reported for many

243 other NAMs such as augite, diopside, enstatite, plagioclase, pyrope and olivine (Yang  
 244 et al., 2011, 2012; Yang and McCammon, 2012; Zhang et al., 2012; Zhao and Yoshino,  
 245 2016). For an OH-bearing mineral with a fixed content of Fe (and other major elements),  
 246 the relative contribution of small polaron conduction is usually much smaller than that  
 247 of proton conduction at relatively low temperature. Eq. (1) can be approximated by:

$$\sigma \propto C_H \cdot \exp(-\Delta H_H / RT) \quad (2)$$

248 where  $\Delta H_H$  is the activation enthalpy by H conduction and is numerically equal to  $\Delta H$   
 249 in Eq. (1). As such, the conductivity of both omphacite and garnet can be modeled for  
 250 a range of H<sub>2</sub>O contents at each of the starting FeO contents (Fig. 4). The essentially  
 251 same conductivity of omphacite with 1.64 and 4.27 wt.% FeO at the same H<sub>2</sub>O content,  
 252 as noted above, indicates a more significant role of H than Fe at such Fe content levels.  
 253 The N-type semiconductor relation is used to model the effect of both Fe and H on the  
 254 conductivity of both minerals:

$$\sigma = A \cdot X_{Fe}^n \cdot C_H \cdot \exp[-(\Delta E - \alpha X_{Fe}^{1/3} - \beta C_H^{1/3}) / RT] \quad (3)$$

255 where  $X_{Fe}$  is the molar fraction of Fe (= Fe/(Fe + Mg)),  $\Delta E$  is the activation energy, and  
 256  $A$ ,  $n$ ,  $\alpha$  and  $\beta$  are constants. The insensitivity of omphacite conductivity to FeO content  
 257 at 1.64-4.27 wt.% makes it hard to model the whole dataset with Eq. (3). In this context,  
 258 the modeling is carried out for omphacite using the conductivity data of our samples  
 259 starting with omp1 and omp3, that restrict the boundaries of Fe contents in subduction-  
 260 related omphacites, and the synthetic omphacite (5.81 wt.% FeO and 420 ppm H<sub>2</sub>O  
 261 with similar OH band positions) of Zhang et al. (2019), and for garnet using the data of  
 262 all our samples (the Dai and Karato (2009) pyropes are not used because of the different  
 263 OH bands). This yields  $\Delta E$ ,  $A$ ,  $n$ ,  $\alpha$  and  $\beta$  of  $106 \pm 6$  kJ/mol,  $2754 \pm 380$  S/m,  $-0.05 \pm 0.09$ ,  
 264  $64 \pm 6$  kJ/mol and  $-26 \pm 8$  kJ/mol for omphacite, and  $89 \pm 3$  kJ/mol,  $61660 \pm 4085$  S/m,  
 265  $4.25 \pm 1.32$ ,  $-4 \pm 31$  kJ/mol and  $15 \pm 9$  kJ/mol for garnet. For both minerals, the modeled

266 results by Eq. (2) and (3) are well consistent at the same H<sub>2</sub>O and boundary Fe contents,  
267 especially at >500 °C (Fig. 4).

268 The bulk conductivity of eclogite is calculated from omphacite and garnet with the  
269 Hashin and Shtrikman (1962) bounds, which offer the narrowest restrictions of a two-  
270 phase composite despite the geometrical configurations:

$$\begin{aligned}\sigma_{HS+} &= \sigma_1 + x_2[(\sigma_2 - \sigma_1)^{-1} + x_1 / (3\sigma_1)]^{-1} \\ \sigma_{HS-} &= \sigma_2 + x_1[(\sigma_1 - \sigma_2)^{-1} + x_2 / (3\sigma_2)]^{-1}\end{aligned}\quad (4)$$

271 where  $\sigma_{HS+}$  and  $\sigma_{HS-}$  are the upper and lower bounds, respectively, given  $\sigma_1 > \sigma_2$  ( $x$  is the  
272 volume proportion). A key prerequisite for the modeling is the equilibrium distribution  
273 of major/minor elements and OH between coexisting minerals. This is a factor that must  
274 be considered when modeling the bulk conductivity of any matrix consisting of several  
275 phases. The calculation is carried out by Eq. (2) using the conductivity data of minerals  
276 at different H<sub>2</sub>O contents for each starting Fe content, in which case the effect of Fe is  
277 constrained at the same time (Fig. 4). Eq. (3) is not adopted, because of the difficulty in  
278 controlling chemical equilibrium between the two minerals. Each of the mineral pairs,  
279 omp1-grt1, omp2-grt2 and omp3-grt3, was separated from the same starting eclogite,  
280 and the major/minor elements are already in equilibrium. The water partition coefficient  
281 between omphacite and garnet in eclogite, ~0.2 from Katayama et al. (2006), is adopted  
282 to obtain the equilibrium H<sub>2</sub>O contents (assuming 100, 200 and 400 ppm in omphacite).  
283 The bulk conductivity of eclogite is not affected by likely minor impurities, because if  
284 present, they are usually isolated (not connected) in the matrix and do not contribute to  
285 bulk conduction. The results are shown in Fig. 5a and b for omphacite-rich and garnet-  
286 rich rocks, respectively. This produces the modeled bulk conductivity at variable Fe and  
287 H<sub>2</sub>O contents and high temperature. In particular, The FeO contents reflect the range of  
288 the two minerals in subducting crusts (as noted before). Given other conditions,  $\sigma_{HS+}$

289 and  $\sigma_{\text{HS}}$  of each sample show a small variation above 600 °C, and are up to ~0.1 S/m  
290 at 900 °C (for the modeled composition). Considering the positive pressure effect on  
291 the conductivity of OH-bearing omphacite and garnet as mentioned above, the bulk  
292 conductivity is only slightly greater at higher pressure.

## 293 **5. Water contents in subducting omphacite and garnet**

294 Electromagnetic induction is sensitive to conductive rather than resistive bodies in  
295 Earth's interior. This makes it hard to accurately resolve the conductivity of subducting  
296 crusts that are highly resistive. Usually, high conductivity is not detected in subducting  
297 slabs, and slab conductivity is much smaller than that of mantle wedge and surrounding  
298 mantle in the inversion results. Conductivity of the subducting crust, either fast or slow  
299 subduction in diverse settings, is estimated to be mostly  $10^{-4}$  to  $10^{-3}$  S/m at ~70-120 km  
300 depth (Vanyan et al., 2002; McGary et al., 2014; Ichiki et al., 2015; Vargas et al., 2019).  
301 This is a reasonable estimate by imposing a 1000, 5000 or 10000  $\Omega$  slab resistivity in  
302 the inversion models (Ichiki et al., 2015). A typical electrical structure of a subducting  
303 crust is given in Fig. 5c (after McGary et al. (2014) who focused on conductive channels  
304 in the wedge). We want to note that, although Evans et al. (2014) have offered a variety  
305 of outputs (mainly in the wedge) with the same electromagnetic data of McGary et al.  
306 (2014) by applying different inversion methods, the crust portion at ~70-120 km depth  
307 is very resistive in most of their models. Moreover, Fig. 5c is for the Cascadia area, but  
308 broadly similar results on resistive slab vs. depth have been reported for other tectonic  
309 environments (e.g., Vanyan et al., 2002; Ichiki et al., 2015; Vargas et al., 2019).

310 It has been shown that, despite a wide range of ages, geometries and convergence  
311 rates, slabs actually share many common features with their thermal structures (Peacock,  
312 1996; Syracuse et al., 2010; Penniston-Dorland et al., 2015). At ~70-120 km depth, the  
313 temperature is mostly ~600-900 °C in the crust layer, for subduction zones over various

314 tectonic settings. The FeO content is ~1-8 wt.% of omphacite and 14-27 wt.% of garnet  
315 in terrane eclogites that were once brought to ~200 km depth (Katayama and Nakashima,  
316 2003; Katayama et al., 2006; Sheng et al., 2007; and Appendix), suggesting that deeply  
317 subducting eclogites are in fact not depleted in Fe. As such, to cause the  $10^{-4}$ - $10^{-3}$  S/m  
318 conductivity of subducting crusts at ~600-900 °C, the only possibility is that omphacite  
319 and garnet are water-poor. According to the modeling in Fig. 5a-b, the H<sub>2</sub>O contents are  
320 <400 ppm in omphacite and <80 ppm in garnet at 600 °C and <100 ppm in omphacite  
321 and <20 ppm in garnet at 900 °C for the bottom Fe boundaries (omp1-grt1), and <30  
322 ppm for the top Fe boundaries (omp3-grt3). These H<sub>2</sub>O-content ranges are meaningful,  
323 in particular if considering the weaker effect of a minor variation in Fe than in H content  
324 (Section 4), and can be even smaller, if the effect of pressure on the conductivity of OH-  
325 bearing omphacite and garnet is considered (Section 3). The maximum <400 ppm H<sub>2</sub>O  
326 content resembles that estimated in Liu et al. (2019), which is based on omp2 in contrast  
327 to omp1 in this study (due to the more significant effect of H at the Fe content levels as  
328 noted above). The inferred H<sub>2</sub>O contents of omphacite and garnet in subducting crusts  
329 (~70-120 km depth) are much less than those up to 2000-3000 ppm in terrane eclogites  
330 (Katayama and Nakashima, 2003; Katayama et al., 2006). This suggests that OH-rich  
331 omphacite and garnet in the terrane samples were H-enriched by secondary processes  
332 during their exhumation to surface, in line with the arguments by analyzing mineral OH  
333 patterns and D/H ratios (Sheng et al., 2007; Schmädicke and Gose, 2017). Interestingly,  
334 the H<sub>2</sub>O values resemble the mostly <360 ppm of omphacite and <65 ppm of garnet in  
335 volcano-hosted xenolith eclogites (Smyth et al., 1991; Bell and Rossman, 1992; Koch-  
336 Müller et al., 2004). This makes more sense if xenolith eclogites represent the various  
337 dehydrated products of subducted crusts that were accidentally entrained by volcanoes  
338 and transferred to the surface; however, things could be complicated because a general

339 consensus has not been reached for the origin of xenolith eclogites, e.g., some may form  
340 by melt accumulation (Jacob, 2004).

341 Consequently, the conductivity data and inferred water contents of omphacite and  
342 garnet are broadly consistent with the constraints from other approaches. The inversion  
343 of electromagnetic data at subduction zones has long been subjected to the difficulty in  
344 adopting a good reference resistor in the starting model. In most cases, the inversion is  
345 carried out by highlighting the conductivity contrast between the resistive slab and the  
346 conductive paths in the wedge (linked to melting and/or dehydrated fluids). In order to  
347 identify fine-scale variation of conductivity in the subducting slab, e.g., due to change  
348 of temperature and release of water with subduction, it is important to impose a reliable  
349 slab conductivity in the inversion model. The water contents of minerals in the slab, the  
350 electrical properties of eclogites and the conductivity laws, as provided here, allow to  
351 model the conductivity of oceanic crusts as a function of temperature and water content  
352 as the starting reference resistor for the inversion of electromagnetic data.

## 353 **6. Implications for deep water recycling**

354 The very low water contents of omphacite and garnet at ~70-120 km depth imply  
355 a much smaller water content in the subducting crust than the original value dominated  
356 by hydrous phases (e.g., amphibole and chlorite), e.g., ~1-2 wt.% H<sub>2</sub>O (Peacock, 1990).  
357 Water in the subducting crust is mostly released at shallow depths, and the dehydration  
358 of hydrous phases and omphacite and garnet in eclogites produces conductive channels  
359 in the wedge (McGary et al., 2014). Eclogite-facies metamorphism, which involves all  
360 materials in the system, occurs at >30 km depths in the subducting crust (e.g., Peacock,  
361 1990; Schmidt and Poli, 2014). The inferred small water contents of omphacite and  
362 garnet are indicative of extremely water-poor conditions, or very low water activity,  
363 during the metamorphism and in the system. Otherwise, the water contents of the two



364 minerals would dramatically increase, due to their strong ability in accommodating OH  
365 (Katayama and Nakashima, 2003; Katayama et al., 2006; Sheng et al., 2007), and their  
366 conductivity would be greatly enhanced, leading to conductive zones in the subducting  
367 slab that is inconsistent with geophysical mappings (Vanyan et al., 2002; McGary et al.,  
368 2014; Ichiki et al., 2015; Vargas et al., 2019). The water-poor conditions also imply  
369 very minor to negligible amounts of hydrous phases in the system, because of their  
370 extensive dehydration and water escape at shallow depths. This agrees with the  
371 arguments that subducting crusts may contain no hydrous minerals beyond ~3 GPa and  
372 700 °C, as advocated by considering dehydration in natural rocks, numerical modeling  
373 and phase stabilities (Liu et al., 1996; Schmidt and Poli, 2014).

374 In this context, the amount of water carried to the deep mantle by subducting crust  
375 is regulated by omphacite and garnet in eclogites, but not hydrous phases. This implies  
376 a minor amount of water that could be brought to the deep mantle, although it might be  
377 significant at very shallow depths (Peacock, 1990; Cai et al., 2018). Assuming the total  
378 length, thickness and density of subducting oceanic crusts of 40000 km (Syracuse et al.,  
379 2010), 6 km and 3 g/cm<sup>3</sup>, respectively, and the average rate of subduction of 5 cm/year,  
380 the mass of water transferred to the mantle exceeding ~70 km depth in the past 3 billion  
381 years would correspond to <3% of the oceans. A direct implication is that the exchange  
382 of water between Earth's exterior and interior reservoirs might not be efficient. This fits  
383 well with the high dehydration efficiency (>92%) of subducting crusts at shallow depths,  
384 estimated from the H<sub>2</sub>O/Ce ratios of mid-ocean ridge basalts (Dixon et al., 2002), and  
385 the preservation of protosolar nebular water in the deep mantle over Earth's evolution,  
386 constrained by the D/H ratios of plume-related lavas (Hallis et al., 2015). The limited  
387 water transfer by crust deep subduction does not mean a water-poor deep Earth, because  
388 surface water (and atmosphere) formed by degassing of the Earth (Allègre et al., 1987)

389 and its deep interior was already hydrous.

390       Consequently, the water flux into the deep mantle is much smaller than that simply  
391 estimated from the stabilities of hydrous minerals such as amphibole, chlorite and dense  
392 hydrous magnesium silicates, by assuming their continuous presence in appreciated  
393 amounts in the downgoing crust. The stability fields of hydrous phases are established  
394 by experiments at water saturated/super-saturated conditions, e.g., with up to >10 wt.%  
395 water in sealed capsules (Liu et al., 1996; Frost, 2006; Schmidt and Poli, 2014). This  
396 requirement is hard to always fulfil in the subducting crust that is not a closed system  
397 and contains an initial H<sub>2</sub>O content of ~1-2 wt.% prior to subduction (Peacock, 1990),  
398 which is mostly released at shallow depths. Thus, there is a gap between the water-poor  
399 state in the deep slab and the water-rich state in laboratory. Finally, if slab mantle bears  
400 serpentine, e.g., by the serpentinization of peridotites providing there is enough water  
401 for serpentine formation prior to subduction, then its role in conveying water to Earth's  
402 interior should be evaluated. Key to this issue is to map the actual amount of serpentine  
403 in the slab mantle and the dynamics of the low density with subduction, e.g., 2.6 g/cm<sup>3</sup>  
404 of serpentine vs. 3.4 g/cm<sup>3</sup> of peridotite and 3.6 g/cm<sup>3</sup> of eclogite. This is beyond the  
405 scope of this work, and more studies are required.

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526 447, 1-9.

527 **Figure captions**

528 **Fig. 1** Representative FTIR spectra of annealed omphacite and garnet. (a) polarized  
529 spectra of omphacite along three orthogonal directions of each grain ( $X'$ ,  $Y'$  and  $Z'$ ), and  
530 (b) unpolarized spectra of garnet from different grains. The omp2 and grt2 spectra were  
531 reproduced from Liu et al. (2019). Spectra were normalized to 1 cm thickness and  
532 vertically offset.

533 **Fig. 2** Representative complex spectra of (a) omphacite (B260) and (b) garnet (B256).  
534  $Z'/Z''$  is the real/imaginary part of complex impedance, and frequency decreases along  
535 the  $Z'$  axis from left (1 MHz) to right (1 Hz). An equivalent circuit of a single  $R$ -CPE,  
536 a resistor ( $R$ ) and a constant phase element (CPE) in parallel, was used to fit the high-  
537 frequency arc. Tail and data scatter at low frequency were due to electrode effects that  
538 did not affect sample lattice resistance measurements. Number close to each spectrum  
539 is temperature ( $^{\circ}\text{C}$ ), and inset in (a) shows spectra at 600-700  $^{\circ}\text{C}$ .

540 **Fig. 3** Measured conductivity of (a) omphacite and (b) garnet at 1-2.5 GPa and 250-  
541 850  $^{\circ}\text{C}$ . The run at 2 GPa was marked, and lines are linear fittings to measured data of  
542 each sample (with  $\text{H}_2\text{O}$  contents labeled) by Eq. (1). Symbols around each line are from  
543 the same sample (different symbols for different cycles). Samples omp2 and grt2 were  
544 from Liu et al. (2019). Uncertainty is usually smaller than/comparable to symbol sizes.  
545 Data sources: assembly background, Yang et al. (2011); Z19, synthetic omphacites in  
546 Zhang et al. (2019) at 3 GPa (0% FeO and 280 ppm  $\text{H}_2\text{O}$  and 5.81% FeO and 420 ppm  
547  $\text{H}_2\text{O}$ , with  $\text{H}_2\text{O}$  contents recalculated (see text)); Y12, augite in Yang and McCammon  
548 (2012) at 1 GPa (8.31% FeO and 75 ppm  $\text{H}_2\text{O}$ ); D09, pyrope in Dai and Karato (2009)  
549 at 8 GPa (7.42% FeO and 160 ppm  $\text{H}_2\text{O}$ , with  $\text{H}_2\text{O}$  content not recalculated owing to  
550 the different OH bands).

551 **Fig. 4** Modeled conductivity of (a) omphacite and (b) garnet at various FeO and  $\text{H}_2\text{O}$

552 contents. Colored solid lines were made using Eq. (2), and dashed and dotted grey lines  
553 were plotted using Eq. (3) for the bottom and top boundaries of FeO content (omp1 and  
554 omp3: see text), respectively. H<sub>2</sub>O contents of 100, 200 and 400 ppm in omphacite and  
555 20, 40 and 80 ppm in garnet at equilibrium OH partitioning were assumed (see text).

556 **Fig. 5** Electrical conductivity of eclogites with different modal composition and Fe  
557 and H<sub>2</sub>O contents. (a) omp-rich (65 vol.% omp + 35 vol.% grt) eclogite, (b) grt-rich (35  
558 vol.% omp + 65 vol.% grt) eclogite, and (c) a typical conductivity-temperature structure  
559 along subduction zones (modified after McGary et al. (2014): note that some reference  
560 resistivities were imposed in their starting inversion model: see text). Modeling in (a,  
561 b) was conducted with the mineral data in Fig. 4 and Eq. (4): blue, red and green colors  
562 highlight the different FeO contents, omp1 + grt1, omp2 + grt2 and omp3 + grt3 (with  
563 labels on the right side), and star symbols denote different OH contents in the minerals.  
564 Horizontal/vertical shadows in (a, b) show the conductivity/temperature range of the  
565 subducting crust at ~70-120 km depth in (c).

566 Table 1 Composition of the starting omphacite and garnet (by wt.%)

	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	Total
omp1	56.84	0.03	9.59	1.64	0.02	10.57	15.39	5.72	0.01	99.81
	<i>0.42</i>	<i>0.02</i>	<i>0.07</i>	<i>0.03</i>	<i>0.01</i>	<i>0.30</i>	<i>0.09</i>	<i>0.03</i>	<i>0.01</i>	
omp2*	55.64	0.07	11.38	4.27	0.02	8.44	12.98	7.31	<0.01	100.11
	<i>0.16</i>	<i>0.03</i>	<i>0.35</i>	<i>0.28</i>	<i>0.02</i>	<i>0.17</i>	<i>0.40</i>	<i>0.24</i>	<i>&lt;0.01</i>	
omp3	55.37	0.05	7.87	7.92	0.04	8.39	13.54	6.64	<0.01	99.82
	<i>0.26</i>	<i>0.01</i>	<i>0.08</i>	<i>0.15</i>	<i>0.04</i>	<i>0.03</i>	<i>0.08</i>	<i>0.16</i>	<i>&lt;0.01</i>	
grt1	39.48	0.05	22.22	15.40	0.29	9.33	12.24	<0.01	<0.01	99.01
	<i>0.15</i>	<i>0.03</i>	<i>0.07</i>	<i>0.17</i>	<i>0.01</i>	<i>0.18</i>	<i>0.19</i>	<i>&lt;0.01</i>	<i>&lt;0.01</i>	
grt2*	37.64	0.03	22.73	26.55	0.74	6.81	5.63	0.03	<0.01	100.16
	<i>0.20</i>	<i>0.02</i>	<i>0.12</i>	<i>0.36</i>	<i>0.08</i>	<i>0.17</i>	<i>0.32</i>	<i>0.02</i>	<i>&lt;0.01</i>	
grt3	37.49	0.07	21.63	26.58	0.14	5.02	8.79	0.04	<0.01	99.76
	<i>0.15</i>	<i>0.04</i>	<i>0.10</i>	<i>0.07</i>	<i>0.04</i>	<i>0.10</i>	<i>0.03</i>	<i>0.02</i>	<i>&lt;0.01</i>	

567 Assuming all Fe as FeO. Data are the average and standard deviation (italic) by electron

568 microprobe analyses. omp, omphacite; grt, garnet.

569 \*: data were reproduced from Liu et al. (2019) (see text).



570 Table 2 Summary of samples and fit parameters

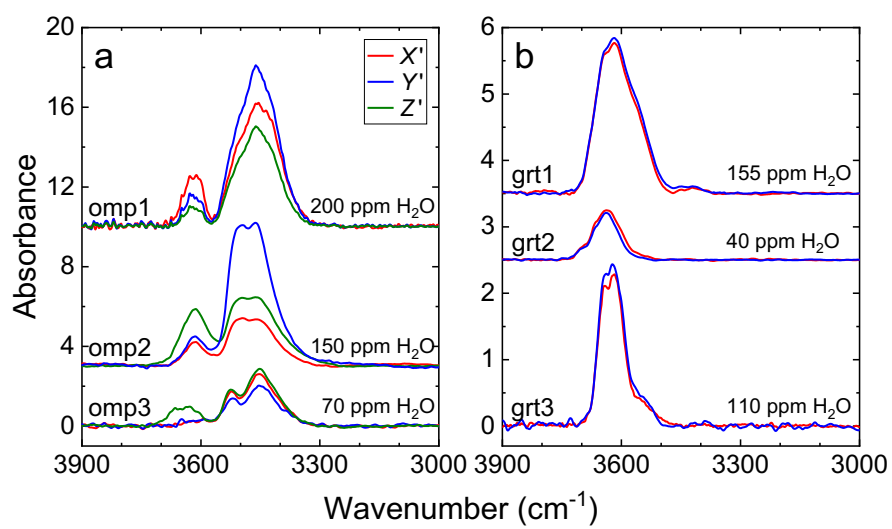
Sample	FeO (wt.%)	$P$ (GPa)	$T$ (°C)	ppm H <sub>2</sub> O (initial)	ppm H <sub>2</sub> O (final)	$L/S$ (m <sup>-1</sup> )	$\log_{10}(A_0(\text{S/m}))$	$\Delta H$ (kJ/mol)
B255 (omp1)	1.64	1	350-700	200	190	288	1.97±0.01	87±2
B185 (omp2)*	4.27	1	350-600	85	80	290	1.38±0.24	84±4
B149 (omp2)*	4.27	1	350-600	150	160	216	1.52±0.14	82±2
B145 (omp2)*	4.27	1	350-650	290	300	269	1.75±0.12	81±2
B260 (omp3)	7.92	1	250-700	70	70	280	1.21±0.07	64±1
B261 (omp3)	7.92	2.5	250-700	70	65	285	1.45±0.04	65±1
B256 (grt1)	15.40	1	350-800	155	150	340	1.89±0.18	93±3
B167 (grt2)*	26.55	1	350-800	40	45	226	1.80±0.17	90±3
B174 (grt2)*	26.55	1	350-800	100	95	245	2.26±0.12	91±2
B267 (grt3)	26.58	1	350-850	110	110	246	2.15±0.13	88±2

571 Water contents are the values prior to (initial) and after (final) conductivity runs, and are rounded to the nearest 5 ppm.  $L/S$  is the ratio of  
572 sample length to cross-section area (by considering the effective contact between electrodes and sample). FeO contents of the samples are  
573 shown (from Table 1).

574 \*: samples were used in Liu et al. (2019) for documenting the water content exponent of 1 in Eq. (2).

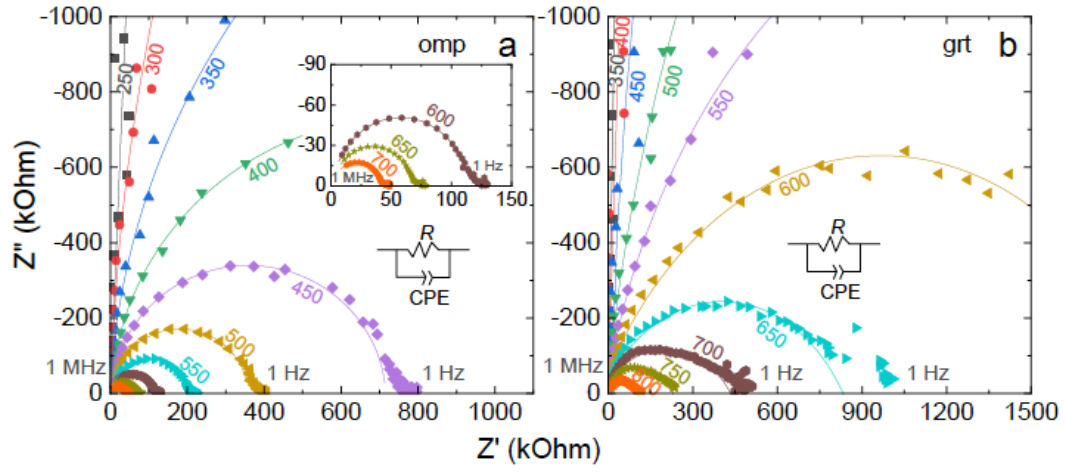
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Fig. 1



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Fig. 2



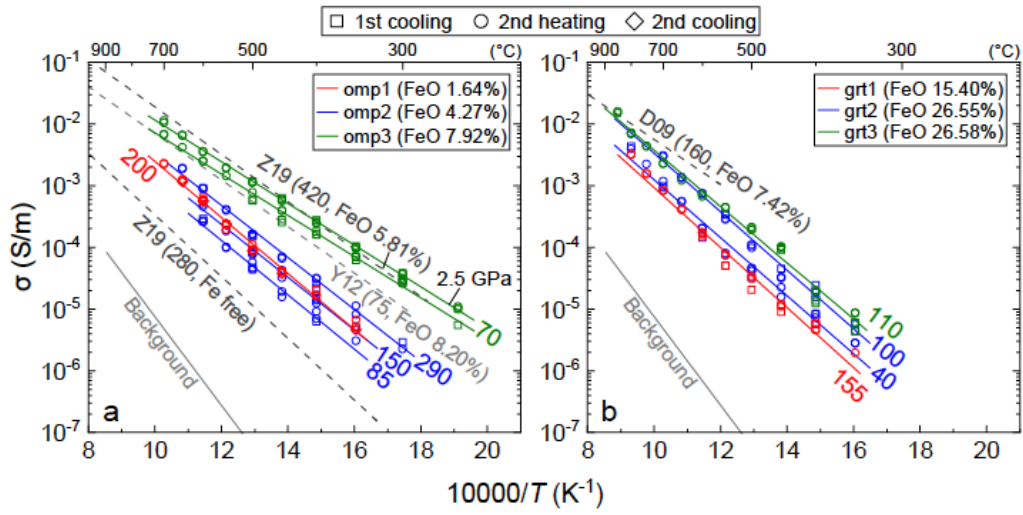
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Fig. 3

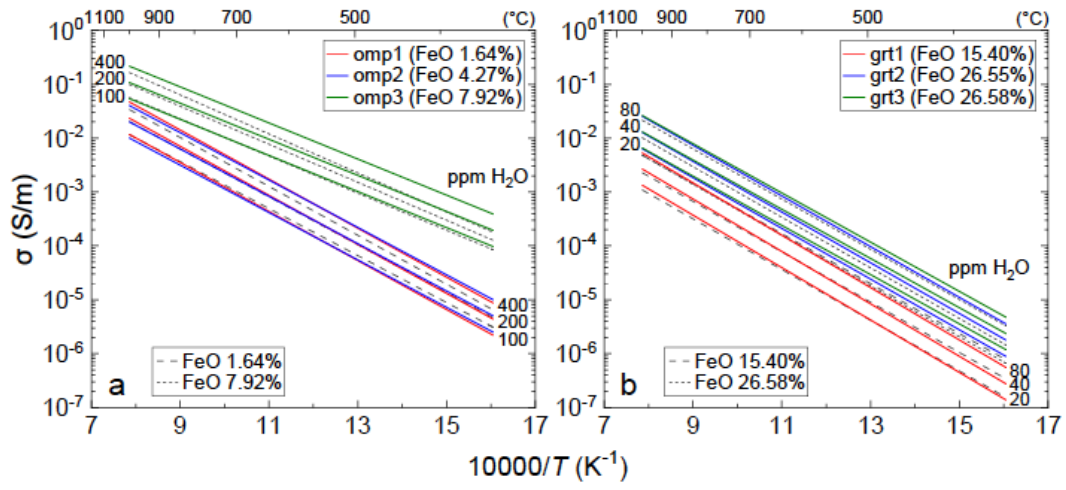


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Fig. 4



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Fig. 5

