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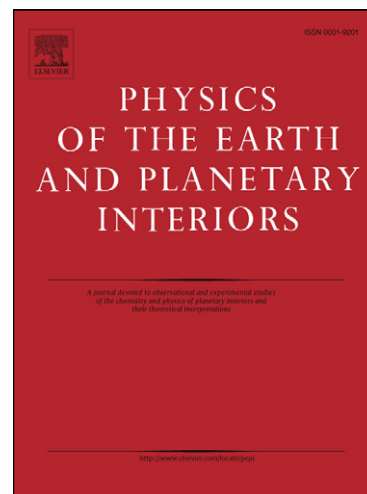
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1 **Electrical conductivity of partial molten carbonate peridotite**

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11 **Abstract** In order to investigate the effect of carbonate-content in partial melt on bulk
12 conductivity under high pressure, electrical conductivity measurements were performed
13 on carbonate melt-bearing peridotites using a Kawai-type multi-anvil apparatus. The
14 starting materials were composed of spinel lherzolite (KLB1) with small amounts of
15 dolomite (1 and 3 wt.%). To obtain various melt fractions, annealing experiments were
16 performed at different temperatures above 1400 K at 3 GPa. At low temperatures (\leq
17 1500 K), the conductivity was distinctly higher than that of carbonate-free peridotite
18 and close to that of the carbonatite melt-bearing olivine aggregates. Although the
19 sample conductivity increased with increasing temperature, the rate at which the
20 conductivity increases was small and the conductivity approached that of silicate melt-
21 bearing peridotite. CO₂ concentration in the partial melt decreased with increasing
22 annealing temperature. Thus, the small increase of the conductivity with annealing
23 temperature is attributed to a decrease of the melt conductivity due to a decrease in

24 carbonate content in the partial melt. As the carbonate concentration in the melt
25 decreases, the estimated melt conductivity approaches that of the basaltic melt.
26 Therefore, conductivity enhancement by the carbonate-bearing melt is very effective at
27 temperature just above that of the carbonate peridotite solidus.

28 *Keywords: Carbonatite; Electrical conductivity; Melt fraction; Peridotite; Upper*
29 *mantle*

30 **1. Introduction**

31 The high conductivity anomaly in the upper mantle is a general feature
32 beneath the oceanic lithosphere as observed by electromagnetic surveys (e.g., Lizarralde
33 et al., 1995; Evans et al., 2005; Baba et al., 2006). There are two major candidates for
34 what causes the raising of the conductivity in that region: partial melt and hydration of
35 nominally anhydrous minerals (e.g., Tyburczy and Waff, 1983; Karato, 1990). However,
36 the electrical conductivity of hydrous olivine is not high enough to explain the
37 conductivity anomaly at the top of the asthenosphere (Yoshino et al., 2006; 2009a; Poe
38 et al., 2009). Recently Gaillard et al. (2008) proposed that a very small amount of
39 carbonatite is an attractive agent for generating the conductivity anomaly based on the
40 conductivity measurement of carbonate melts at atmospheric pressure. Later research
41 showed the conductivity measurement of carbonate melt-bearing olivine aggregates at 3
42 GPa had an order of magnitude higher conductivity than the silicate melt-bearing
43 olivine aggregates for the same melt fraction (Yoshino et al., 2010). In addition, carbon
44 in peridotite can significantly reduce the solidus temperature (e.g., Falloon and Green,
45 1989; Dasgupta and Hirschmann, 2006). Thus the presence or absence of carbonatite
46 melt will affect the estimation of mantle temperature from the conductivity structure of
47 the upper mantle obtained from geophysical surveys.

48 Yoshino et al. (2010) reported on the effect of carbonatitic partial melt with
49 higher carbon concentration on the bulk conductivity. However, partial melts of
50 carbonate-bearing mantle peridotites may have variable carbonate concentrations under
51 a wide range of mantle conditions. Thus, to assess conductivity anomalies in the upper
52 mantle, we need to know the effect of the carbonate component in partial melt on the
53 bulk conductivity of the partially molten peridotite under high pressure. In this study,
54 we determined the electrical conductivities of carbonate-bearing peridotite with variable
55 melt fractions. A variation of melt fractions was obtained by annealing under different
56 temperature conditions. Since we used starting materials with a fixed carbonate
57 concentration, the variation in the degree of partial melting can provide the variation in
58 carbonate concentration in the partial melt. In addition, we measured conductivity of
59 dolomite melt in order to constrain the absolute conductivity and activation enthalpy of
60 carbonate melt at high pressure. The electrical conductivity of carbonate-bearing melt
61 was estimated as a function of CO₂ concentration in the melt. We obtained CO₂
62 concentration dependence of electrical conductivity by comparing the data obtained
63 from our previous results, in which the conductivity was obtained for the partial molten
64 carbonate peridotite with fixed CO₂ concentration in melt. Such an argument provides
65 the constraints on the presence of carbonate partial melt and on the thermal structure of
66 the upper mantle.

67 **2. Experimental Methods**

68 Starting materials were powder mixtures of natural spinel lherzolite (KLB1)
69 with 1 and 3 wt. % natural dolomite from Austria, which have approximately 0.5 and
70 1.5 wt.% bulk CO₂, respectively. For one experiment, a powder of the KLB1 without
71 dolomite was used as a starting material to investigate the effect of carbonate on
72 electrical conductivity. Dolomite powder was also used as a starting material to measure
73 conductivity of dolomite melt. The particle size of the powder was less than a few

74 micrometers. The powdered sample was encapsulated in a cylindrical MgO sleeve and
75 was sandwiched by two graphite electrodes in contact with two sets of $W_{97}Re_3$ - $W_{75}Re_{25}$
76 thermocouples. Two sets of thermocouples were also used for the four-pole resistance
77 method of electrical conductivity measurement. The design of the cell assembly is the
78 same as that given in Yoshino et al. (2010).

79 Conductivity measurement with impedance spectroscopy was carried out using a
80 Solartron 1260 impedance Gain-Phase Analyzer combined with a Solartron 1296
81 interface. Complex impedances were obtained at frequencies ranging from 0.1 Hz to 1
82 MHz and applied voltages of 1 or 1.41 V. We applied the pseudo 4-pole electrode
83 system to the conductivity measurement, because the resistance of the electrode and
84 long metallic wires (thermocouple) can be comparable to that of the highly conductive
85 melt (Pommier et al., 2010).

86 Conductivity measurements were conducted through several heating-cooling
87 cycles at 3 GPa in a Kawai-type multianvil press. The temperature was increased and
88 decreased in steps of 25-50 K to the desired temperature. Impedance spectra were
89 obtained at each temperature step. Firstly the sample was heated and held at 1000 K to
90 dehydrate the sample and the surrounding materials. Then the sample was heated to the
91 desired temperature, which is above the solidus of carbonate-bearing peridotite (~1350
92 K at 3 GPa; Dasgupta and Hirschmann, 2006). To achieve textural equilibrium of the
93 solid-liquid composites, the samples were annealed at the desired temperature, by
94 continuous monitoring of electrical conductivity, until the sample conductivity became
95 constant. After annealing, the sample was cooled to less than 1000 K. Subsequent
96 heating cycles using step-wise temperature increments were also conducted to confirm
97 reversibility. In order to retain the partial molten texture, the sample was quenched from
98 the highest temperature to ambient temperature.

99 Retrieved samples were mounted in epoxy and ground parallel to the axis of
100 the cylindrical heater. The chemical compositions of the carbonate and silicate phases in
101 the recovered sample were obtained using electron microprobe analyzer (EPMA). The
102 carbonate (CO₂) concentration in the melt was predicted from the total weight deficit of
103 the EPMA data (Table 2). However, the near-solidus partial melt composition could not
104 be determined because of its small size (< 1 μm). The microstructure on the polished
105 section of the run products was observed by secondary electron and back-scattered
106 electron images (BEI) using a field-emission scanning electron microscope (FE-SEM).
107 The melt fraction of the samples was determined from the image analysis after the
108 experiments (for detail, see Yoshino et al. (2005)).

109 **3. Results**

110 3.1. Texture and chemical composition

111 The melt fraction of carbonate-bearing peridotite increased with increasing the
112 annealing temperature (Table 1), whereas the carbonate-free peridotite annealed at 1700
113 K showed the absence of a melt phase. Microstructures on the polished section are
114 shown in Fig. 1. Carbonate-bearing melt was located at the triple junction of the olivine
115 and pyroxene crystals (Fig. 1a-c). As the melt fraction increased, the melt completely
116 surrounded the olivine crystals on the polished section (Fig. 1d). The apparent dihedral
117 angle was around 20°, which implies a three-dimensional interconnection of the melt
118 network. Infiltration of the melt into the MgO capsule was not observed for all run
119 products. Partial melt was homogeneously distributed in the center of the sample,
120 whereas the melt-less olivine layer was also developed adjacent to the MgO container
121 with a thickness of less than 100 μm. Forsterite content (Fo₉₆) in the olivine adjacent to
122 the MgO container was relatively higher because of iron loss from the sample to the
123 MgO capsule. The samples showed relatively homogeneous distribution of the melt, and
124 therefore we believe the measured conductivity values should represent the whole part

125 of the partially molten zone. As mentioned above, the outer part of the sample had less
126 iron contents. The sample conductivity of the uncontaminated part of the sample should
127 be slightly higher than the obtained value (less than 10 %).

128 For the 3 wt.% dolomite-bearing peridotite system, the dolomite completely
129 decomposed below 1600 K. Orthopyroxene disappeared because of a decarbonation
130 reaction such as MgSiO_3 (in orthopyroxene) + MgCO_3 (in dolomite) = Mg_2SiO_4 (in
131 olivine) + CO_2 . Clinopyroxene was present up to 1700 K, although by volume the
132 proportion of clinopyroxene decreased with increasing temperature. The carbonate
133 concentration (CO_2) in the partial melt of the samples decreased from 45 to 13 wt.%
134 with increasing melt fraction from 0.02 to 0.2 (Tables 1 and 2). In the case of the KLB1
135 + 1 wt.% dolomite system, the CO_2 concentration in melt decreased to 5 wt.% when the
136 melt fraction was 0.20. We were not able to measure the melt composition accurately
137 for samples containing the lowest melt fraction (≤ 1 vol.%) using the electron
138 microprobe. However, the carbonate component of the sample is usually expected to be
139 40 wt.% just above the solidus (Dasgupta et al., 2007). Thus the CO_2 concentration in
140 the partial melt largely decreased with increasing annealing temperature and melt
141 fraction. The CO_2 concentrations in the melt estimated by EPMA were slightly higher
142 than those expected from the bulk composition. For example, when we assume that all
143 carbons partition into the melt phase, and the densities of the carbonate and basaltic
144 melt range from 2 to 2.7 g/cm^3 (e.g., Dobson et al., 1996; Rigden et al., 1986), the
145 KLB1 + dolomite 3 wt.% system with 20 vol.% melting requires around 10 wt.% CO_2
146 concentration in the partial melt. The CO_2 concentrations in the melt were increased by
147 partial solidification of the melt due to reaction with the MgO capsule.

148 3.2. Electrical conductivity

149 The impedance spectra generally show one arc at higher frequencies and an
150 additional part appearing at lower frequencies (Fig. 2). If the melt phase is

151 interconnected in a solid matrix, it forms an electrical pathway in parallel to the solid
152 matrix (Roberts and Tyburczy, 2000). Thus the high-frequency arc reflects the sample
153 properties, and the low frequency tail is accordingly interpreted as an effect of the
154 electrodes. Therefore, only the first arc was used to determine the conductivity of the
155 sample. As shown in Fig. 2b, the impedance spectra were sometimes observed to
156 contain a predominantly inductive reactance (inductive loop) possibly due to the mass
157 transport and electrochemical reaction at the electrode interface (Hampton et al., 1980;
158 van Hassel et al., 1991). However, errors in conductivity values created by the induction
159 component are small (less than 10 %), and therefore we reserve a detailed argument on
160 the inductive component.

161 Fig. 3 shows the conductivity of dolomite up to 1800 K in Arrhenian plot. In the
162 first heating, the conductivity largely increased by nearly two orders of magnitude at
163 1300 K and then increased by one order of magnitude at 1500 K. Above 1600 K, an
164 increase of conductivity with temperature became small. After annealing at 1800 K, the
165 conductivity slightly decreased with decreasing temperature. The high conductivity
166 values (10 S/m) were retained even after cooling below the melting temperature. Above
167 1500 K the good linear relation in Arrhenius plot was observed. Conductivity-
168 temperature relationships were determined from Arrhenian fits to the data for each
169 sample

$$170 \quad \sigma = \sigma_0 \exp\left(-\frac{H}{kT}\right) \quad (1)$$

171 where k is the Boltzmann constant and T is temperature in K. Activation enthalpies (H)
172 and pre-exponential terms (σ_0) resulting from fitting Eq. (1) to the data are listed in
173 Table 1. The calculated activation enthalpy for electrical conduction in dolomite melt is
174 38 kJ/mol, which is slightly higher than the values (30–35 kJ/mol) reported from
175 Gaillard et al. (2008). The pre-exponential factor (σ_0) for electrical conduction in

176 dolomite melt is 1343 S/m, which is lower than the values (3440 S/m) reported from
177 Gaillard et al. (2008). Therefore, the absolute conductivity value of the dolomite melt
178 was slightly lower than those of alkali carbonate melt [$\text{KCa}_{0.5}\text{CO}_3$]₂,
179 ($\text{NaKCa}_{0.5}\text{CO}_3$)₃, (NaKCaCO_3)₂, (NaKCO_3)₂, and (LiNaKCO_3)₃] reported by
180 Gaillard et al. (2008).

181 An example of the conductivity measurements for the partial molten system is
182 shown in Fig. 4a. The conductivity values were initially high and rapidly decreased
183 during annealing at 1000 K because of the dehydration of surrounding materials. In the
184 second heating, the conductivity largely increased by nearly two orders of magnitude up
185 to 1100 K. This temperature is consistent with the temperature at which the conductivity
186 of Ca-rich carbonate abruptly increases during heating (e.g. Gaillard et al., 2008).
187 Above 1200 K, the conductivity slightly increased with increasing temperature up to
188 1500 K. At the beginning of annealing at 1500 K, the sample conductivity rapidly
189 decreased and then became constant within 2 hours, similar to our previous studies
190 (Yoshino et al., 2010). At the beginning of partial melting, the powder with large
191 porosity was instantaneously filled with melt. Therefore, the conductivity was initially
192 high. As the system established textural equilibrium during annealing, some pores were
193 closed to minimize the total interfacial energy in the system. This process led to the
194 reduction of the bulk conductivity of the sample. The impedance spectra showed part of
195 a semicircular shape at higher frequencies, suggesting that the conductive phase forms
196 an electrical pathway parallel to the solid matrix, and an additional part derived from
197 electrode reaction at the interface between the sample and electrode at lower
198 frequencies (Fig. 2). In the cooling path, the electrical conductivity decreased with
199 decreasing temperature. The temperature-conductivity path in the Arrhenius plot
200 showed linear trends with some different slopes, suggesting the conduction mechanism
201 changes in a certain temperature range. In the subsequent heating path, the
202 reproducibility was confirmed.

203 In the third heating, the conductivity values at the highest temperature were
204 identical to those just after the annealing for a few hours at the same temperature in the
205 second heating (Fig. 4a). The samples were quenched from the highest temperature to
206 obtain the partial molten texture of the final state. The conductivity values shown in
207 Table 1 were obtained just before quenching. Therefore, the sample texture and the
208 conductivity values represent the same condition as the second heating. Fig. 5 shows a
209 plot of the conductivity values at the maximum temperature and melt fraction against
210 the highest temperature experienced for each sample. The electrical conductivity of the
211 partial molten sample generally increased with increasing temperature. The melt
212 fraction increased from 0.01 to 0.20 in the investigated temperature range from 1400 to
213 1700 K.

214 Fig. 4b shows the logarithmic plot of conductivity versus reciprocal temperature
215 for the KLB1 + dolomite 3 wt.% system in the second cooling path. The electrical
216 conductivity of the carbonate-bearing peridotite is distinctly higher than that of
217 carbonate- and melt-free peridotite, which is similar to that of olivine (Constable 2006;
218 Yoshino et al., 2006; 2009). The samples annealed at higher temperatures showed lower
219 conductivity values at the same temperature, and larger temperature dependence
220 compared with the sample annealed at lower temperatures. Although samples have the
221 same bulk composition, the fact that the cooling paths in the Arrhenius plot are largely
222 different suggests that the chemical composition of the melt phase did not change
223 significantly during cooling. In other words, chemical equilibrium in the system was
224 established at the highest temperature, but not held during cooling. The activation
225 enthalpy for electrical conductivity of carbonate melt is smaller than those of silicate
226 melts (e.g., Presnall et al., 1972; Gaillard et al., 2008). A small temperature dependence
227 on the conductivity of samples annealed at lower temperature was quite similar to that
228 of dolomite end member, suggesting that carbonate concentration in the melt was
229 relatively high. This trend is consistent with a variation of the carbonate concentration

230 in the partial melt determined by the weight deficiency of the microprobe analysis
231 (Table 2).

232 4. Discussion

233 4.1. Relationship between conductivity and melt fraction

234 Carbonate-bearing melts have high wetting properties (e.g., Hunter and
235 McKenzie, 1989; Minarik and Watson, 1995; Hammouda and Laporte, 2000). The
236 previous works which determined dihedral angle of olivine-basalt and olivine-
237 carbonatite systems have shown similar values (20~30°), which is much less than a
238 critical value (60°) for interconnection (Hunter and McKenzie, 1989; Waff and Bulau,
239 1979; Yoshino et al., 2009b). Partial melt should form interconnected liquid networks at
240 olivine grain boundaries even at very low-volume fractions and should therefore
241 contribute to the bulk rock conductivity. Thus, a large change of melt connectivity with
242 melt fraction is not expected in the present system. If there was no threshold for the
243 interconnection, a relationship between conductivity (σ_{bulk}) and melt fraction (ϕ) can be
244 expressed by mixing models such as Hashin-Shtrikman upper bound. The previous
245 studies of the partial molten system with the constant melt composition demonstrated a
246 linear relationship in the log σ - log ϕ plot, which is known as the Archie's law (Archie,
247 1942). Archie's relationship can be expressed as follows:

$$248 \quad \sigma_{\text{bulk}} = C\phi^n\sigma_m \quad (2)$$

249 where C and n are constants (e.g., Watanabe and Kurita, 1993). The power exponent in
250 Archie's relation should be close to unity for partial molten rocks with well-
251 interconnected melt geometry and the constant melt composition (Watanabe and Kurita,
252 1993; ten Grotenhuis et al., 2005; Yoshino et al., 2010). The exponent close to unity is

253 also valid for the other mixing models in the system containing the well-interconnected
254 phase.

255 In this study, the melt fraction in the sample changes with temperature under the
256 constant bulk CO₂ composition (Fig. 6). The conductivity of the bulk rock increases
257 with increasing melt fraction in this study, which is in the same tendency with that in
258 the previous study, where CO₂ concentration is constant not in the bulk rock but in the
259 melt (Yoshino et al., 2010). However, an increase of conductivity values with melt
260 fraction in this study is smaller than that in the previous study. The present system
261 yields much smaller exponent for Eq. (2), namely $n = 0.58$. In addition, the conductivity
262 values of dolomite melt, which is an end member of this system, does not agree with the
263 extrapolated trend (Fig. 6).

264 The Archie's law is only applicable to the system with constant melt
265 conductivity. Thus, the "apparent" low power exponent ($n = 0.58$) obtained in this study
266 contains information about variation in melt conductivity with melt fraction. There are
267 two factors to vary the melt conductivity. One is temperature and the other is the CO₂
268 concentration in the partial melt. First we consider thermal effect. The melt conductivity
269 generally increases with increasing temperature followed by Arrhenian relation. If we
270 consider the sample conductivity at 1700 K in order to directly compare the results of
271 our previous study (Yoshino et al., 2010), the power exponent would be much smaller
272 than 0.58, because the conductivity measured at lower temperature is relatively higher
273 than that determined at higher temperature. Such a low exponent value (< 0.58) further
274 increases the deviation from that (~ 1) expected from the partial molten rocks with a
275 constant melt composition. Therefore, temperature is not controlling factor.

276 Next we consider the effect of melt composition on the "apparent" low power
277 exponent. Gaillard et al. (2008) reported that carbonate melt has a distinctly higher
278 electrical conductivity than silicate melt. In the carbonate-bearing peridotite system, the

279 conductivity of the partial melt is expected to be controlled by the CO₂ content in the
280 partial melt. In general, carbonatitic partial melts containing ~40 wt.% CO₂ are stable
281 only in a limited temperature range above the solidus (Dalton and Presnall, 1998;
282 Gudfinnsson and Presnall, 2005; Dasgupta and Hirschmann, 2006; 2007; Dasgupta et
283 al., 2007). As the temperature approaches to the carbonate-free dry peridotite solidus
284 temperature, the reaction of carbonatite melts with silicate minerals in peridotite
285 produces carbonated silicate melts with a lower CO₂ concentration (Moore and Wood,
286 1998; Dalton and Presnall, 1998; Gudfinnsson and Presnall, 2005; Dasgupta et al.,
287 2007). For example, Dasgupta et al. (2007) showed that the transition from carbonatite
288 (40 wt.% CO₂) to carbonated silicate melt (≤ 25 wt.% CO₂) in carbonate-bearing
289 peridotite at 3 GPa occurs abruptly at 1623 K. This study also showed that the CO₂
290 content in the partial melt is even lower at higher temperatures (~13 wt.% CO₂ at 1700
291 K). Our measurement demonstrated that, at low melt fraction (~1 vol. %), the
292 conductivity values are similar to those of the olivine-carbonatite system (at 1650 K),
293 whereas at higher melt fraction (> 10 vol.%), the conductivity values are rather closer to
294 those of the olivine-basalt system measured at 1.5 GPa (Yoshino et al., 2010). Thus the
295 expected trend of the conductivity-melt fraction of the present system approaches to that
296 of the olivine-carbonatite (40 wt.% CO₂) and the olivine-basalt trends at lower (< 0.3
297 vol.%) and higher (> 10 vol.%) melt fractions, respectively. As a result the small n
298 value obtained in the investigated range of melt fraction can be attributed to the
299 decrease of conductivity with carbonate content in the melt by increasing the annealing
300 temperature.

301 4.2. Estimation of liquid conductivity as a function of carbonate content

302 We next estimate the electrical conductivity of the carbonate-bearing melt itself
303 as a function of the CO₂ concentration in the melt to compare it directly with melts with

304 various CO₂ concentrations at constant temperature (1700 K). To do this, we made the
305 following two simple assumptions:

306 1) Electrical conductivity of the partial melt is proportional to the melt fraction.
307 For example, when the bulk conductivity of the sample with 10 vol.% melt fraction is
308 10 S/m, the melt conductivity can be calculated as 100 S/m. This assumption can be
309 justified since the power exponent n in Eq. (2) for partially molten rocks has a value
310 close to unity (Watanabe and Kurita, 1993; Roberts and Tyburczy, 2000; ten Grotenhuis
311 et al., 2005; Yoshino et al., 2010). In addition, if a difference between solid and melt
312 conductivities exceeds 3 orders of magnitude, the exponent estimated from the Hashin-
313 Shtrikman upper bound, which is frequently used for the melt-bearing system, yields
314 approximately unity for a case that melt fraction is above 0.01.

315 2) The effect of temperature on the conductivity of the melt is controlled by the
316 activation enthalpy for electrical conductivity of the melts. The activation enthalpy of
317 dolomite melts was 38 kJ/mol at 3 GPa. On the other hand, the average activation
318 enthalpies of basaltic melt are more than three times higher (110-150 kJ/mol) (Tyburczy
319 and Waff, 1983; Gaillard and Marziano, 2005). Thus, the temperature dependence of
320 the conductivity of the carbonate melts is much smaller than that of silicate melts. Since
321 silicate melts are likely to polymerize as the SiO₂ component in the melt increases, its
322 activation enthalpy would increase with the increasing SiO₂ component in the carbonate
323 melt. In the present study, the CO₂ concentration in the melt varies considerably.
324 Therefore, the activation enthalpy is expected to gradually increase with decreasing CO₂
325 concentration in the melt, because there is a negative-correlation between the SiO₂ and
326 CO₂ concentrations in the carbonate melt (Table 2).

327 The calculation scheme was as follows. First of all, the absolute conductivity
328 values of melt at temperature we measured were calculated based on the first
329 assumption that conductivity is proportional to melt fraction. To compare the

330 conductivity for each carbonate melt at constant temperature, the Arrhenian relation (Eq.
331 1) was used. The pre-exponential factors (σ_0) for each carbonate melt were calculated
332 from the present data for two fixed activation enthalpies (38 and 150 kJ/mol), because
333 once the activation enthalpy is assumed, the unknown parameter in Eq. (1) is only σ_0 .
334 The calculated parameters are shown in Table 3. The σ_0 calculated using high activation
335 energy (150 kJ/mol) yields higher values. Indeed, the activation enthalpy should change
336 with the carbonate component in the melt. To estimate more realistic melt conductivity,
337 the activation enthalpy was assumed to be a linear relation between pure carbonate (38
338 kJ/mol) and pure silicate melts (150 kJ/mol). Further calculation scheme was the same
339 as those presented earlier. The used parameters are also shown in Table 3. The
340 conductivities at 1700 K were calculated from Eq. (1) using the σ_0 determined by this
341 study.

342 Fig. 7a shows the logarithmic plot of the electrical conductivity of the partial
343 melt in the KLB1 + dolomite 3 wt.% system as a function of the CO₂ concentration in
344 the melt at 1700 K. Although the log of electrical conductivity was calculated using a
345 possible range of activation enthalpies, the activation enthalpy for the melt conductivity
346 with higher CO₂ concentration (> 40 wt. %) should be low and close to that of dolomite
347 melt (38 kJ/mol), whereas that with lower CO₂ concentration should be higher and
348 approach that of silicate melt (110-150 kJ/mol). Considering the effect of the CO₂
349 concentration on activation enthalpy for the melt conductivity, the expected trend of the
350 electrical conductivity of the carbonate-bearing melt, which is in equilibrium with
351 peridotite, shows a large decrease of the electrical conductivity below 40 wt.% of CO₂
352 (yellow arrows in Fig. 7a). The extrapolation of the melt conductivity to zero CO₂
353 agrees well with the basaltic melt conductivity (TW83: Tyburczy and Waff, 1983;
354 KAB: Pommier et al., 2010). A variation of the estimated melt conductivity as a
355 function of CO₂ concentration shows a plateau between 33 and 20 wt.% CO₂. If there
356 exists a threshold for interconnection of chains of silicate tetrahedral in melt, an abrupt

357 change of melt conductivity and its activation enthalpy might be present. On the other
358 hand, the conductivity of the melt with higher CO₂ concentration is quite consistent
359 with that of dolomite melt measured at 3 GPa, which is lower than those of the alkali
360 carbonate melt determined at atmospheric pressure (Gaillard et al., 2008). One
361 explanation for this discrepancy between dolomite and alkali carbonate melt is a
362 negative pressure dependence on the melt conductivity. Although Tyburczy and Waff
363 (1983) reported a negative pressure effect on silicate melts, which are highly
364 polymerized, there has been no relevant conductivity data for carbonate melts as a
365 function of pressure. According to Gaillard et al. (2008), there is a strong correlation
366 between viscosity and conductivity. However, the viscosity of carbonate melts has little
367 pressure effect (Dobson et al., 1996). Another explanation is that the carbonatite melt
368 has a relatively lower conductivity than the carbonate compound composed of alkali
369 elements such as Na and K measured by Gaillard et al. (2008).

370 Fig. 7b shows the conductivity enhancement by the carbonate component in the
371 melt as a function of temperature. The conductivity of carbonate-bearing melt increases
372 more than an order of magnitude with increasing CO₂ concentration in the melt from 13
373 to 45 wt.%. The effect of the carbonate component in the partial melt becomes
374 significantly larger with increasing CO₂ concentration, especially for higher than 40
375 wt.% of CO₂. As the temperature increases, the conductivity dependence on CO₂
376 becomes smaller because of the much lower activation enthalpies for melts with higher
377 CO₂. It is concluded that the effect of the carbonate melt on the bulk rock conductivity
378 is larger at relatively lower temperatures.

379 4.3. Geophysical implications

380 The present study showed that electrical conductivity in carbonate-bearing
381 peridotite increases by a limited magnitude with increasing temperature. Only at very
382 low melt fractions and low temperatures just above the carbonate-bearing peridotite

383 solidus, the partial melting is very effective in raising the electrical conductivity of the
384 upper mantle. Therefore, in this section, we discuss only geological settings in which
385 the carbonate melt has very high CO₂ concentration (> 40 vol. %). Fig. 8 shows the
386 conductivity-depth profile as a function of the melt fraction of carbonatite based on the
387 extrapolation of the trend in Fig. 6. In this figure, the temperature and pressure effects
388 on the conductivity were ignored, because activation enthalpy and volume for electrical
389 conductivity of carbonate melt are very small (Gaillard et al., 2008).

390 Dasgupta and Hirschmann (2006) experimentally demonstrated that the solidus
391 temperature of carbonate peridotite is lower than the upper mantle adiabatic geotherm
392 below the depth of 330 km. In addition, Hirschmann (2010) predicted that carbonatite
393 melt can exist below 150 km depth beneath the oceanic lithosphere based on the
394 thermodynamic model assuming the typical volatile contents (100 wt. ppm for H₂O and
395 60 wt. ppm for CO₂). Thus the conductivity anomaly in such a depth region (150–330
396 km) can be explained by the presence of very small amount of carbonate melt in
397 peridotite. The deep electrical conductivity profile beneath the northwestern Pacific
398 from Hawaii to North America, showed a conductivity anomaly of 10⁻¹ S/m at a depth
399 of 200–250 km (Lizarralde et al., 1995). The conductivity of the partial molten
400 peridotite with a trace amount of carbonatitic melt should be similar to that of the
401 olivine-carbonatite system, because the carbonate concentration in the partial melt of
402 the carbonate peridotite is significantly high when the degree of melting is very low. If
403 this anomaly originates from the presence of carbonatite melt, the melt fraction can be
404 estimated to be less than 0.5 vol.% at that depth (Fig. 8). However, only peridotite with
405 at least 3000 ppm of CO₂ could achieve this value from a carbonatite melt with high
406 CO₂ concentration.

407 A recent one-dimensional (1-D) electrical conductivity model beneath the
408 Philippine Sea revealed an abrupt increase in the conductivity at around 75 km of the

409 upper mantle depths and a constant conductivity at a depth of 75–300km with
410 approximately 0.03 S/m (Baba et al., 2010). The seismic tomography indicates cold
411 slabs currently stagnant beneath the region around the Philippine Sea (Fukao et al.,
412 1992). The subducted slab can add carbonate materials through degassing or melting to
413 the above mantle wedge. The added CO₂ or carbonate melt can raise the conductivity in
414 this region even if the temperature in this region is significantly lower than that in the
415 surrounding mantle. The conductivity value 0.03 S/m can be explained by a presence of
416 0.1 vol.% of carbonatitic melt (Fig. 8). Note that the depth of the abrupt increase in the
417 conductivity (75 km depth) is close to the lower bound of the stabilization of the
418 carbonate melt (e.g., Dalton and Presnall, 1998). We suggest that the large decrease in
419 conductivity above this depth should be attributed to the release of CO₂ from the
420 carbonate melt because of its strong pressure dependence. Seismic evidence for a sharp
421 lithosphere–asthenosphere boundary beneath the Philippine Sea (Kawakatsu et al.,
422 2009) could also imply the presence of carbonate melt in the upper part of the
423 asthenosphere.

424 5. Conclusion

425 We performed electrical conductivity measurement at 3 GPa to elucidate the
426 changes in the electrical conductivity of partially molten carbonate-bearing peridotite as
427 functions of the melt fraction and CO₂ concentration in the melt. When degree of
428 melting is low (< a few volume %), the conductivity of the carbonatitic melt is distinctly
429 higher than that of carbon-free silicate melt. Although the conductivity of the partial
430 molten peridotite increases with increasing temperature, the degree of the increase in
431 the electrical conductivity is obviously small compared with a case in which the melt
432 composition is constant. We identified that the conductivity changes as a consequence
433 of both an increase in the melt fraction and a decrease in the correlated carbonate
434 concentration in the partial melt with increasing temperature. Carbonate is identified as

435 the dominant charge carrier and the conductivity changes of the partial molten rocks are
436 attributed to change in the carbonate mobility controlled by the chemical composition of
437 the melt. When CO₂ concentration in the melt decreases to 0 wt.%, the estimated melt
438 conductivity approaches to that of basaltic melt. Although the partial melt with 10 wt.%
439 CO₂ at higher temperatures has slightly higher conductivity than basaltic melt, the
440 enhancement of electrical conductivity by the carbonate melt is the most effective in a
441 temperature range just above the carbonate peridotite solidus temperature. The electrical
442 conductivity of the partially molten region with carbonate in the upper mantle would
443 increase significantly for the case where the degree of melting is very small (< 1 vol.%).
444 Petrological knowledge of carbonate concentration in the partial molten rocks is needed
445 to estimate the melt fraction from the geophysical signature of the upper mantle.

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551 basaltic and carbonatite melt-bearing peridotites at high pressures: implications for melt
552 distribution and melt fraction in the upper mantle. *Earth Planet. Sci. Lett.* 295, 593-602.

553 Figure captions

554 Fig. 1. Back-scattered electron images (BEI) of polished samples with a chemical
555 composition of KLB1 + dolomite 3 wt.% and with various proportions of carbonate-
556 bearing melt. (a) Run# 1K1364, annealed at 1400 K. (b) Run# 1K1173, annealed at
557 1500 K. (c) Run# 1K1174, annealed at 1600 K. (d) Run# 1K1361, annealed at 1700 K.
558 White bars correspond to 10 μm .

559 Fig. 2. Impedance spectra of carbonate-bearing peridotite samples obtained during
560 cooling as a function of temperature. All samples have a bulk composition of the KLB1
561 + dolomite 3 wt.%. Samples were annealed at 1500 K (a), 1600 K (b), and 1700 K (c).
562 Note the first semicircular arc at high frequencies followed by a pseudo-inductive part
563 and an additional tail derived from electrode reaction at low frequencies.

564 Fig. 3. Electrical conductivity of $(\text{Ca,Mg})\text{CO}_3$ as a function of reciprocal temperature.
565 Note that the conductivity after melting is much higher than that before melting.

566 Fig. 4. Electrical conductivity of the peridotite-dolomite system as a function of
567 reciprocal temperature. (a) Run# 1K1173, peridotite with 3 wt.% carbonatite during the
568 heating (open symbols)-cooling (closed symbols) cycles. (b) All samples for peridotite
569 with and without 3 wt.% dolomite. The symbols indicate raw data of the cooling path
570 after annealing at the maximum temperature for each sample with different melt
571 fractions. Abbreviations; C06: the latest model of olivine electrical conductivity at 0.1
572 MPa under IW (iron-wüstite) buffers from *Constable* (2006). YMYK06: a conductivity
573 range of electrical conductivity of olivine single crystal at 3 GPa under Ni-NiO buffer
574 from Yoshino et al. (2006).

575 Fig. 5. Electrical conductivity and melt fraction versus temperature for the system with
576 KLB1 + 3 wt.% dolomite.

577 Fig. 6. Relationship between the melt fractions and electrical conductivities for the
578 carbonate-bearing partial molten peridotite. Data of the olivine + carbonatitic melts
579 (1650 K and 3 GPa) with constant CO₂ concentration in the melt, and the olivine +
580 basalt melts (1.5 GPa and 1600 K; i.e. no CO₂) are also plotted (Yoshino et al., 2010).
581 The shaded region indicates a typical range of conductivity values of basaltic melt (e.g.,
582 Presnell et al., 1972). The thick dashed line indicates an expected conductivity-melt
583 fraction relation for the carbonate-bearing peridotite system. Star symbol denotes
584 conductivity of dolomite melt, which is an end member of the system used for the
585 present study at 1700 K. Note that the trend of the carbonate-bearing partial molten
586 peridotite (KLB1 + dolomite 3 wt.%) is inconsistent with the conductivity value of
587 dolomite melt.

588 Fig. 7. Log electrical conductivity σ of melt in the KLB1 + dolomite 3 wt.% system as a
589 function of CO₂ concentration in the melt. (a) Symbols represent the log σ of the
590 carbonate-bearing melt calculated at 1700 K assuming various activation enthalpies for
591 the electrical conductivity of the melt. Orange circle represents the conductivity value
592 of dolomite melt at 1700 K. The dashed line denotes the CO₂ concentration in dolomite
593 (Ca,Mg)CO₃. The red square symbols indicate the conductivity values of silicate melt
594 (TW83: Tyburezy and Waff, 1983; KAB, Kilauea alkali basalt: Pommier et al., 2010).
595 The green area indicates the conductivity range of the carbonate melt determined at
596 atmospheric pressure (Gaillard et al., 2008). Note that as CO₂ concentration in the melt
597 increases, the conductivity of the partial melt in carbonate-bearing peridotite increases
598 by nearly an order of magnitude. The yellow dashed arrows denote a predicted trend of
599 the melt conductivity. (b) The log σ of the carbonate-bearing melt as a function of CO₂
600 concentration in the melt for different temperatures based on a case for variable
601 activation enthalpy (Table 3). Note that as temperature decreases, the conductivity
602 depends more strongly on the CO₂ concentration in the melt.

603 Fig. 8. Melt fraction of carbonatite melt with a high CO₂ concentration (> 40 wt. %)
604 estimated from the reference one-dimensional (1-D) conductivity models obtained by
605 geophysical observations. The yellow colored area denotes the reference 1-D models for
606 the north Pacific obtained by Lizarralde et al. (1995). The red line indicates the 1-D
607 model for the Philippine Sea mantle (Baba et al., 2010).

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608 Table 1. Summary of runs

609	Run No.	vol.% ^a	T _{max} (K)	log σ_{\max} (S/m) ^b	CO ₂ (wt.%) ^c	Phase assemblage ^d	Remarks
610	KLB1						
611	1K1178	0	1700	-1.87	0	ol(Fe ₉₀)-opx-cpx-grt	
612	KLB1 + Dolomite 1 wt.%						
613	1K1161	1.0	1400	-0.25	n.d.	ol(Fe ₉₀)-opx-cpx-grt-melt	
614	1K1158	20	-	0.50	5(1)	ol(Fe ₉₃)-melt	Incorrect T reading
615	KLB1 + Dolomite 3 wt.%						
616	1K1364	0.5	1400	-0.48	~50	ol(Fe ₉₀)-opx-cpx-grt-dol-melt	
617	1K1173	1.8	1500	-0.21	45(3)	ol(Fe ₉₁)-opx-cpx-grt-dol-melt	
618	1K1174	11	1600	0.18	33(2)	ol(Fe ₉₃)-cpx-melt	
619	1K1363	14.7	1650	0.49	20(1)	ol(Fe ₉₃)-cpx-melt	
620	1K1361	20.3	1700	0.29	13(3)	ol(Fe ₉₂)-cpx-melt	
621	Dolomite						
622	S2367	100	1800	2.02	47.7	melt	$\Delta H = 38$ kJ/mol $\sigma_0 = 1343$ S/m

624 All experiments were conducted at 3 GPa.

625 a: Volume percent of melt phase in run products determined by image analysis.

626 b: Log conductivity measured at maximum temperature just before quenching.

627 c: CO₂ concentration in melt estimated from the total weight deficit of the EPMA analysis.

628 d: Abbreviations: ol; olivine, opx; orthopyroxene, cpx; clinopyroxene, grt; garnet, dol; dolomite

629

630 Table 2. Melt composition of run products

	1K1173	1K1174	1K1363	1K1361	
	1500 K	1600 K	1650 K	1700 K	
633	SiO ₂	5.10(26)	18.90(89)	32.18(151)	38.94(93)
634	TiO ₂	0.42(10)	0.02(23)	0.55(12)	0.48(10)
635	Al ₂ O ₃	2.03(62)	2.15(48)	12.18(53)	11.05(51)
636	FeO*	7.47(141)	4.52 (101)	3.57(102)	7.07(89)
637	NiO	0.02(2)	0.02(3)	0.06(6)	0.03(3)
638	MnO	0.17(5)	0.13(2)	0.18(1)	0.16(1)
639	MgO	13.58(491)	14.20(114)	7.26(115)	13.01(242)
640	CaO	24.99(225)	24.95(372)	24.05(134)	14.68(503)
641	Na ₂ O	1.10(23)	1.57(23)	1.07(4)	1.54(34)
642	K ₂ O	0.35(7)	0.58(5)	0.45(4)	0.14(7)
643	Total	55.24(269)	66.97(103)	81.55(128)	87.10(165)

644 The chemical compositions of melt were measured by the electron probe
 645 microanalyzer under the operating condition of 15 kV and 12 nA.

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649

650 Table 3. Parameters used for calculation of melt conductivity

	1K1364	1K1173	1K1174	1K1363	1K1361	
	1400 K	1500 K	1600 K	1650 K	1700 K	
653	Cases for the fixed activation enthalpy					
654	150 kJ/mol					
655	log σ_0 (S/m)	7.21	6.76	6.03	5.96	5.59
656	log σ_{1700K} (S/m)	2.60	2.15	1.43	1.35	0.98
657	38 kJ/mol					
658	log σ_0 (S/m)	3.03	2.86	2.38	2.41	2.15
659	log σ_{1700K} (S/m)	1.87	1.69	1.21	1.25	0.98
660	A case for variable activation enthalpy					
661	ΔH (kJ/mol)	38	49	76	105	121
662	log σ_0 (S/m)	3.03	3.25	3.62	4.54	4.70

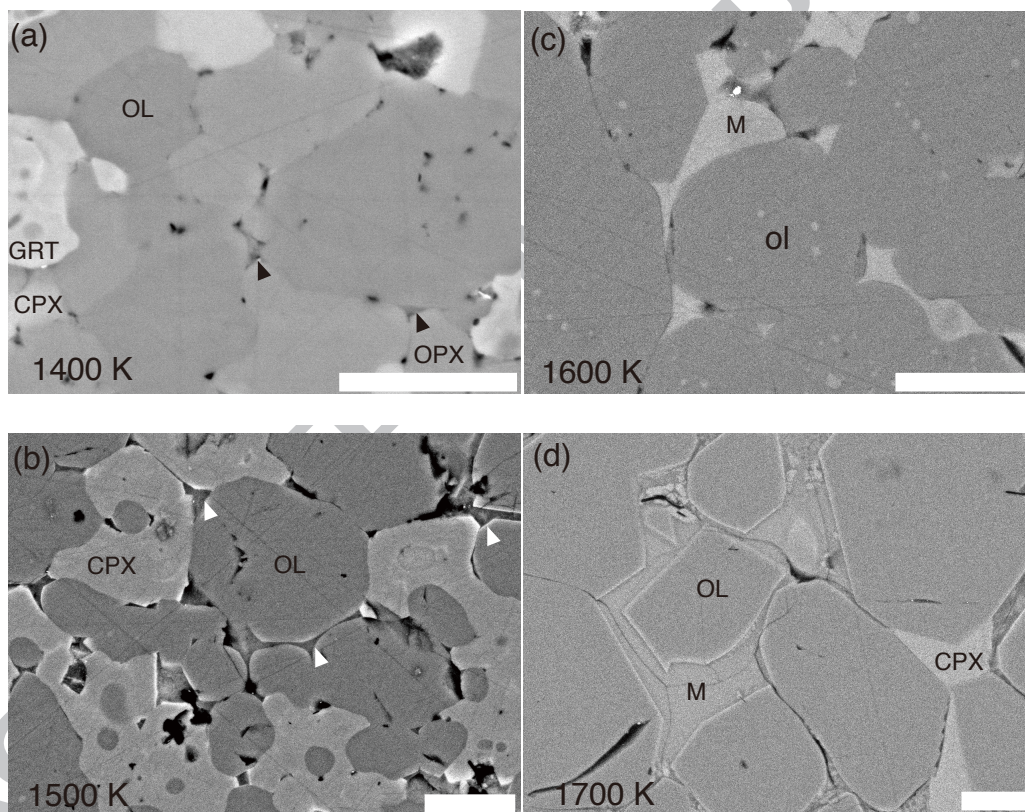


Fig. 1. Yoshino et al.

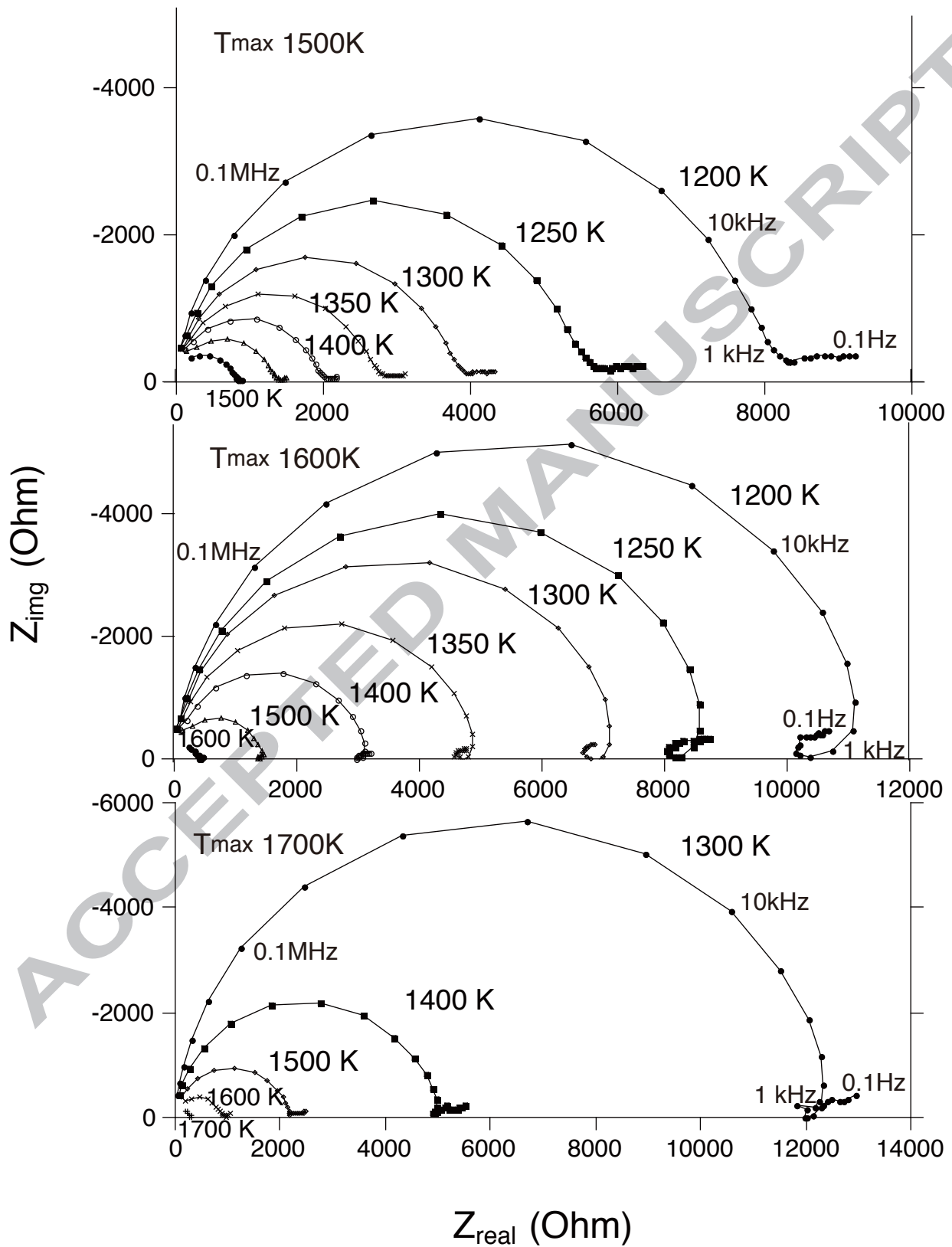


Fig. 2. Yoshino et al.

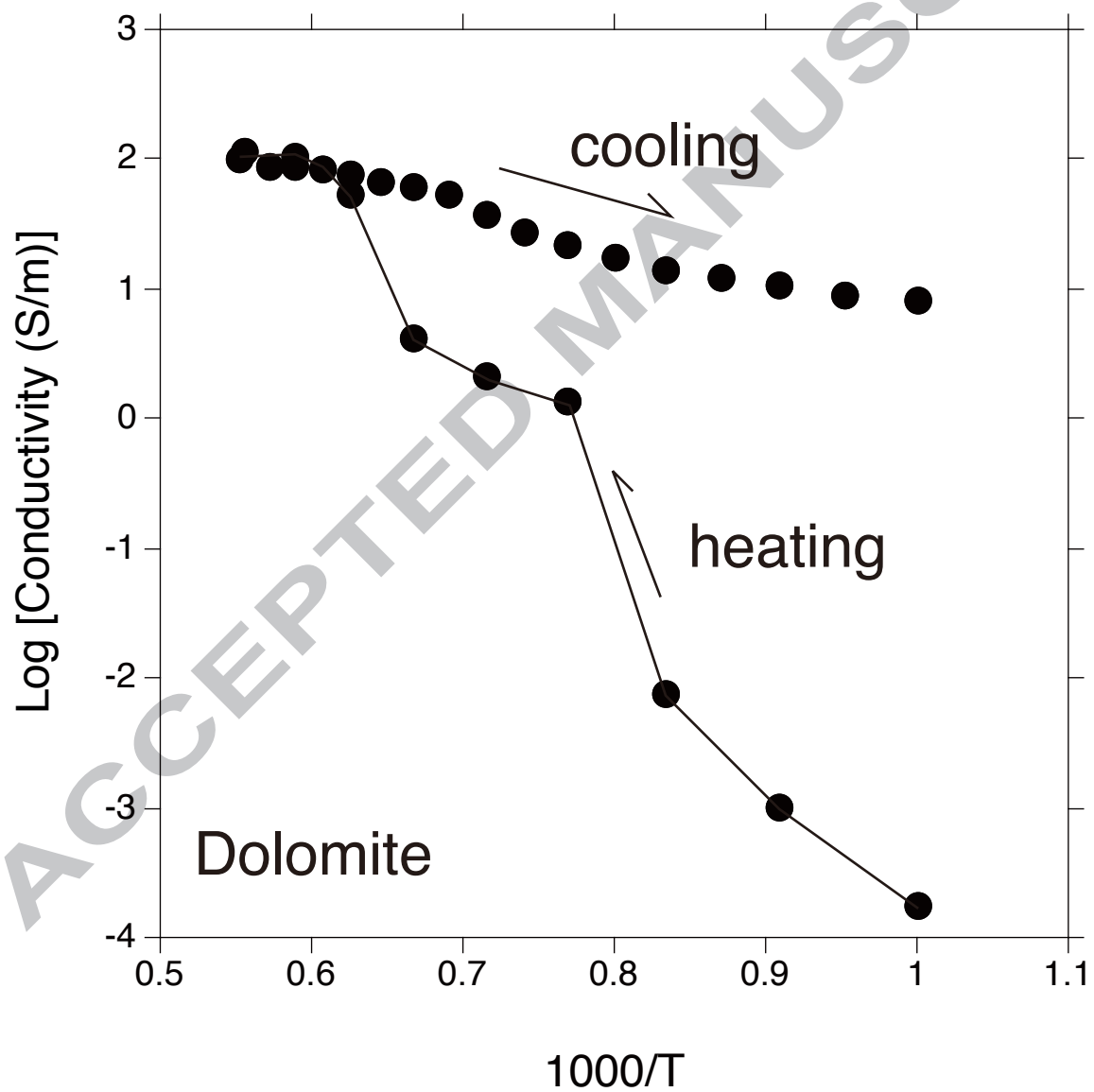


Fig. 3. Yoshino et al.

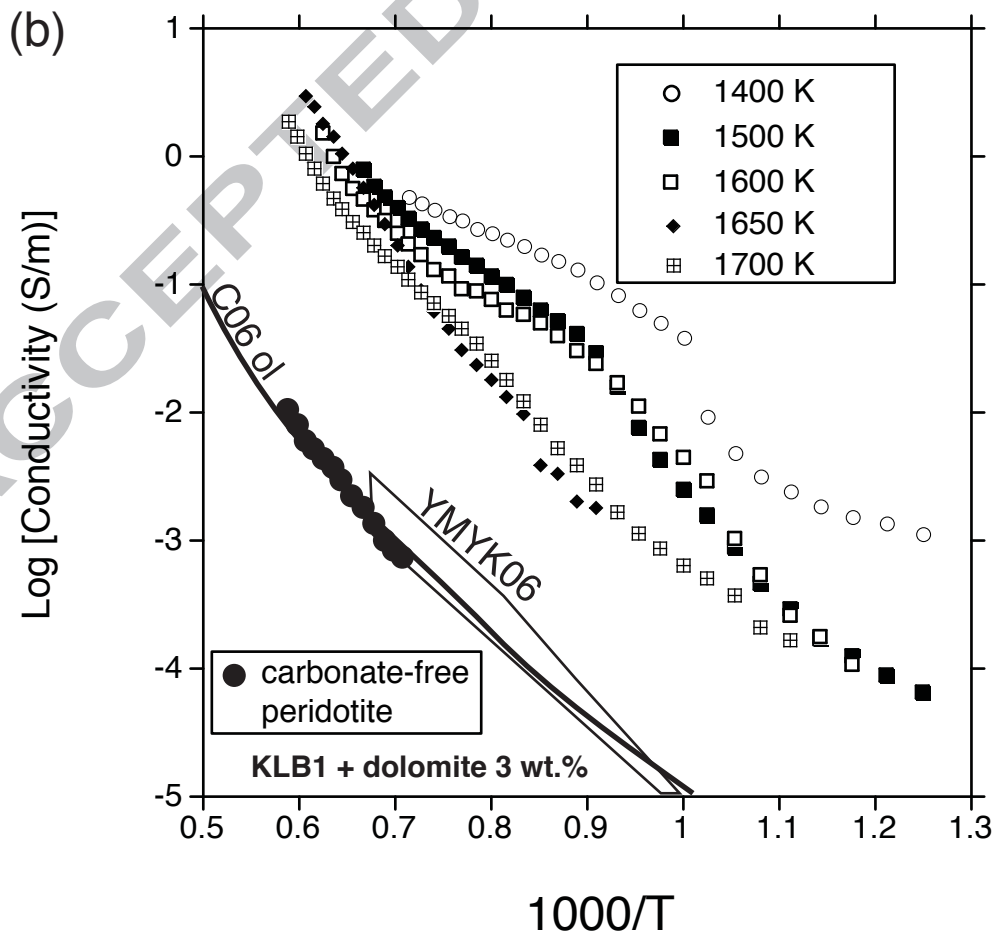
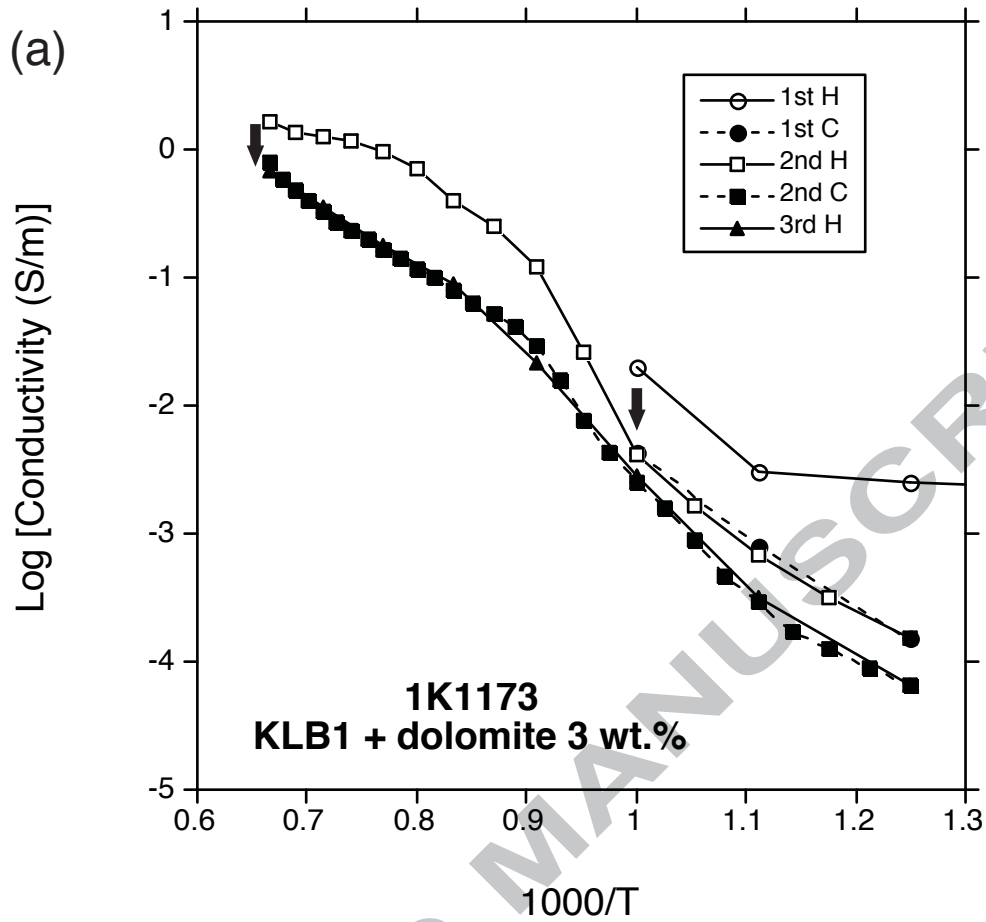


Fig. 4. Yoshino et al.

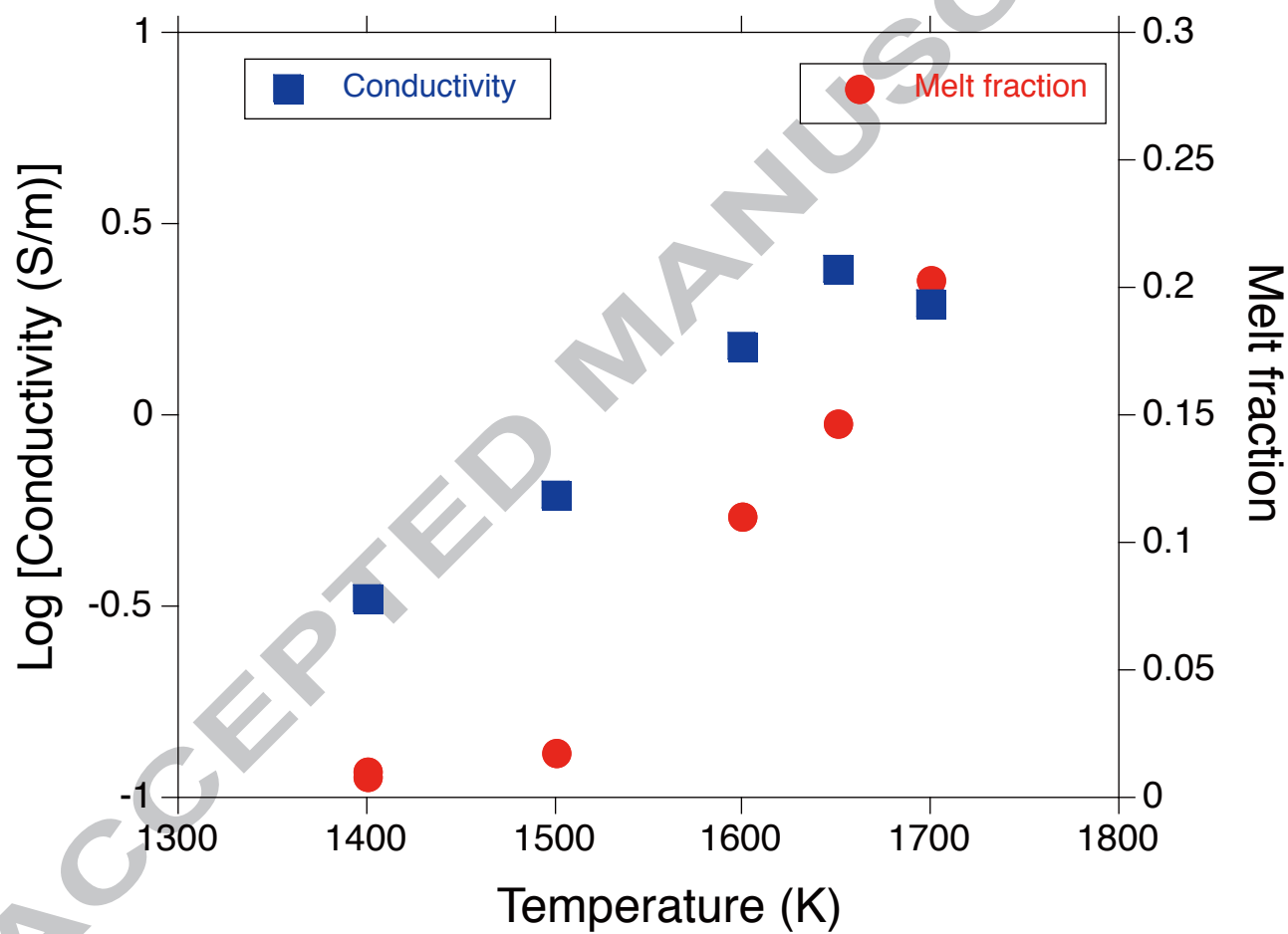


Fig. 5. Yoshino et al.

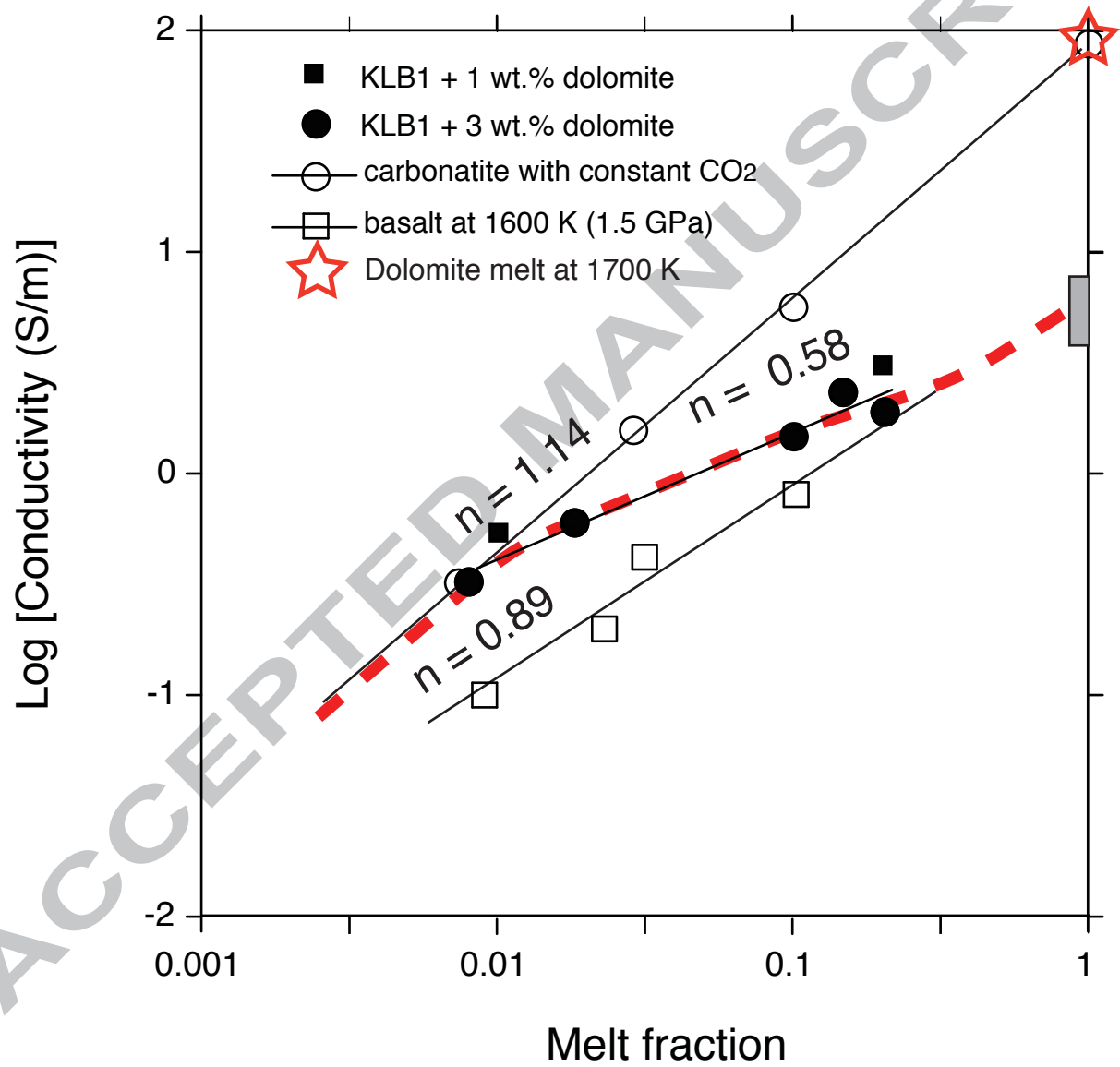


Fig. 6. Yoshino et al.

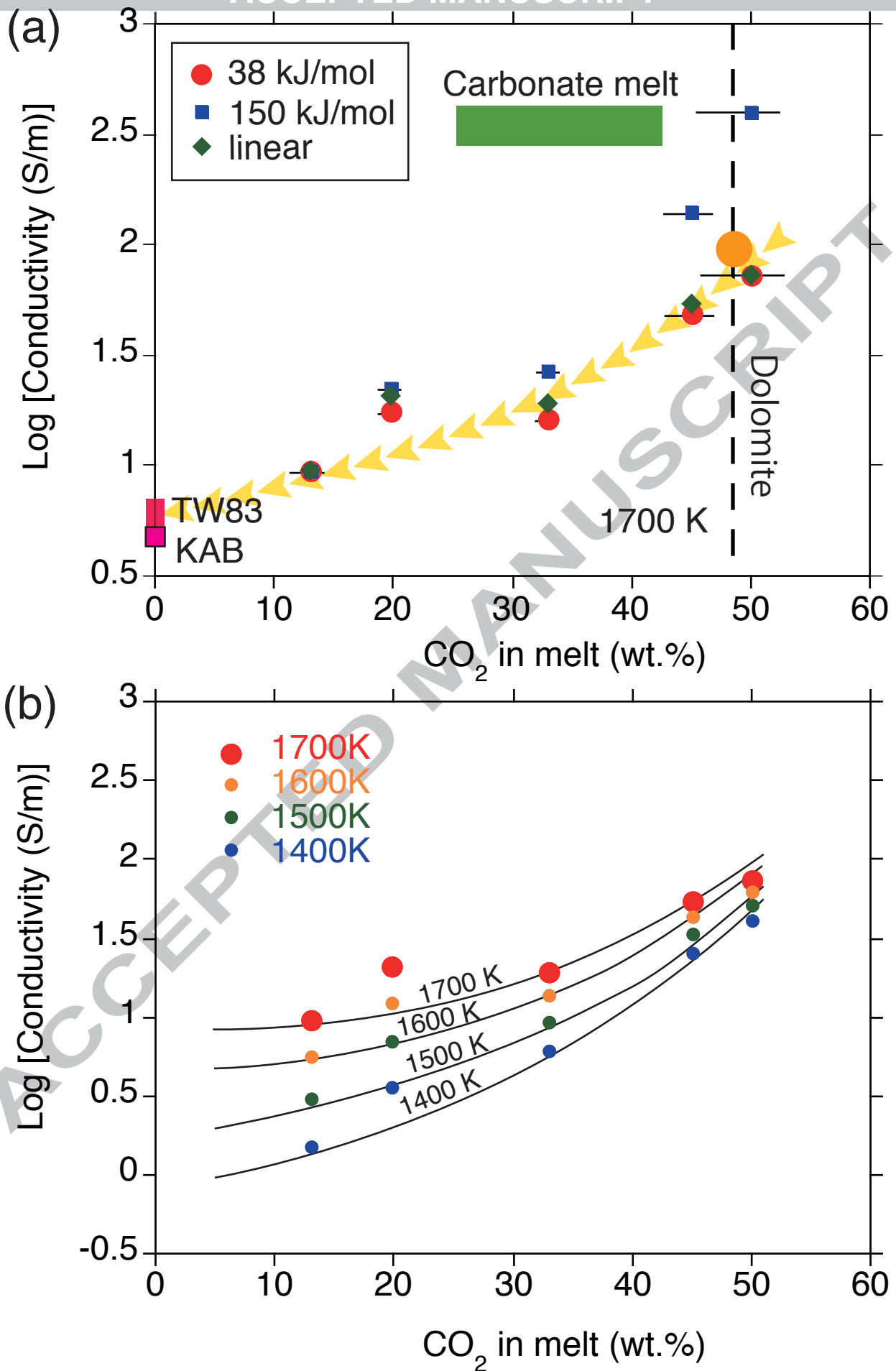


Fig. 7. Yoshino et al.

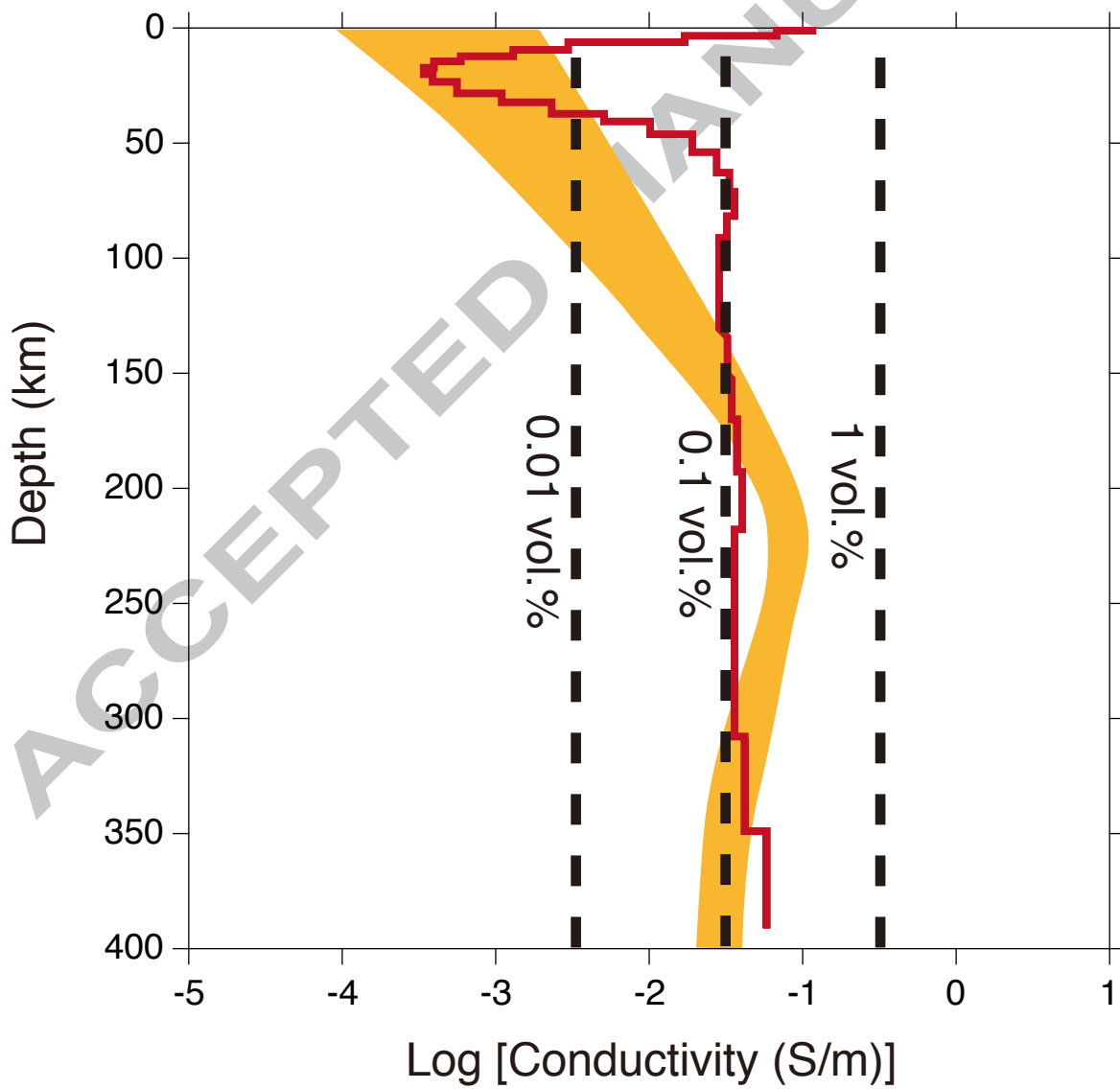


Fig. 8. Yoshino et al.

663 Highlights

- 664 1. We investigated electrical conductivity of partially molten carbonate peridotite.
- 665 2. On the solidus, the conductivity was markedly higher than that of carbonate-free
666 peridotite.
- 667 3. Electrical conductivity is not markedly increased by higher melting degree.
- 668 4. The moderate increase is attributed to a decrease in carbonate content in the partial melt.
- 669 5. The conductivity in the upper mantle is enhanced by very small degree of melting.
- 670

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