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1 Electrical conductivity of partial molten carbonate peridotite

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Effect of carbonate content on electrical conductivity

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

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30 1. Introduction

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

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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 a wide range of mantle conditions. Thus, to assess conductivity anomalies in the upper 51 mantle, we need to know the effect of the carbonate component in partial melt on the 52 bulk conductivity of the partially molten peridotite under high pressure. In this study, 53 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 temperature conditions. Since we used starting materials with a fixed carbonate 56 concentration, the variation in the degree of partial melting can provide the variation in 57 carbonate concentration in the partial melt. In addition, we measured conductivity of 58 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 was estimated as a function of CO₂ concentration in the melt. We obtained CO₂ 61 62 concentration dependence of electrical conductivity by comparing the data obtained from our previous results, in which the conductivity was obtained for the partial molten 63 carbonate peridotite with fixed CO₂ concentration in melt. Such an argument provides 64 the constraints on the presence of carbonate partial melt and on the thermal structure of 65 the upper mantle. 66

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

4

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

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

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

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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 (CO ₂) 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 (< 1μ m). 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

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

of the partially molten zone. As mentioned above, the outer part of the sample had less iron contents. The sample conductivity of the uncontaminated part of the sample should

be slightly higher than the obtained value (less than 10 %).

For the 3 wt.% dolomite-bearing peridotite system, the dolomite completely 128 decomposed below 1600 K. Orthopyroxene disappeared because of a decarbonation 129 reaction such as MgSiO₃ (in orthopyroxene) + MgCO₃ (in dolomite) = Mg₂SiO₄ (in 130 olivine) + CO₂. Clinopyroxene was present up to 1700 K, although by volume the 131 132 proportion of clinopyroxene decreased with increasing temperature. The carbonate concentration (CO₂) in the partial melt of the samples decreased from 45 to 13 wt.% 133 with increasing melt fraction from 0.02 to 0.2 (Tables 1 and 2). In the case of the KLB1 134 + 1 wt.% dolomite system, the CO₂ concentration in melt decreased to 5 wt.% when the 135 melt fraction was 0.20. We were not able to measure the melt composition accurately 136 for samples containing the lowest melt fraction ($\leq 1 \text{ vol.}\%$) using the electron 137 microprobe. However, the carbonate component of the sample is usually expected to be 138 40 wt.% just above the solidus (Dasgupta et al., 2007). Thus the CO₂ concentration in 139 the partial melt largely decreased with increasing annealing temperature and melt 140 141 fraction. The CO₂ concentrations in the melt estimated by EPMA were slightly higher than those expected from the bulk composition. For example, when we assume that all 142 carbons partition into the melt phase, and the densities of the carbonate and basaltic 143 melt range from 2 to 2.7 g/cm³ (e.g., Dobson et al., 1996; Rigden et al., 1986), the 144 145 KLB1 + dolomite 3 wt.% system with 20 vol.% melting requires around 10 wt.% CO₂ concentration in the partial melt. The CO_2 concentrations in the melt were increased by 146 147 partial solidification of the melt due to reaction with the MgO capsule.

148 3.2. Electrical conductivity

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

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

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

170
$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_0 \exp\left(-\frac{H}{kT}\right)$$

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

(1)

dolomite melt is 1343 S/m, which is lower than the values (3440 S/m) reported from
Gaillard et al. (2008). Therefore, the absolute conductivity value of the dolomite melt
was slightly lower than those of alkali carbonate melt [KCa_{0.5}]₂(CO₃)₂,
(NaKCa_{0.5})₂(CO₃)₃, (NaKCa)(CO₃)₂, (NaK)₂(CO₃)₂, and (LiNaK)₂(CO₃)₃] reported by
Gaillard et al. (2008).

8

An example of the conductivity measurements for the partial molten system is 181 182 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 second heating, the conductivity largely increased by nearly two orders of magnitude up 184 to 1100 K. This temperature is consistent with the temperature at which the conductivity 185 of Ca-rich carbonate abruptly increases during heating (e.g. Gaillard et al., 2008). 186 Above 1200 K, the conductivity slightly increased with increasing temperature up to 187 1500 K. At the beginning of annealing at 1500 K, the sample conductivity rapidly 188 decreased and then became constant within 2 hours, similar to our previous studies 189 (Yoshino et al., 2010). At the beginning of partial melting, the powder with large 190 porosity was instantaneously filled with melt. Therefore, the conductivity was initially 191 high. As the system established textural equilibrium during annealing, some pores were 192 closed to minimize the total interfacial energy in the system. This process led to the 193 reduction of the bulk conductivity of the sample. The impedance spectra showed part of 194 195 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 showed linear trends with some different slopes, suggesting the conduction mechanism 200 201 changes in a certain temperature range. In the subsequent heating path, the 202 reproducibility was confirmed.

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In the third heating, the conductivity values at the highest temperature were 203 204 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 obtain the partial molten texture of the final state. The conductivity values shown in 206 Table 1 were obtained just before quenching. Therefore, the sample texture and the 207 conductivity values represent the same condition as the second heating. Fig. 5 shows a 208 209 plot of the conductivity values at the maximum temperature and melt fraction against the highest temperature experienced for each sample. The electrical conductivity of the 210 211 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 1700 K. 213

Fig. 4b shows the logarithmic plot of conductivity versus reciprocal temperature 214 for the KLB1 + dolomite 3 wt.% system in the second cooling path. The electrical 215 conductivity of the carbonate-bearing peridotite is distinctly higher than that of 216 carbonate- and melt-free peridotite, which is similar to that of olivine (Constable 2006; 217 Yoshino et al., 2006; 2009). The samples annealed at higher temperatures showed lower 218 219 conductivity values at the same temperature, and larger temperature dependence compared with the sample annealed at lower temperatures. Although samples have the 220 221 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 melts (e.g., Presnall et al., 1972; Gaillard et al., 2008). A small temperature dependence 226 on the conductivity of samples annealed at lower temperature was quite similar to that 227 of dolomite end member, suggesting that carbonate concentration in the melt was 228 229 relatively high. This trend is consistent with a variation of the carbonate concentration

in the partial melt determined by the weight deficiency of the microprobe analysis

231 (Table 2).

232 **4. Discussion**

4.1. Relationship between conductivity and melt fraction

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

248
$$\sigma_{\text{bulk}} = C\phi^n \sigma_{\text{m}} \tag{2}$$

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

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also valid for the other mixing models in the system containing the well-interconnectedphase.

In this study, the melt fraction in the sample changes with temperature under the 255 constant bulk CO_2 composition (Fig. 6). The conductivity of the bulk rock increases 256 with increasing melt fraction in this study, which is in the same tendency with that in 257 the previous study, where CO_2 concentration is constant not in the bulk rock but in the 258 259 melt (Yoshino et al., 2010). However, an increase of conductivity values with melt fraction in this study is smaller than that in the previous study. The present system 260 yields much smaller exponent for Eq. (2), namely n = 0.58. In addition, the conductivity 261 values of dolomite melt, which is an end member of this system, does not agree with the 262 extrapolated trend (Fig. 6). 263

264 The Archie's law is only applicable to the system with constant melt conductivity. Thus, the "apparent" low power exponent (n = 0.58) obtained in this study 265 contains information about variation in melt conductivity with melt fraction. There are 266 two factors to vary the melt conductivity. One is temperature and the other is the CO_2 267 concentration in the partial melt. First we consider thermal effect. The melt conductivity 268 generally increases with increasing temperature followed by Arrhenian relation. If we 269 consider the sample conductivity at 1700 K in order to directly compare the results of 270 271 our previous study (Yoshino et al., 2010), the power exponent would be much smaller than 0.58, because the conductivity measured at lower temperature is relatively higher 272 273 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

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279	conductivity of the partial melt is expected to be controlled by the CO_2 content in the
280	partial melt. In general, carbonatitic partial melts containing ~ 40 wt.% CO ₂ 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.% CO ₂) to carbonated silicate melt (\leq 25 wt.% CO ₂) in carbonate-bearing
289	peridotiteat 3 GPa occurs abruptly at 1623 K. This study also showed that the CO_2
290	content in the partial melt is even lower at higher temperatures (~13 wt.% CO_2 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.% CO_2) 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.

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

We next estimate the electrical conductivity of the carbonate-bearing melt itself as a function of the CO_2 concentration in the melt to compare it directly with melts with

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

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

315 2) The effect of temperature on the conductivity of the melt is controlled by the activation enthalpy for electrical conductivity of the melts. The activation enthalpy of 316 dolomite melts was 38 kJ/mol at 3 GPa. On the other hand, the average activation 317 enthalpies of basaltic melt are more than three times higher (110-150 kJ/mol) (Tyburczy 318 and Waff, 1983; Gaillard and Marziano, 2005). Thus, the temperature dependence of 319 the conductivity of the carbonate melts is much smaller than that of silicate melts. Since 320 silicate melts are likely to polymerize as the SiO₂ component in the melt increases, its 321 activation enthalpy would increase with the increasing SiO₂ component in the carbonate 322 melt. In the present study, the CO_2 concentration in the melt varies considerably. 323 324 Therefore, the activation enthalpy is expected to gradually increase with decreasing CO_2 325 concentration in the melt, because there is a negative-correlation between the SiO_2 and 326 CO₂ concentrations in the carbonate melt (Table 2).

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

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

Fig. 7a shows the logarithmic plot of the electrical conductivity of the partial 342 melt in the KLB1 + dolomite 3 wt.% system as a function of the CO_2 concentration in 343 the melt at 1700 K. Although the log of electrical conductivity was calculated using a 344 345 possible range of activation enthalpies, the activation enthalpy for the melt conductivity with higher CO_2 concentration (> 40 wt. %) should be low and close to that of dolomite 346 melt (38 kJ/mol), whereas that with lower CO₂ concentration should be higher and 347 approach that of silicate melt (110-150 kJ/mol). Considering the effect of the CO₂ 348 349 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 CO_2 agrees well with the basaltic melt conductivity (TW83: Tyburczy and Waff, 1983; 353 KAB: Pommier et al., 2010). A variation of the estimated melt conductivity as a 354 function of CO₂ concentration shows a plateau between 33 and 20 wt.% CO₂. If there 355 356 exists a threshold for interconnection of chains of silicate tetrahedral in melt, an abrupt

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change of melt conductivity and its activation enthalpy might be present. On the other 357 358 hand, the conductivity of the melt with higher CO_2 concentration is quite consistent 359 with that of dolomite melt measured at 3 GPa, which is lower than those of the alkali carbonate melt determined at atmospheric pressure (Gaillard et al., 2008). One 360 explanation for this discrepancy between dolomite and alkali carbonate melt is a 361 negative pressure dependence on the melt conductivity. Although Tyburczy and Waff 362 (1983) reported a negative pressure effect on silicate melts, which are highly 363 polymerized, there has been no relevant conductivity data for carbonate melts as a 364 365 function of pressure. According to Gaillard et al. (2008), there is a strong correlation between viscosity and conductivity. However, the viscosity of carbonate melts has little 366 pressure effect (Dobson et al., 1996). Another explanation is that the carbonatite melt 367 has a relatively lower conductivity than the carbonate compound composed of alkali 368 369 elements such as Na and K measured by Gaillard et al. (2008).

Fig. 7b shows the conductivity enhancement by the carbonate component in the 370 371 melt as a function of temperature. The conductivity of carbonate-bearing melt increases more than an order of magnitude with increasing CO₂ concentration in the melt from 13 372 373 to 45 wt.%. The effect of the carbonate component in the partial melt becomes significantly larger with increasing CO₂ concentration, especially for higher than 40 374 wt.% of CO₂. As the temperature increases, the conductivity dependence on CO₂ 375 376 becomes smaller because of the much lower activation enthalpies for melts with higher 377 CO_2 . It is concluded that the effect of the carbonate melt on the bulk rock conductivity is larger at relatively lower temperatures. 378

379 4.3. Geophysical implications

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

solidus, the partial melting is very effective in raising the electrical conductivity of the upper mantle. Therefore, in this section, we discuss only geological settings in which the carbonate melt has very high CO_2 concentration (> 40 vol. %). Fig. 8 shows the conductivity-depth profile as a function of the melt fraction of carbonatite based on the extrapolation of the trend in Fig. 6. In this figure, the temperature and pressure effects on the conductivity were ignored, because activation enthalpy and volume for electrical conductivity of carbonate melt are very small (Gaillard et al., 2008).

Dasgupta and Hirschmann (2006) experimentally demonstrated that the solidus 390 temperature of carbonate peridotite is lower than the upper mantle adiabatic geotherm 391 below the depth of 330 km. In addition, Hirschmann (2010) predicted that carbonatite 392 melt can exist below 150 km depth beneath the oceanic lithosphere based on the 393 thermodynamic model assuming the typical volatile contents (100 wt. ppm for H_2O and 394 60 wt. ppm for CO_2). Thus the conductivity anomaly in such a depth region (150~330 395 km) can be explained by the presence of very small amount of carbonate melt in 396 peridotite. The deep electrical conductivity profile beneath the northwestern Pacific 397 from Hawaii to North America, showed a conductivity anomaly of 10^{-1} S/m at a depth 398 of 200-250 km (Lizarralde et al., 1995). The conductivity of the partial molten 399 peridotite with a trace amount of carbonatitic melt should be similar to that of the 400 olivine-carbonatite system, because the carbonate concentration in the partial melt of 401 402 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 CO_2 could achieve this value from a carbonatite melt with high CO_2 concentration. 406

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

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17

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 CO_2 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 CO ₂ 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

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

the dominant charge carrier and the conductivity changes of the partial molten rocks are 435 436 attributed to change in the carbonate mobility controlled by the chemical composition of 437 the melt. When CO_2 concentration in the melt decreases to 0 wt.%, the estimated melt conductivity approaches to that of basaltic melt. Although the partial melt with 10 wt.% 438 CO₂ at higher temperatures has slightly higher conductivity than basaltic melt, the 439 enhancement of electrical conductivity by the carbonate melt is the most effective in a 440 441 temperature range just above the carbonate peridotite solidus temperature. The electrical conductivity of the partially molten region with carbonate in the upper mantle would 442 443 increase significantly for the case where the degree of melting is very small (< 1 vol.%). Petrological knowledge of carbonate concentration in the partial molten rocks is needed 444 to estimate the melt fraction from the geophysical signature of the upper mantle. 445

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553 Figure captions

- Fig. 1. Back-scattered electron images (BEI) of polished samples with a chemical
- composition of KLB1 + dolomite 3 wt.% and with various proportions of carbonate-
- 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 $10 \,\mu m$.
- 559 Fig. 2. Impedance spectra of carbonate-bearing peridotite samples obtained during
- cooling as a function of temperature. All samples have a bulk composition of the KLB1
- + 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
- and an additional tail derived from electrode reaction at low frequencies.
- Fig. 3. Electrical conductivity of $(Ca,Mg)CO_3$ as a function of reciprocal temperature.
- Note that the conductivity after melting is much higher than that before melting.

Fig. 4. Electrical conductivity of the peridotite-dolomite system as a function of 566 reciprocal temperature. (a) Run# 1K1173, peridotite with 3 wt.% carbonatite during the 567 heating (open symbols)-cooling (closed symbols) cycles. (b) All samples for peridotite 568 with and without 3 wt.% dolomite. The symbols indicate raw data of the cooling path 569 after annealing at the maximum temperature for each sample with different melt 570 571 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 from Yoshino et al. (2006). 574

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

Effect of carbonate content on electrical conductivity

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 CO_2 concentration in the melt, and the olivine + basalt melts (1.5 GPa and 1600 K; i.e. no CO₂) are also plotted (Yoshino et al., 2010). 580 The shaded region indicates a typical range of conductivity values of basaltic melt (e.g., 581 Presnell et al., 1972). The thick dashed line indicates an expected conductivity-melt 582 583 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 present study at 1700 K. Note that the trend of the carbonate-bearing partial molten 585 peridotite (KLB1 + dolomite 3 wt.%) is inconsistent with the conductivity value of 586 dolomite melt. 587

Fig. 7. Log electrical conductivity σ of melt in the KLB1 + dolomite 3 wt.% system as a 588 function of CO₂ concentration in the melt. (a) Symbols represent the log σ of the 589 carbonate-bearing melt calculated at 1700 K assuming various activation enthalpies for 590 the electrical conductivity of the melt. Orange circle represents the conductivity value 591 of dolomite melt at 1700 K. The dashed line denotes the CO₂ concentration in dolomite 592 593 (Ca,Mg)CO₃. The red square symbols indicate the conductivity values of silicate melt (TW83: Tyburczy and Waff, 1983; KAB, Kilauea alkali basalt: Pommier et al., 2010). 594 595 The green area indicates the conductivity range of the carbonate melt determined at 596 atmospheric pressure (Gaillard et al., 2008). Note that as CO_2 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 concentration in the melt for different temperatures based on a case for variable 600 activation enthalpy (Table 3). Note that as temperature decreases, the conductivity 601 depends more strongly on the CO₂ concentration in the melt. 602

26

- Fig. 8. Melt fraction of carbonatite melt with a high CO_2 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
- the north Pacific obtained by Lizarralde et al. (1995). The red line indicates the 1-D
- model for the Philippine Sea mantle (Baba et al., 2010).

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Run No.	vol.% ^a	T _{max} (K)	$log\sigma_{max}$ (S/m)	^b CO ₂ (wt.%) ^c	Phase assemblage d	Remarks
KLB1						
1K1178	0	1700	-1.87	0	ol(Fo90)-opx-cpx-grt	
KLB1 + Do	olomite 1 wt.%					
1K1161	1.0	1400	-0.25	n.d.	ol(Fo90)-opx-cpx-grt-melt	
1K1158	20	-	0.50	5(1)	ol(Fo ₉₃)-melt	Incorrect T reading
KLB1 + Do	olomite 3 wt.%					
1K1364	0.5	1400	-0.48	~50	ol(Fo90)-opx-cpx-grt-dol-melt	
1K1173	1.8	1500	-0.21	45(3)	ol(Fo91)-opx-cpx-grt-dol-melt	
1K1174	11	1600	0.18	33(2)	ol(Fo93)-cpx-melt	
1K1363	14.7	1650	0.49	20(1)	ol(Fo ₉₃)-cpx-melt	
1K1361	20.3	1700	0.29	13(3)	ol(Fo92)-cpx-melt	
Dolomite						
S2367	100	1800	2.02	47.7	melt	$\Delta H = 38 \text{ kJ/mol}$
						$\sigma_0 = 1343 \text{ S/m}$

All experiments were conducted at 3 GPa.

624 625 626 627 a: Volume percent of melt phase in run products determined by image analysis. .are ju. otal weigt. .ene, epx; elino,

b: Log conductivity measured at maximum temperature just before quenching.
c: CO₂ concentration in melt estimated from the total weight deficit of the EPMA analysis.

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

Table 2. Melt composition of run products

	1K1173 1500 K	1K1174 1600 K	1K1363 1650 K	1K1361 1700 K	
SiO ₂	5.10(26)	18.90(89)	32.18(151)	38.94(93)	
TiO ₂	0.42(10)	0.02(23)	0.55(12)	0.48(10)	
Al_2O_3	2.03(62)	2.15(48)	12.18(53)	11.05(51)	
FeO*	7.47(141)	4.52 (101)	3.57(102)	7.07(89)	
NiO	0.02(2)	0.02(3)	0.06(6)	0.03(3)	
MnO	0.17(5)	0.13(2)	0.18(1)	0.16(1)	
MgO	13.58(491)	14.20(114)	7.26(115)	13.01(242)	
CaO	24.99(225)	24.95(372)	24.05(134)	14.68(503)	
Na ₂ O	1.10(23)	1.57(23)	1.07(4)	1.54(34)	
K_2O	0.35(7)	0.58(5)	0.45(4)	0.14(7)	
Total	55.24(269)	66.97(103)	81.55(128)	87.10(165)	
The cher	mical composition	ns of melt we	re measured b	y the electron prob	be la
microan	alvzer under the o	operating cond	dition of 15 kV	and 12 nA.	
	,	1 0			

650	Table 3. Parameters used for calculation of melt conductivity							
651		1K1364	1K1173	1K1174	1K1363	1K1361		
652		1400 K	1500 K	1600 K	1650 K	1700 K		
653	Cases for the	fixed activation	on enthalpy					
654	150 kJ/mol							
655	$\log \sigma_0$ (S/m)	7.21	6.76	6.03	5.96	5.59		
656	$\log \sigma_{1700\mathrm{K}}$ (S/r	n)2.60	2.15	1.43	1.35	0.98		
57	38 kJ/mol			4				
58	$\log \sigma_0$ (S/m)	3.03	2.86	2.38	2.41	2.15		
59	$\log \sigma_{1700K}$ (S/r	n) 1.87	1.69	1.21	1.25	0.98		
000	A case for va	riable activation	on enthalpy		105	101		
100	ΔH (kJ/mol)	38	49	76	105	121		
	10800 (0,111)							
	C		2					
(6							



Fig. 1. 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.

Effect of carbonate content on electrical conductivity

- 663 Highlights
- 1. We investigated electrical conductivity of partially molten carbonate peridotite. 664
- 2. On the solidus, the conductivity was markedly higher than that of carbonate-free 665 peridotite. 666
- 3. Electrical conductivity is not markedly increased by higher melting degree. 667

- 4. The moderate increase is attributed to a decrease in carbonate content in the partial melt. 668
- 5. The conductivity in the upper mantle is enhanced by very small degree of melting. 669
- 670