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Electrical conductivity of metasomatized lithology in subcontinental lithosphere

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Abstract

A plausible origin of the seismically observed mid-lithospheric discontinuity (MLD) in the subcontinental lithosphere is mantle metasomatism. The metasomatized mantle is likely to stabilize hydrous phases such as amphiboles. The existing electrical conductivity data on amphiboles vary significantly. The electrical conductivity data on hornblende is much higher than the recent data on tremolite. Thus, if hornblende truly represents the amphibole varieties in MLD regions, then it is likely that amphibole will cause high electrical conductivity anomalies at MLD depths. However, this is inconsistent with the magnetotelluric observations across MLD depths. Hence, to better understand this discrepancy in electrical conductivity data of amphiboles and to evaluate whether MLD could be caused by metasomatism, we determined the electrical conductivity of a natural metasomatized rock sample. The metasomatized rock sample consists of ~87% diopside pyroxene, ~9% sodium-bearing tremolite amphibole, and ~3% albite feldspar. We

22 collected the electrical conductivity data at ~ 3.0 GPa, i.e., the depth relevant to MLD. We also
23 spanned a temperature range between 400 to 1000 K. We found that the electrical conductivity of
24 this metasomatized rock sample increases with temperature. The temperature dependence of the
25 electrical conductivity exhibits two distinct regimes- at low temperatures < 700 K, the electrical
26 conductivity is dominated by the conduction in the solid state. At temperatures > 775 K, the
27 conductivity increases, and it is likely to be dominated by the conduction of aqueous fluids due to
28 partial dehydration. The main distinction between the current study and the prior studies on the
29 electrical conductivity of amphiboles or amphibole-bearing rocks is the sodium (Na) content in
30 amphiboles of the assemblage. And it is likely that the higher Na content in amphiboles leads to
31 the higher electrical conductivity. Pargasite and edenite amphiboles are the most common
32 amphibole varieties in the metasomatized mantle and our study on Na-bearing tremolite is the
33 closest analog of these amphiboles. Comparison of the electrical conductivity results with the
34 magnetotelluric observations limits the amphibole abundance at MLD depths to $< 1.5\%$. Such a
35 low modal proportion of amphiboles could only reduce the shear seismic wave velocity by 0.4-
36 0.5%, which is significantly lower than the observed velocity reduction of 2-6%. Thus, it might be
37 challenging to explain both seismic and magnetotelluric observations at MLD simultaneously.

38

39 **Key Points:** Mid-lithospheric discontinuity (MLD), Electrical conductivity, Metasomatized rock,
40 Diopside, Amphibole

41

42 **Introduction**

43 Recent high-frequency seismic data have revealed a seismic discontinuity in the
44 subcontinental lithosphere at a depth range of 60-160 km. This is often referred to as the mid-
45 lithospheric discontinuity (MLD) and is characterized by a 2-6% reduction in the shear seismic
46 wave velocity (Rychert and Shearer, 2009; Romanowicz, 2009; Abt et al., 2010; Fischer et al.,
47 2010). The thickness of the discontinuity typically varies between 10-20 km (Fischer et al., 2010).

48 Although MLDs are found across all cratons, their origin is poorly understood. A variety
49 of mechanisms have been proposed to explain the observed shear velocity anomaly at MLD depths
50 (Selway et al., 2015; Karato et al., 2015). The most plausible mechanisms include: the presence of
51 partial melts (Thybo, 2006), elastically accommodated grain boundary sliding (Karato et al., 2015;
52 Karato and Park, 2019), seismic anisotropy (Yuan and Romanowicz, 2010; Sodoudi et al., 2013;
53 Wirth and Long, 2014; Ford et al., 2016), and mantle metasomatism or frozen-in melts (Savage
54 and Silver, 2008; Sodoudi et al., 2013; Hansen et al., 2015; Rader et al., 2015, Selway et al., 2015;
55 Saha et al., 2018; Peng and Mookherjee, 2020; Saha et al., 2021).

56 Mantle metasomatism alters the “dry” mantle lithology and often stabilizes carbonates and
57 hydrous minerals such as amphiboles and phlogopite. The xenoliths formed at pressures associated
58 with MLD depths indicate that at least ~25% of these are amphibole-bearing, ~90% are phlogopite-
59 bearing, and less contains carbonates (Rader et al., 2015). While the presence of amphiboles in
60 MLD tends to lower the shear seismic wave velocity (Selway et al., 2015; Peng and Mookherjee,
61 2020; Saha et al., 2021), it may also lead to anomalously high electrical conductivity based on the
62 measurement of hornblendite (Wang et al., 2012). The study showed that when the temperature is
63 higher than ~760-840 K, the electrical conductivity of hornblendite is enhanced due to the
64 dehydration-oxidation reaction, which is about 10^4 - 10^6 higher than that of olivine (Wang et al.,
65 2012). This, however, is contrary to magnetotelluric (MT) observations, since there is no globally

66 recognized anomaly in the electrical conductivity at MLD depths (Meqbel et al., 2014; Yang et al.,
67 2015; Selway, 2019, Karato and Park, 2019). The inconsistency between the MT observations and
68 expected electrical conductivity is one of the biggest criticisms of the mantle metasomatism
69 mechanism (Karato and Park, 2019). However, a recent experimental study found that the
70 electrical conductivity of tremolite amphibole is much lower than previously thought, about 10^3 -
71 10^5 lower than that of hornblende. It only exceeds the conductivity of olivine by one order of
72 magnitude at temperatures <1123 K, i.e., the melting temperature (Shen et al., 2020). Therefore,
73 the role of amphiboles in the electrical conductivity of the mantle is ambiguous and it certainly
74 requires further study.

75 To better understand the effect of hydrous phases on the electrical conductivity of
76 metasomatized lithology, we conducted experiments to constrain the electrical conductivity on a
77 natural metasomatized rock consisting dominantly of pyroxene, amphibole, and feldspar under
78 conditions relevant for MLD, i.e., at ~ 3.0 GPa and ~ 400 - 1000 K. We then used the experimental
79 results to determine the conductivity-depth profiles for the metasomatized cratonic mantle and
80 evaluate whether mantle metasomatism, i.e., metasomatic amphiboles, will cause anomalously
81 high electrical conductivity at MLD depths.

82

83 **Methods**

84 We characterized the mineral phases of the natural rock sample using Raman spectroscopy
85 at ambient conditions (**Figure 1**). To identify the minerals, we used HORIBA LabRAM HR
86 Evolution Raman spectrometer with a Nd:YAG laser ($\lambda = 532$ nm) at the Earth Materials
87 Laboratory, Department of Earth, Ocean, and Atmospheric Sciences, Florida State University,
88 Tallahassee. We also investigated the composition of each mineral phase using electron probe

89 microanalysis (EPMA). We used Cameca SxFive-Tactis at the Laboratoire Magmas et Volcans,
90 Clermont-Ferrand, France, with an accelerating voltage of 15 kV and a steady beam current of 20
91 nA. We also obtained the energy dispersive X-ray spectroscopy (EDS) chemical mapping of the
92 sample using the scanning electron microscope (SEM) with JEOL JSM-5910LV (**Figure S1**). We
93 determined the modal abundances using ImageJ open-source package (Schneider et al., 2012). We
94 found that the sample consists of ~87% pyroxene, ~9% amphibole, and ~3% feldspar (**Table 1**).
95 The stoichiometry of pyroxene is dominated by diopside [(Mg_{1.02}Ca_{0.85}Na_{0.09}Al_{0.06})(Si_{1.99}Al_{0.01})
96 O₆], while amphibole is Na-rich tremolite [(Na_{0.19}K_{0.03})(Ca_{1.61}Na_{0.39})(Mg_{5.09}Fe_{0.02}Ti_{0.01})(Si_{7.75}
97 Al_{0.29})O₂₂(OH)₂], and feldspar is dominantly albite [Na_{1.09}K_{0.01}Al_{1.03}Si_{2.94}O₈] (**Table 1**).

98 We performed the high pressure and temperature electrical conductivity experiment in a
99 1500-ton split-cylinder type multi-anvil apparatus installed at the Laboratoire Magmas et Volcans,
100 Clermont-Ferrand, France. We used an 18/11 assembly, i.e., an edge length of 18 mm for the
101 Cr₂O₃-doped MgO octahedral pressure medium and a truncated edge length of 11 mm for the eight
102 tungsten carbide (WC) anvils. We placed the cylindrical sample specimen in the MgO cylindrical
103 sleeve to electrically insulate the sample from the furnace during the measurements (**Figure S1**).
104 In addition, we placed two nickel (Ni) disks at two ends of the sample. These two Ni disks serve
105 as electrodes and also buffer the oxygen fugacity (f_{O_2}) close to Ni-NiO buffer. We monitored the
106 temperature of the sample using a W₉₅Re₅-W₇₄Re₂₆ thermocouple junction placed at one side of
107 the sample. We placed another W₉₅Re₅ wire at the opposite side of the sample and used two
108 opposing W₉₅Re₅ wires that emerged at the top and bottom of the sample to collect the impedance
109 spectra for the electrical conductivity measurements. We used a 25 μm thick Re foil as the furnace.
110 To enhance effective heating and minimize thermal loss, i.e., to insulate, we also used a ZrO₂
111 sleeve surrounding the Re-furnace (**Figure S1**). To eliminate the influence of absorbed moisture

112 from the atmosphere and exposure to contaminations, we baked the ceramic assembly parts in a
113 high-temperature furnace at 1273 K for >12 hours. We stored all the heat-treated ceramic parts in
114 a high-vacuum furnace (<100 mTorr) at 398 K overnight.

115 For the electrical conductivity measurements, we analyzed the impedance spectroscopy
116 using the ModuLabMTS impedance/gain-phase analyzer over a frequency range of 10^6 - 10^1 Hz.
117 Before measuring the temperature dependence of the electrical conductivity of the sample, first,
118 we held the pressure constant at 3.0 GPa and kept the sample at 500 K for more than 6 hours.
119 While maintaining a constant temperature of 500 K, we measured the electrical resistance of the
120 sample at regular time intervals to ensure that the sample resistance reached a steady value. We
121 noted that the steady-state resistance is about 1 order of magnitude lower than the resistance
122 measured at the very beginning (**Figure 2**). This step is crucial to ensure the removal of the
123 absorbed moisture in the assembly, which could otherwise influence the electrical conductivity
124 measurements at higher temperatures (Manthilake et al., 2015). Then we collected the impedance
125 spectra over a temperature range of 300-1000 K at a step of ~50 K. Below 700 K, we heated and
126 cooled the sample and found that the electrical resistance for the heating and cooling path was
127 reproducible. This minimizes the uncertainty of the electrical conductivity measurements. After
128 this heating and cooling cycle, we collected the spectra in a continuous heating and cooling cycle
129 reaching 1000 K.

130 Following the electrical conductivity measurements, we investigated the cross-section of
131 the experimental run product using EPMA and EDS chemical mapping using SEM. We identified
132 the chemical compositions of each mineral phase after dehydration reaction (**Table 1**).

133 The impedance spectra of a polycrystalline sample can be better understood in terms of a
134 resistor-capacitor (R-C) or resistor-constant phase element (R-CPE) circuit. We obtained the

135 resistance of the polycrystalline sample by fitting the impedance spectra to appropriate RC/R-CPE
136 circuits. We determined the electrical conductivity of the sample by the formulation: $\sigma=L/(RS)$,
137 where σ is the electrical conductivity in “S/m”, R is the resistance in “ Ω ”, L is the length of the
138 cylindrical sample in “m”, and S is the cross-sectional area of the cylindrical sample in “m²”. The
139 sample length and diameter measured before the experiment were ~ 1.7 mm in length and ~ 1.0 mm
140 in diameter. Similar aspect ratios have been successfully used in previous electrical conductivity
141 measurements, particularly when the samples are cored out of natural rocks rather than synthesized
142 using hot-pressed methods from powdered samples (Manthilake et al., 2015, 2016, 2021). After
143 we recovered the sample, we measured the sample length ~ 1.7 mm, i.e., it remained unchanged.
144 This is very likely due to the facts that the sample used in our study is a natural rock rather than a
145 powdered sample, and the experimental conditions were modest ~ 3 GPa which didn’t affect the
146 length of the sample. Since the sample is cut and polished along the length of the cylindrical core,
147 we were unable to directly measure the diameter of the recovered sample. Based on the fact that
148 the length remained unaffected, we used the length and diameter of the sample before the
149 experiments to estimate the electrical conductivity.

150 To ensure that we are measuring the electrical conductivity of the sample and our results
151 are not affected by the sample assembly, we also measured the electrical resistivity of the assembly.
152 The electrical resistivity of the sample is much lower than that of the insulation resistance of the
153 sample assembly (MgO), i.e., the electrical conductivity of the sample is significantly greater than
154 that of the assembly (**Figure S2**).

155

156 **Results**

157 We analyzed the electrical conductivity of the sample using the impedance spectra which
158 are often represented in a Cole-Cole plot with the imaginary part of the impedance plotted in the
159 ordinate and the real part of the impedance plotted in the abscissa. At low temperatures, <700 K,
160 all the spectra display a semicircular arc over the whole frequency range (**Figure S3**). The radius
161 of the semicircular arc is proportional to the electrical resistivity, i.e., inversely proportional to the
162 electrical conductivity. With increasing temperature, the radius of the semicircular arc continues
163 to shrink, indicating that the electrical resistivity reduces or conversely the electrical conductivity
164 increases with temperature. At temperatures >775 K, an additional arc is observed in the low-
165 frequency range (**Figure S3**). The separation of the two arcs possibly indicates two conductive
166 paths, one through the grain interior and the other one along the grain boundary. This is very likely
167 due to the dehydration of the amphibole-bearing rock sample which generates two conductive
168 phases operating in series, i.e., the original solid phase and the aqueous fluid phase interconnected
169 along the grain boundaries.

170 The temperature dependence of the electrical conductivity exhibits a linear relationship
171 before and after dehydration and both could be well described by the Arrhenius relationship:

172
$$\sigma = \sigma_0 \exp\left(\frac{-\Delta H}{RT}\right), \quad (1)$$

173 where σ_0 is a pre-exponential constant, R is the gas constant, and ΔH is the activation enthalpy.
174 The activation enthalpies are respectively 0.188 ± 0.003 eV and 0.48 ± 0.03 eV before and after
175 dehydration (**Figure 2, Table 2**). The increase in the activation enthalpy of the different segments
176 is likely due to the dehydration reaction of the amphibole phase in the sample. The release of
177 aqueous fluids often causes the change of slope i.e., activation enthalpy in the temperature-
178 dependent electrical conductivity of hydrous phases. The activation enthalpy after dehydration for
179 our study, i.e., the T₂ segment between 775 K and 1000 K, is very similar to the activation enthalpy

180 ~0.5 eV in the T₃ segment of the tremolite sample, where the conduction mechanism is dominant
181 by the flow of aqueous fluids (Shen et al., 2020) (**Figure 3, Table 2**).

182 The temperature range where the slope change occurs generally depends on the specific
183 mineral assemblage and its thermal stability (Manthilake et al., 2015, 2016; Pommier and Evans,
184 2017; Pommier et al., 2019). The experimental study on pargasitic-edenitic amphiboles indicates
185 that these amphibole phases are stable till 1373 K (Mandler and Grove, 2016). However, in our
186 study, amphibole starts to dehydrate at ~775 K. An earlier study on the hornblende-actinolite-
187 bearing rock (hornblendite) at 0.5 GPa exhibits two distinct stages of the temperature dependence
188 of electrical conductivity, and the slope changes at ~736-760 K. Similarly, the plagioclase-
189 hornblende-bearing rock (plagioclase hornblendite) also undergoes the discontinuous increase of
190 the electrical conductivity at similar temperatures, i.e., ~797-840 K (**Figure 3, Table 2**) (Wang et
191 al, 2012). It is not surprising that amphiboles show much lower dehydration temperatures in a
192 multi-phase rock. The stability field of amphiboles decreases when competing with other phases,
193 like diopside and albite.

194 We also noted that the electrical conductivity of our diopside-tremolite-albite sample is
195 higher than that of pure clinopyroxene (Yang et al., 2011) or pure tremolite (Shen et al., 2020) by
196 a factor of 10³-10⁴, especially when the temperature is lower than 1000 K (**Figure 3**). These
197 electrical conductivity experiments on clinopyroxene, plagioclase, and amphibole were conducted
198 at different pressures. The effect of pressure on the electrical conductivity is found to be negligible
199 (Yang et al., 2011, 2012; Shen et al., 2020; Hu et al., 2018). Recent experiments on Fe-bearing
200 amphibole indicated that the total Fe content influences the electrical conductivity of amphiboles.
201 Upon heating, the sample loses hydrogen and in the process oxidizes the iron (Fe²⁺ → Fe³⁺), which
202 in turn enhances the electrical conductivity via hopping of electrons (Hu et al., 2018). However,

203 the total Fe content of our sample is as low as ~0.12 wt.% and yet we found that our electrical
204 conductivity results are very high, especially in the solid state (**Figure 3**). This high electrical
205 conductivity cannot be attributed to Fe content. We found that the Na₂O content in our sample,
206 2.24 wt.%, is greater than that of all the amphibole samples examined in earlier electrical
207 conductivity studies, i.e., 0.12 wt.% in tremolite (Shen et al., 2020); ~0.38 wt.% in Fe-bearing
208 amphibole (Hu et al., 2018) and 1.44-1.75 wt.% in amphibole-bearing rocks (Wang et al., 2012).
209 The alkali cation, i.e., Na, occupies the alkali (A) site in the crystal structure of amphiboles and
210 the A site is often vacant in many amphibole end members including tremolite. It is very likely
211 that Na is loosely bound in the large A site in amphiboles. The A sites are connected to each other
212 to form channels along [001] direction that is likely to further facilitate the mobility of the Na⁺
213 ions. Similar high electrical conductivity has been observed in liebermannite which also has
214 tunnels that promote the migration of alkali cations (He et al., 2016; Manthilake et al., 2020). Also,
215 fast ionic conduction in solid state has been observed in albite, where Na⁺ ions undergo faster
216 diffusion via the alkali ion cavities in the albite aluminosilicate framework (Hu et al., 2011). Thus,
217 it is very likely that Na content significantly affects the electrical conductivity of amphiboles
218 (**Figure 3**). When we compare our electrical conductivity data with prior results on amphibole-
219 bearing samples, we found a strong correlation between the electrical conductivity and the Na₂O
220 content, i.e., as the Na₂O content in the amphibole phase of the sample increases, the electrical
221 conductivity is also enhanced (**Figure 3**). The activation enthalpy for our sample at lower
222 temperatures i.e., in the solid-state is also much lower than that of the samples with lower Na
223 content (**Figure 3, Table 2**).

224 The electrical conductivity of major mantle mineral phases (nominally anhydrous mineral
225 (NAM) phases), including olivine (Gardés et al., 2014; Xu et al., 2000; Wang et al., 2006; Yoshino

226 et al., 2006, 2009; Dai et al., 2010; Dai and Karato, 2014a, b; Fei et al., 2020), pyroxene (Dai and
227 Karato, 2009a), and garnet (Dai and Karato, 2009b) is lower than that of amphiboles or amphibole-
228 bearing rocks (**Figure S4**). One exception is that the electrical conductivity of orthopyroxene is
229 slightly higher than that of pure tremolite (Yang et al., 2012; Shen et al., 2020). However, trace
230 amounts of water/hydrogen often have a significant impact on the electrical conductivity, because
231 hydrogen atoms in NAMs occur as defects and are not structurally bonded (Karato, 1990; Wang
232 et al., 2006). For example, the electrical conductivity of garnet with ~50 ppm of water is quite
233 comparable to those of Na-rich amphiboles or amphibole-bearing rocks at mid-lithosphere
234 temperatures. While the electrical conductivity of volumetrically dominant mantle phases at MLD
235 conditions, i.e., olivine and pyroxene with ~50 ppm of water, is still lower than those of Na-rich
236 amphiboles, but comparable with Na-poor amphiboles (**Figure S4**).

237

238 **Discussions**

239 To test whether mantle metasomatism could be a plausible cause for MLD, we selected a
240 natural metasomatized rock sample that consists of ~87% diopside, ~9% tremolite, and ~3% albite.
241 The electrical conductivity of the diopside is similar to that of orthopyroxene (Yang et al., 2011,
242 2012) and slightly higher than that of olivine (Gardés et al., 2014). The metasomatized cratonic
243 mantle often contains <10% amphiboles. To recreate chemistry and modal abundances of mineral
244 phases in the metasomatized mantle, recent experimental studies have demonstrated that 90 wt.%
245 depleted mantle rock and 10 wt.% rhyolitic melts could generate 3.6% to 9.4% amphiboles over
246 2-3 GPa and 1223-1323 K (Saha et al., 2018). Thus, our selected metasomatized sample with ~9%
247 amphibole is a good representative for the metasomatized cratonic mantle rock.

248 Owing to the low modal abundance of amphiboles in the metasomatized mantle, it is very
249 unlikely that they are interconnected. However, amphibole-bearing xenoliths do suggest that
250 amphiboles might be connected via veins (Ionov and Hofmann, 1995). In our experiment, the
251 texture of the starting sample does not show any evidence for the interconnectivity of the
252 amphibole (**Figure S1**). However, the SEM image of the sample after the electrical conductivity
253 measurement shows that tremolite develops a texture that resembles discontinuous and locally
254 interconnected thin veins that are near perpendicular to the electrodes (**Figure 2**). These thin veins
255 are not inherent to the sample and very likely represent quenched aqueous fluids along the grain
256 boundaries.

257 In the earlier electrical conductivity study on tremolite, the sample had little Na. In contrast,
258 our sample assemblage contains Na-bearing tremolite and albite which is more representative of
259 the metasomatized mantle rock in MLD. It is well known that tremolite and albite may form
260 edenite amphiboles at higher pressures (Spear, 1981). Tremolite + tschermakite amphibole + albite
261 could form pargasite amphibole at higher pressures (Bhadra and Bhattacharya, 2007). It is also
262 well known that the most relevant phase of amphiboles stable at MLD conditions is pargasitic-
263 edenitic amphiboles (Mandler and Grove, 2016; Saha et al., 2018), which translates to 3.4-4.4 wt.%
264 Na₂O content in amphiboles (Saha et al., 2018). The even higher Na content in MLD indicates that
265 the electrical conductivity is likely to be similar or higher than our electrical conductivity results.
266 Such a high conductivity is very likely to cause a positive anomaly. This is however not detected
267 by MT observations and thus puts severe constraints on metasomatism as a viable mechanism to
268 explain MLD.

269

270 **Implication**

271 Our experimental electrical conductivity data can provide independent and important
272 constraints on the debated mechanism of MLD, i.e., mantle metasomatism. To better evaluate the
273 electrical conductivity of the metasomatized mantle, we used our results on the temperature
274 dependence of the electrical conductivity of diopside-tremolite-albite rock and extrapolated to
275 conditions relevant for MLD. We calculated the electrical conductivity vs. depth along a typical
276 steady-state conductive geotherm of the cratonic lithosphere, with a surface heat flux of 45 mW/m²
277 (Lee et al., 2011) (**Figure 4**). We extrapolated the electrical conductivity of the solid-state regime
278 to MLD depths. To compare with pristine mantle free from any metasomatism, we determined
279 electrical conductivity-depth profiles using a depleted peridotite composition consisting of ~48.5%
280 olivine, ~41.5% orthopyroxene, ~9.1% garnet, and ~0.9% clinopyroxene. This mantle rock model
281 is based on the average composition of the global cratonic xenoliths (Saha et al., 2018; Lee et al.,
282 2011; Luguet et al., 2015; Maier et al., 2012; Pearson and Wittig, 2013). We also considered the
283 effect of enhanced electrical conductivity due to hydrogen defects in NAMs. As we all know,
284 cratons are stable which indicates cratons are rheologically stiff and often dry so that they resist
285 being swept away by mantle convection. However, recent studies have shown that most of the
286 cratonic lithospheric mantle is not as dry as assumed and can contain a small amount of water in
287 NAMs such that it will not affect the overall rheological strength of the craton, i.e., it can sustain
288 over geological times (Selway, 2019; Demouchy and Bolfan-Casanova, 2016; Griffin et al., 2009;
289 Saal et al., 2002; Simons et al., 2002; Freitas and Manthilake, 2019). Thus, we considered two
290 distinct cases for the unmetasomatized “background” mantle- (a) major mantle minerals are “dry”
291 and (b) major mantle minerals are “wet” with an aggregate water content of 50 ppm (Saal et al.,
292 2002; Simons et al., 2002; Freitas and Manthilake, 2019). We also considered the partition
293 coefficients of water among these major mantle phases from earlier studies for the “wet” model

294 (Ardia et al., 2012; Férot and Bolfan-Casanova, 2012; Demouchy and Bolfan-Casanova, 2016;
295 Mookherjee and Karato, 2010).

296 To estimate the electrical conductivity of the unmetasomatized “background” mantle, we
297 used the generalized Archie’s Law (Glover, 2010):

$$298 \quad \sigma = \sum_i^n \sigma_i \phi_i^{m_i}, \quad (2)$$

299 where σ_i is the electrical conductivity of the i^{th} mineral phase ($i=1,2,\dots,n$), ϕ_i is the modal
300 abundance of the i^{th} mineral phase, and m_i is the connectivity exponent of the i^{th} mineral phase
301 which is inversely related to the physical connectivity of mineral phase. We used the connectivity
302 exponent $m_i= 1.3$ for the i^{th} phase comprising $\geq 20\%$ of the whole rock, $m_i= 2$ for the phase
303 comprising 10-20%, and $m_i= 3$ for the phase comprising $<10\%$ (Selway, 2019). For an ideal
304 connection, $m= 1$.

305 We calculated the electrical conductivity of two distinct lithologies by combining the
306 electrical conductivity of metasomatized rock, i.e., our study, with (a) “dry background” mantle
307 and (b) “wet background” mantle. We compared the predicted electrical conductivity-depth
308 profiles with available MT data (Yang et al., 2015) near two distinct seismic stations JFWS and
309 EYMN located in the northern US (Selway, 2019). These two stations have reported MLDs at
310 depths of 59 km and 94 km, respectively (Abt et al., 2010). To explain the MT observations, we
311 varied the proportion of the depleted peridotite rock and the metasomatized rock.

312 We found that at the MLD depth, i.e., ~60-95 km in the northern US, the conductivity of
313 our metasomatized rock is 3.1-4.6 orders of magnitude greater than the “dry” depleted mantle and
314 1.6-2.4 orders of magnitude greater than the “wet” depleted mantle with 50 ppm of water (**Figure**
315 **4**). The difference between the electrical conductivity of the “dry” and “wet” depleted mantle is
316 due to the effect of extrinsic hydrogen defects. We also found that if the sub cratonic mantle is

317 “dry”, the lower bound of the observed MT profile could be explained by ~7% of partial
318 metasomatism. This means ~0.6% amphiboles in the bulk rock. While the upper bound could be
319 explained by partial metasomatism not exceeding 17%, which is equivalent to ~1.5% amphibole
320 in the bulk rock. In the “wet” mantle, we noted that without the effect of mantle metasomatism, it
321 is already sufficient to explain the observed MT profiles. And the upper bound of the MT profiles
322 could be explained <14% of partial metasomatism, i.e., <1.3% amphibole in the bulk rock (**Figure**
323 **4**). However, ~1.5% amphibole in the bulk rock could only reduce the shear seismic wave velocity
324 by ~0.4-0.5% at MLD depths (Saha et al., 2021; Peng and Mookherjee, 2020), while a 2-6%
325 reduction in the shear seismic wave velocity has been observed in most MLD regions (Rychert
326 and Shearer, 2009; Romanowicz, 2009; Abt et al., 2010; Fischer et al., 2010). Hence, our results
327 do indicate that irrespective of whether the sub-cratonic mantle is “dry” or “wet”, the presence of
328 Na-rich amphibole-bearing rock, or mantle metasomatism cannot explain the shear seismic wave
329 velocity and electrical conductivity of MLD simultaneously.

330

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338

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546

547 **Figures caption**

548 **Figure 1.** Raman spectrum of the natural metasomatized rock sample containing diopside,
549 tremolite, and albite (Di-Tr-Ab) (red) at ambient conditions. It is compared with the reference
550 Raman spectra for diopside (Di) (RRUFF ID: R040109; black), tremolite (Tr) (RRUFF ID:
551 R040009; blue), and albite (Ab) (RRUFF ID: R040068; green) (Lafuente et al., 2015).

552

553 **Figure 2. (a)** Electrical conductivity of diopside-tremolite-albite sample as a function of reciprocal
554 temperature. The sample was subjected to several heating-cooling cycles. The lines represent
555 Arrhenius fits to the data, except for the first heating-cooling cycle. The colored numbers indicate
556 the activation enthalpy associated with each regime, i.e., solid-state conduction and partial
557 dehydration. **(b)** SEM image of the sample after the electrical conductivity experiment. The
558 mineral phases are labeled. Locally interconnected thin bands of tremolite phase are noted.
559 Abbreviation: Di- diopside, Tr- tremolite, Ab- albite, and Fls- fluids.

560

561 **Figure 3.** Plot of electrical conductivity of the diopside-tremolite-albite (Di-Tr-Ab) sample as a
562 function of reciprocal temperature. The colored numbers indicate the Na₂O content in the
563 amphibole phase of the samples and the total FeO content of the samples. Note: the filled red
564 symbols- data from this study; the bold red lines- Arrhenius fit to data from this study; the dark
565 blue line- hornblende-actinolite-bearing hornblendite (Hbl-Act) (W12: Wang et al., 2012); the
566 intermediate blue line- plagioclase-hornblende-bearing hornblendite parallel to the lineation
567 within the sample (Pl-Hbl//); the light blue line- plagioclase-hornblende-bearing hornblendite
568 perpendicular to the lineation within the sample (Pl-Hbl⊥); the orange line- clinopyroxene (Cpx)
569 (Y11: Yang et al., 2011); the purple line- tremolite (Tr) (S20: Shen et al., 2020); and the green
570 line- amphibole (Amp) (H18: Hu et al., 2018). *We note an unreasonable high CaO content ~20.43
571 wt.% in the amphibole sample (Hu et al., 2018). This might indicate that the amphibole sample is
572 not representative of the lithosphere. The relevant CaO content for amphiboles that are likely to
573 be stable in the metasomatized mantle ranges between 9.2-12.6 wt.% (Saha et al., 2021).

574

575 **Figure 4.** The conductivity-depth profile of the diopside-tremolite-albite sample (Di-Tr-Ab)
576 (green lines) and predicted conductivity-depth profiles of the (a) “dry” and (b) “wet” (50 ppm of
577 water) depleted peridotite (DP) based on the generalized Archie’s Law. The lighter red and blue
578 lines are the bulk conductivity of the mixed model. For example, DP+7% MR means the bulk rock
579 consists of 93% depleted peridotite (DP) and 7% of our metasomatized rock (MR), i.e., the
580 diopside-tremolite-albite sample. The dashed lines indicate the conductivity-depth profiles from
581 the three-dimensional MT models near seismic stations JFWS and EYMN in the northern US
582 (Yang et al., 2015). The cyan shaded area shows the depth range of MLDs reported at the two
583 seismic stations (Abt et al., 2010).

Table 1. Chemical composition (wt.%) of mineral phases in the sample assemblage determined by EPMA before and after the electrical conductivity measurements. The chemical composition of each phase is slightly modified after the experiment.

Sample	SiO ₂	MgO	CaO	Al ₂ O ₃	Na ₂ O	K ₂ O	FeO	TiO ₂	Cr ₂ O ₃	NiO	MnO	Total	Modal abundance
Before													
Di	$(\text{Mg}_{1.02}\text{Ca}_{0.85}\text{Na}_{0.09}\text{Al}_{0.06})(\text{Si}_{1.99}\text{Al}_{0.01})\text{O}_6$												87.5%
	55.58	19.10	22.03	1.62	1.25	0.00	0.12	0.02	0.04	0.09	-	99.86	
Tr	$(\text{Na}_{0.19}\text{K}_{0.03})(\text{Ca}_{1.61}\text{Na}_{0.39})(\text{Mg}_{5.09}\text{Fe}_{0.02}\text{Ti}_{0.01})(\text{Si}_{7.75}\text{Al}_{0.29})\text{O}_{22}(\text{OH})_2$												9.1%
	58.27	25.68	11.28	1.83	2.24	0.17	0.17	0.14	0.04	0.04	-	99.87	
Ab	$(\text{Na}_{1.09}\text{K}_{0.01})\text{Al}_{1.03}\text{Si}_{2.94}\text{O}_8$												3.4%
	66.70	0.37	0.07	19.77	12.79	0.11	0.04	0.03	0.02	0.09	-	99.97	
After													
Di	$(\text{Ca}_{0.96}\text{Na}_{0.04})(\text{Mg}_{0.95}\text{Al}_{0.04})\text{Si}_{2.00}\text{O}_6$												87.2%
	55.94	17.76	24.95	0.87	0.54	0.00	0.18	0.03	0.04	-	0.17	100.49	
Tr	$(\text{Na}_{0.38}\text{K}_{0.05})(\text{Ca}_{1.73}\text{Na}_{0.27})(\text{Mg}_{4.69}\text{Al}_{0.17}\text{Ti}_{0.06}\text{Fe}_{0.03}\text{Mn}_{0.02})(\text{Si}_{7.61}\text{Al}_{0.39})\text{O}_{22}(\text{OH})_2$												9.1%
	57.15	23.24	12.29	2.50	1.69	0.19	0.26	0.34	0.03	-	0.18	97.89	
Ab	$(\text{Na}_{0.96}\text{K}_{0.01}\text{Mg}_{0.01}\text{Ca}_{0.01})\text{Al}_{1.01}\text{Si}_{2.99}\text{O}_8$												3.4%
	69.32	0.03	0.32	19.88	11.41	0.16	0.01	0.00	0.00	-	0.01	101.15	
Loss													0.3%

Note: abbreviation: Di- diopside, Tr- tremolite, and Ab- albite.

Table 2. Arrhenius fit parameters of the diopside-tremolite sample in this study and previous studies with experimental pressure and temperature conditions. The numbers in parentheses is the error of the last digit with one standard deviation.

Mineral phase	P (GPa)	T_1 (K)	$\text{Log } \sigma_1$ (S/m)	ΔH_1 (eV)	T_2 (K)	$\text{Log } \sigma_2$ (S/m)	ΔH_2 (eV)	T_3 (K)	$\text{Log } \sigma_3$ (S/m)	ΔH_3 (eV)	T_4 (K)	$\text{Log } \sigma_4$ (S/m)	ΔH_4 (eV)	Reference
Di-Tr-Ab	3.0	450-700	-0.59(3)	0.188(3)	775-1000	1.4(2)	0.48(3)	-	-	-	-	-	-	This study
Hbl-Act	0.5	420-736	2.03(9)	0.69(2)	760-824	24(2)	3.9(4)	-	-	-	-	-	-	Wang et al., 2012
Pl+Hbl(\perp)	1.0	443-824	1.18(7)	0.68(1)	840-900	18.0(7)	3.5(2)	-	-	-	-	-	-	Wang et al., 2012
Pl+Hbl(\parallel)	1.0	395-771	1.49(7)	0.66(1)	797-857	18(2)	3.3(4)	-	-	-	-	-	-	Wang et al., 2012
Tr	2.0	648-898	-0.8(2)	0.75(3)	923-998	5.0(9)	1.7(2)	1023-1123	-1(1)	0.5(2)	1148-1373	14.2(6)	3.8(2)	Shen et al., 2020
Cpx	1.2	773-1273	2.2(3)	1.06(5)	-	-	-	-	-	-	-	-	-	Yang et al., 2011
Pl	1.2	773-1273	4.1(3)	1.67(6)	-	-	-	-	-	-	-	-	-	Yang et al., 2012
Amp*	2.0	623-873	2.08	0.81	923-1173	1.88	0.68	-	-	-	-	-	-	Hu et al., 2018

Note: abbreviation: Di- diopside, Tr- tremolite, Ab- albite, Hbl- hornblende, Act- actinolite, Pl- plagioclase, Cpx- clinopyroxene, and Amp- amphibole. *We note an unreasonable high CaO content ~ 20.43 wt.% in the amphibole sample (Hu et al., 2018). This might indicate that the amphibole sample is not representative of the lithosphere. The relevant CaO content for amphiboles that are likely to be stable in the metasomatized mantle ranges between 9.2-12.6 wt.% (Saha et al., 2021).

Intensity (a.u.)

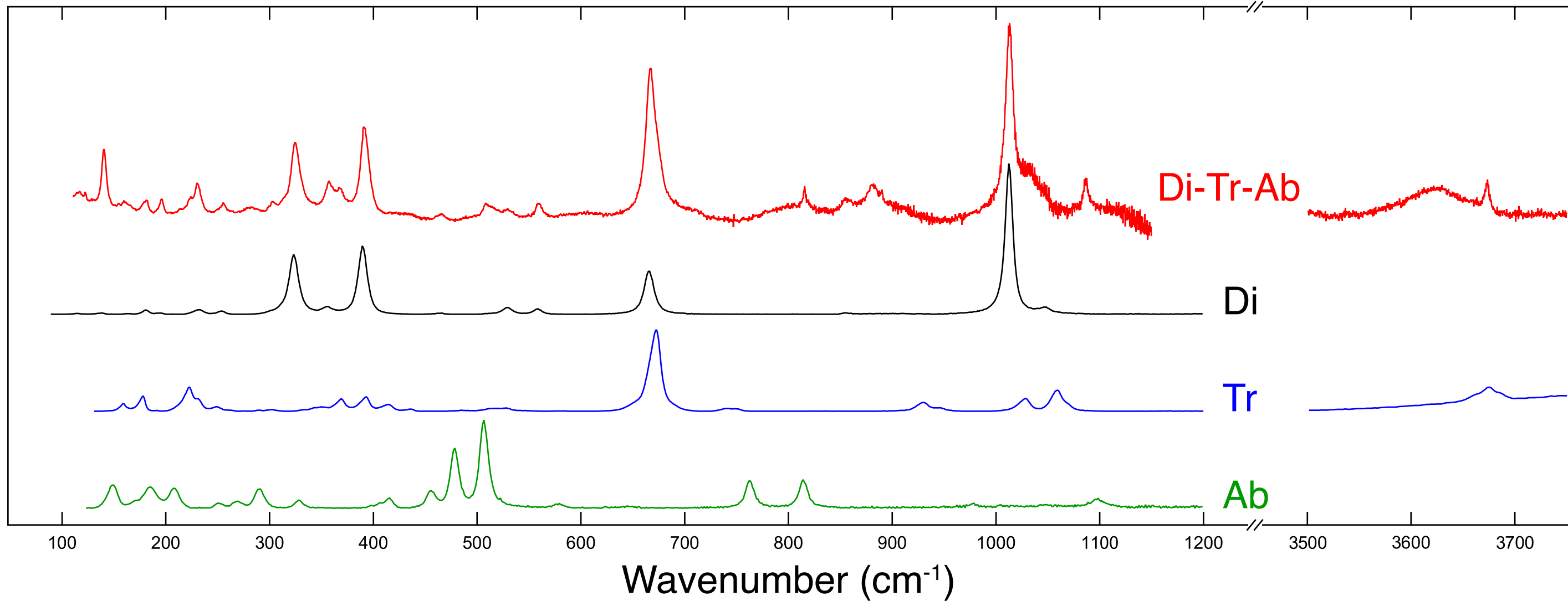


Figure 1

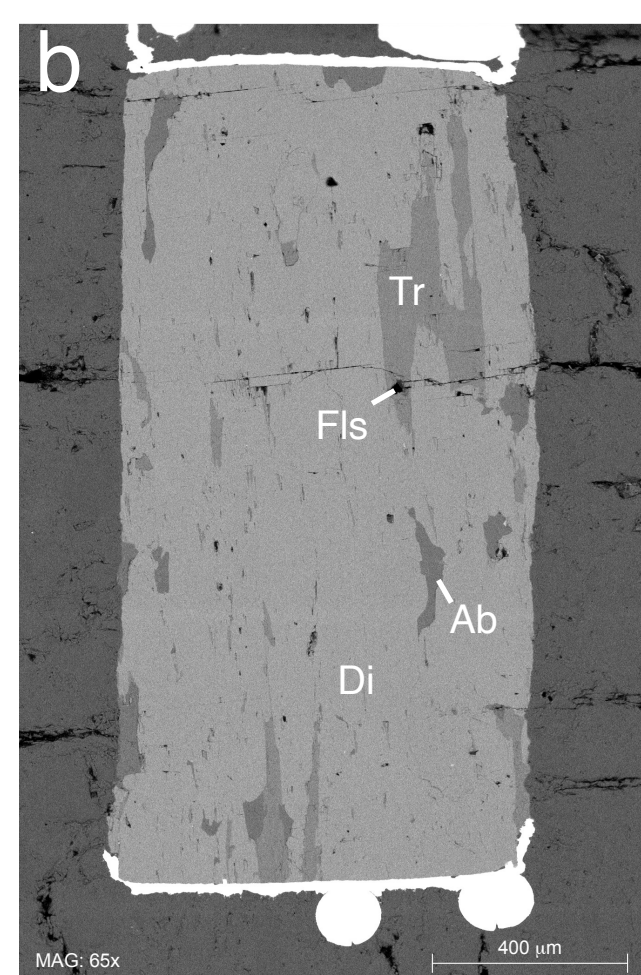
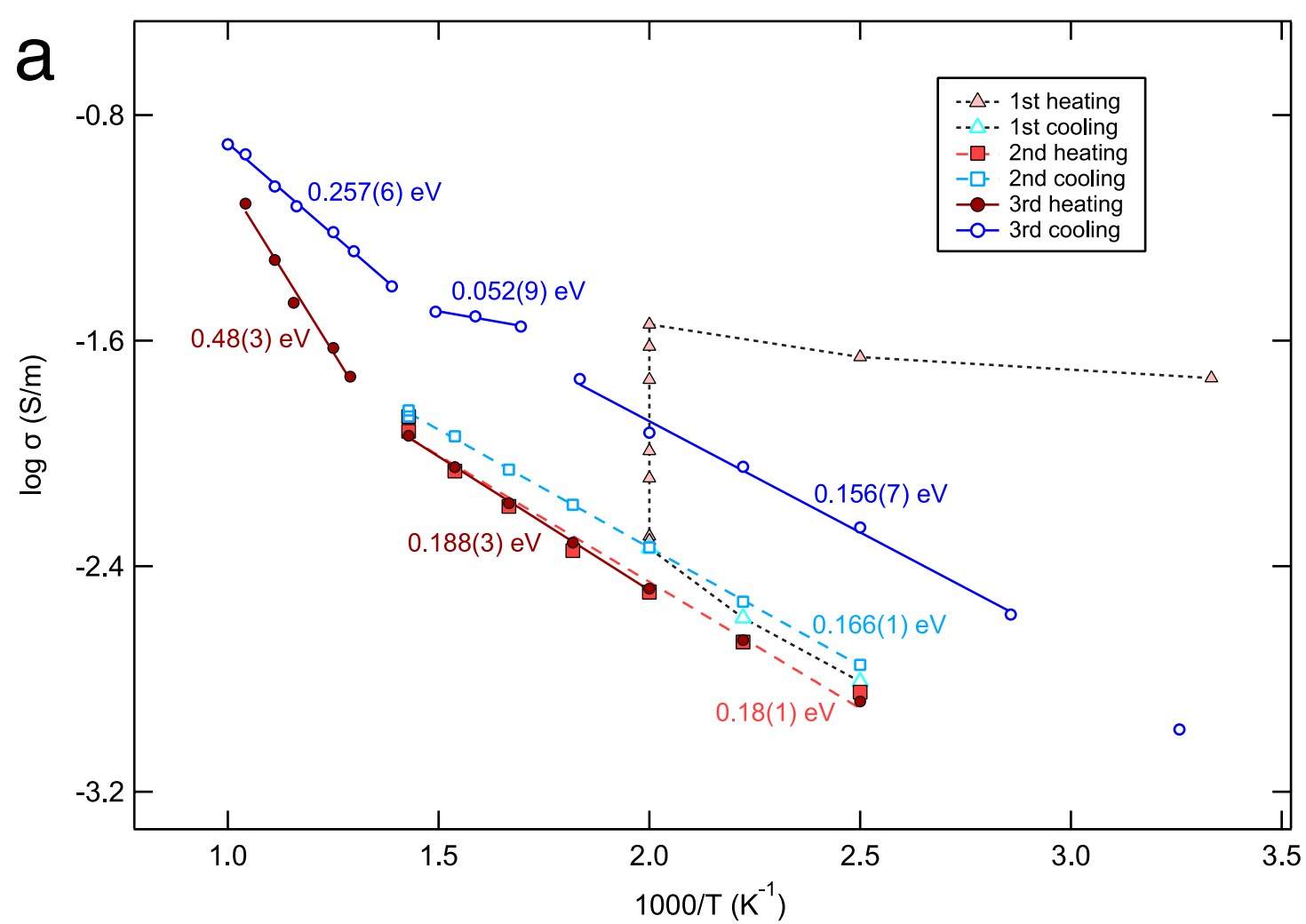
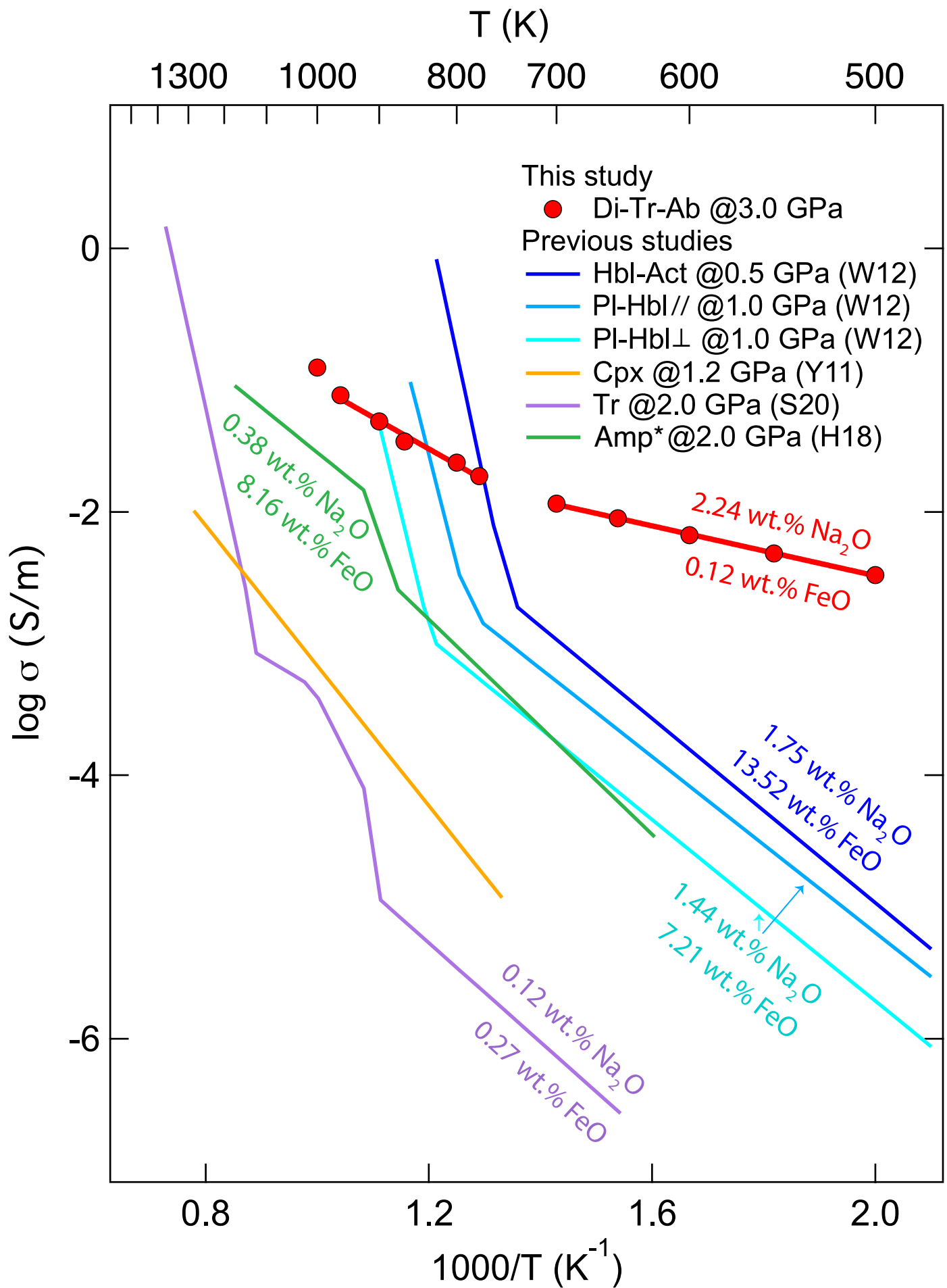


Figure 2



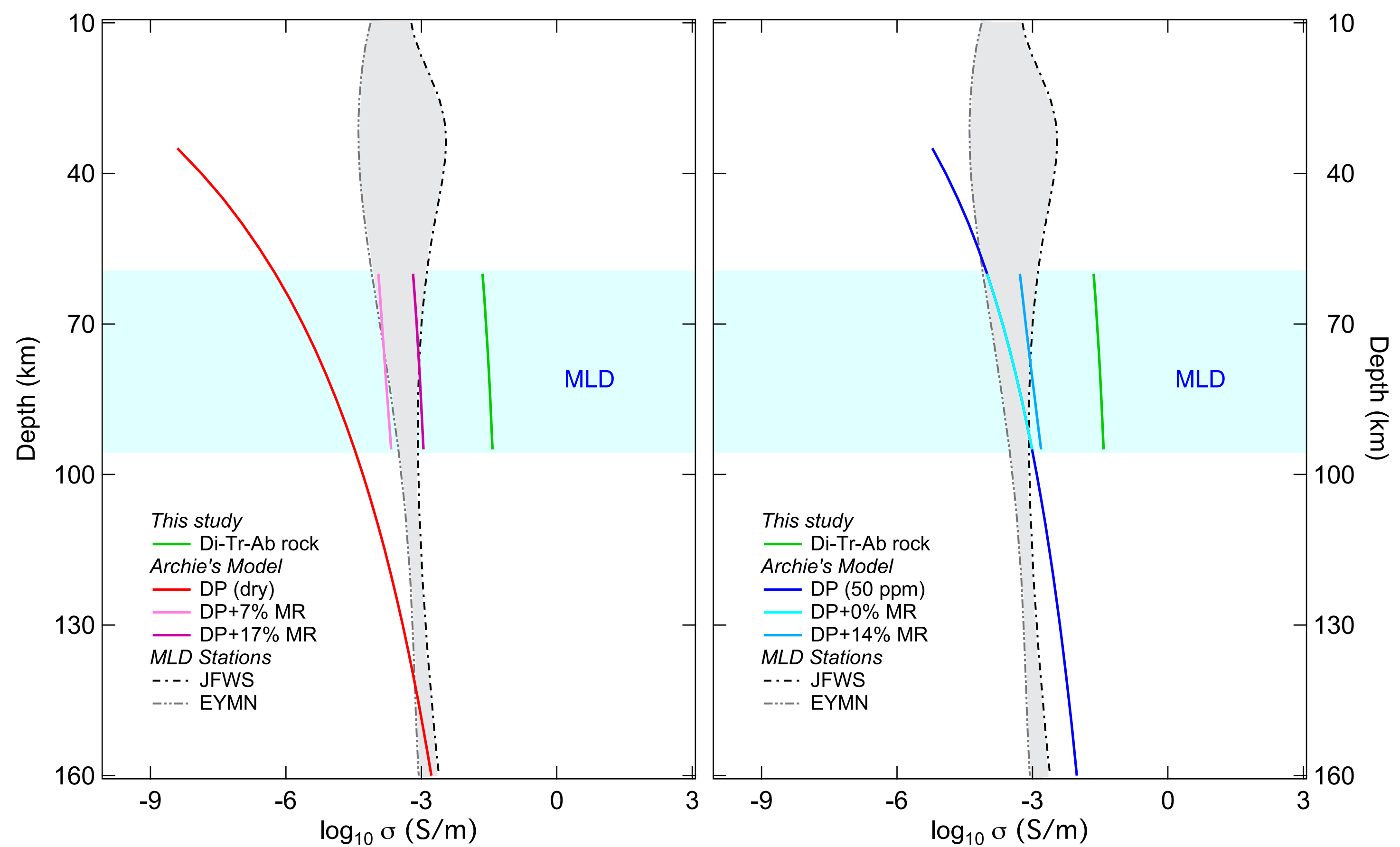


Figure 4

Supporting Information

Electrical conductivity of metasomatized lithology in subcontinental lithosphere

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Figures S1 to S4

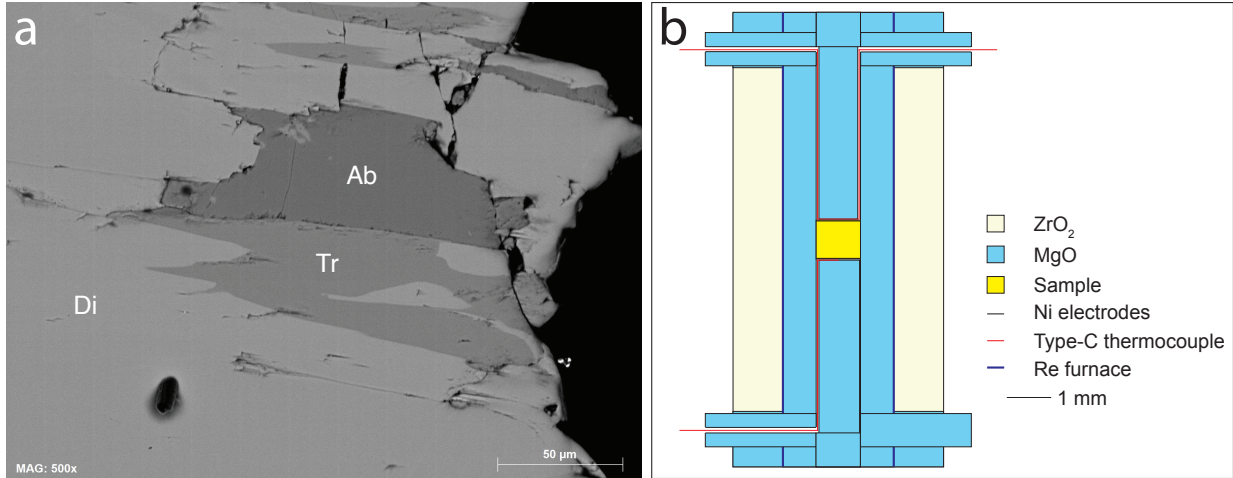


Figure S1. (a) SEM image of the diopside-tremolite-albite sample before the experiment. The mineral phases are labeled. Abbreviation: Di- diopside, Tr- tremolite, and Ab- albite. (b) Cross-section of the sample assembly for high-pressure and high-temperature electrical conductivity measurements.

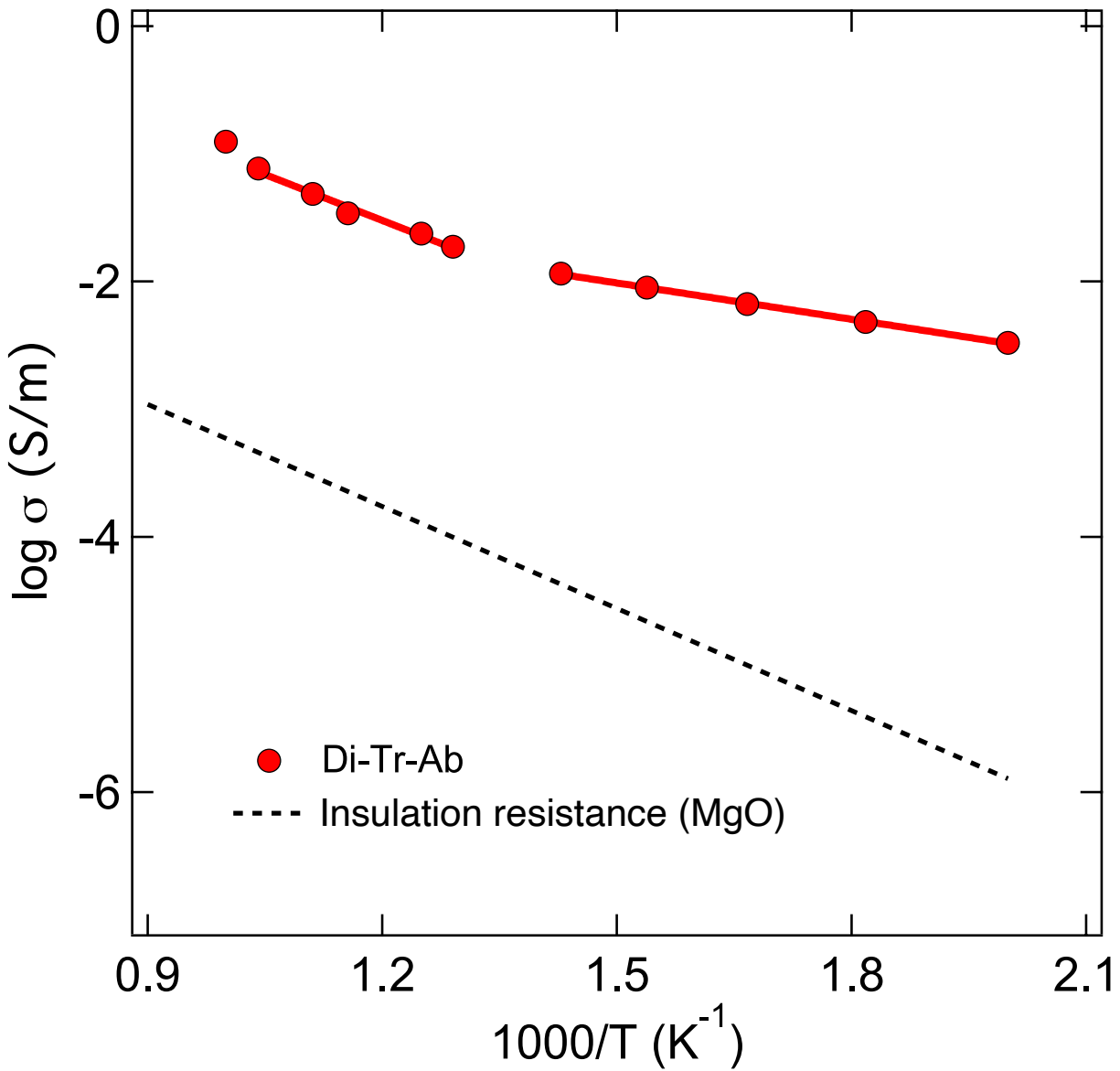


Figure S2. Comparison between the electrical conductivity of the diopside-tremolite-albite (Di-Tr-Ab) sample and the conductivity (insulation resistance) of the assembly (MgO) as a function of reciprocal temperature.

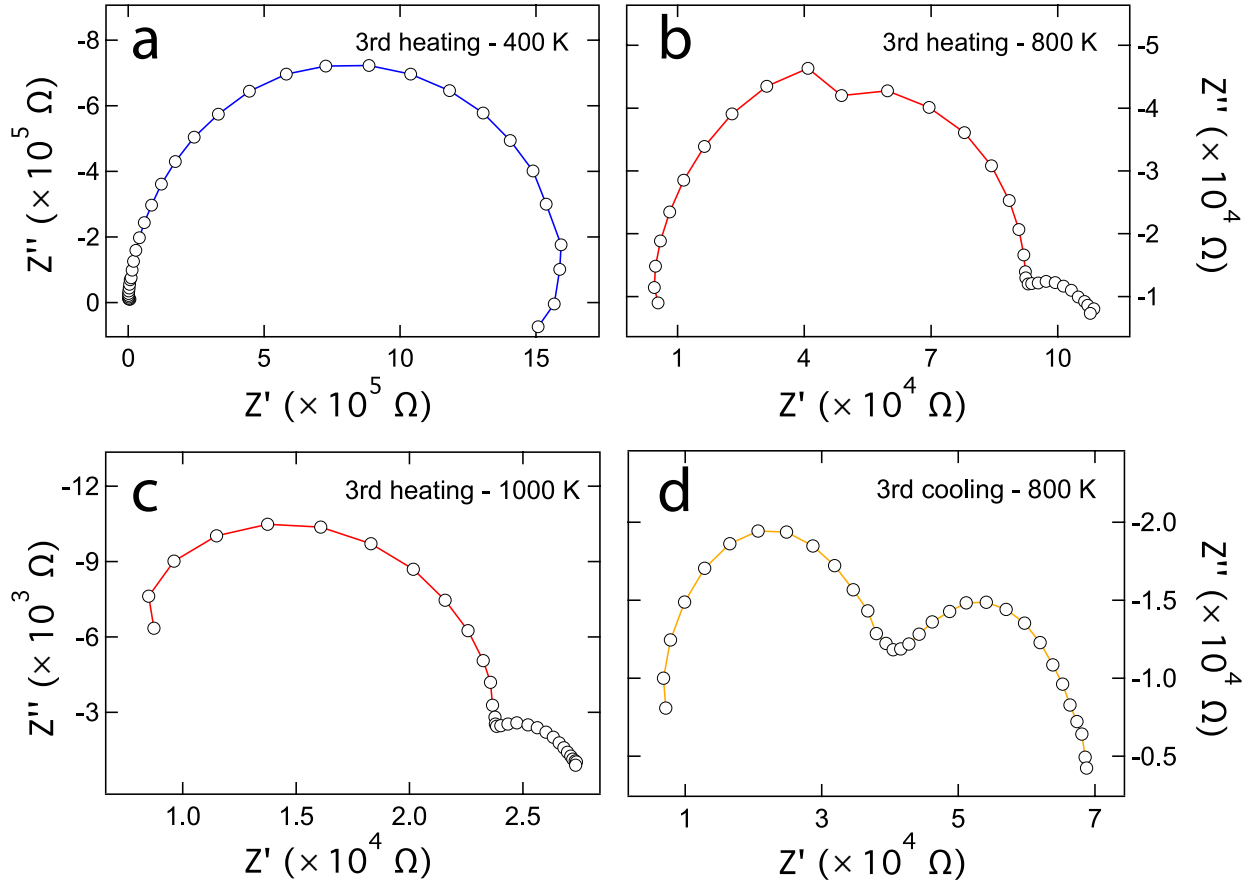


Figure S3. Cole-Cole plot for the diopside-tremolite-albite sample at 3.0 GPa. The axis represents the real (Z') and imaginary part (Z'') of the complex impedance. **(a)** The sample is in the solid state at 400 K of the 3rd heating path. **(b-c)** The radius of the arc continues to shrink and an additional arc is observed in the low-frequency range at 800 K and 1000 K of the 3rd heating path. **(d)** The separation of the two arcs is very clear at 800 K of the 3rd cooling path. The separation of the two arcs in panel **(b-d)** possibly indicates two conductive paths, one through the grain interior and the other one along the grain boundary. This probably reflects the dehydration of the amphibole-bearing rock sample which generates two conductive phases at high temperatures, i.e., the original solid phase and the aqueous fluid phase interconnected along the grain boundaries.

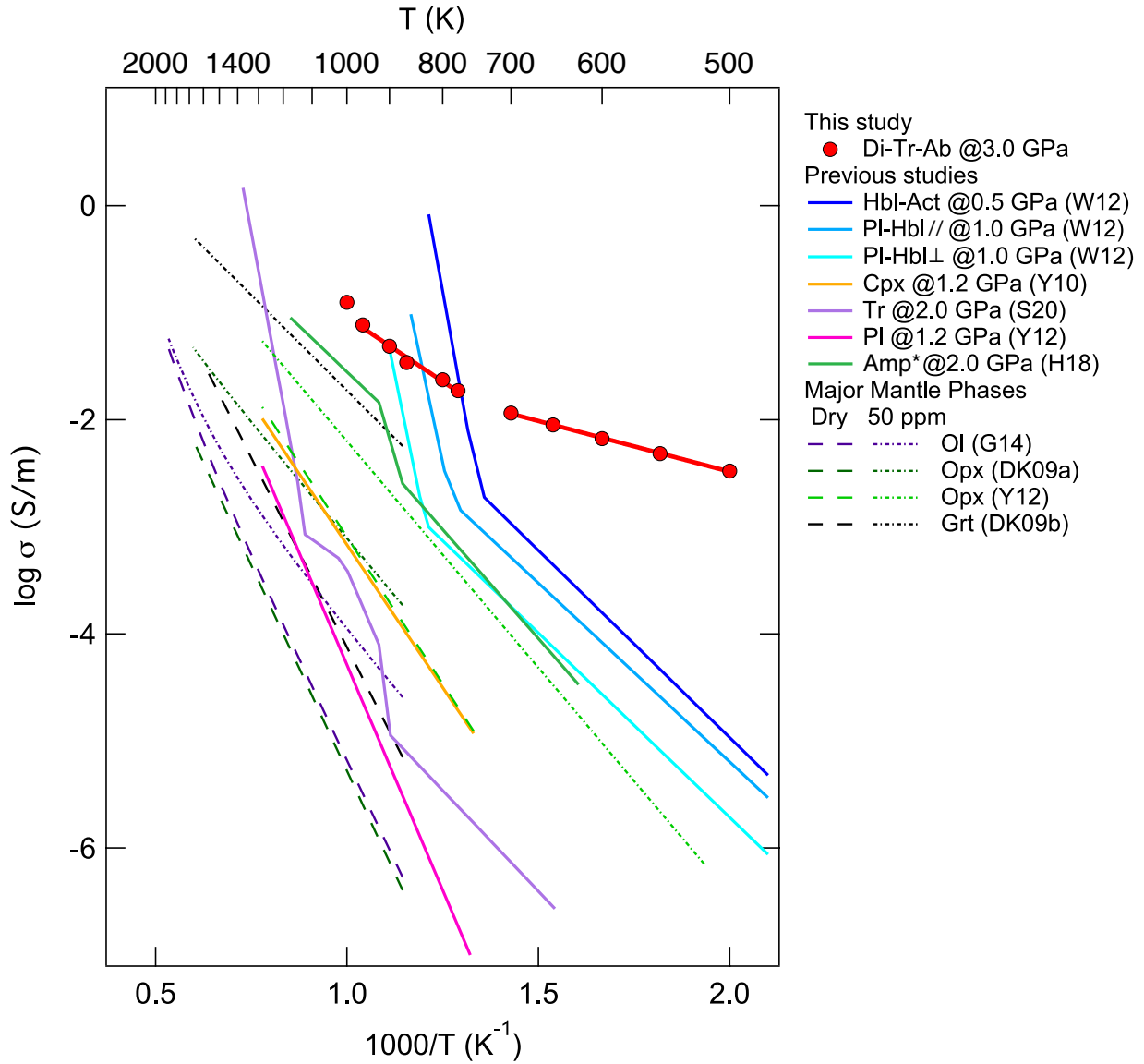


Figure S4. Plot of electrical conductivity of the diopside-tremolite-albite (Di-Tr-Ab) sample as a function of reciprocal temperature. Note: the filled red symbols- data from this study; the bold red lines- Arrhenius fit to data from this study; the dark blue line- hornblende-actinolite-bearing hornblendite (Hbl-Act) (W12: Wang et al., 2012); the intermediate blue line- plagioclase-hornblende-bearing hornblendite parallel to the lineation within the sample (PI-Hbl//); the light blue line- plagioclase-hornblende-bearing hornblendite perpendicular to the lineation within the sample (PI-Hbl⊥); the orange line- clinopyroxene (Cpx) (Y11: Yang et al., 2011); the purple line-

tremolite (Tr) (S20: Shen et al., 2020); the magenta line- plagioclase (Pl) (Y12: Yang et al., 2012); the green line- amphibole (Amp*) (H18: Hu et al., 2018); the thin dashed and dotted lines represent “dry” NAMs and “wet” NAMs with 50 ppm water, respectively: olivine (Ol) (G14: Gardés et al., 2014), orthopyroxene (Opx) (DK09a: Dai and Karato, 2009a; Y12: Yang et al., 2012), and garnet (Grt) (DK09b: Dai and Karato, 2009b).