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**Experimental Investigation of the Effect of Nickel on the Electrical Resistivity  
of Fe-Ni and Fe-Ni-S Alloys under Pressure**

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24 **Abstract**

25 Electrical resistivity experiments were conducted on three alloys in the iron-rich side of the Fe-  
26 Ni(-S) system (Fe-5 wt.% Ni, Fe-10 wt.% Ni, Fe-10 wt.% Ni-5 wt.% S) at 4.5 and 8 GPa and up  
27 to 1900 K using the multi-anvil apparatus and the 4-electrode technique. For all samples,  
28 increasing temperature increases resistivity, and the effect of pressure on resistivity is less  
29 important than the effect of temperature. At defined temperature, Fe-Ni(-S) alloys are more  
30 resistive than Fe by a factor of about 3. Fe-Ni alloys containing 5 and 10 wt.% Ni present  
31 comparable electrical resistivity values. The resistivity of Fe-Ni(-S) alloys is comparable to the  
32 one of Fe – 5 wt.% S at 4.5 GPa and is about 3 times higher than the resistivity of Fe – 5 wt.% S  
33 at 8 GPa, due to a different pressure dependence of electrical resistivity between Fe-Ni and Fe-S  
34 alloys. Based on these electrical results and experimentally-determined thermal conductivity  
35 values from the literature, lower and upper bounds of thermal conductivity were calculated. For  
36 all Ni-bearing alloys, thermal conductivity estimates range between ~12 and 20 W/m.K over the  
37 considered pressure and temperature ranges. Adiabatic heat fluxes were computed for both  
38 Ganymede's core and the Lunar core, and heat flux values suggest a significant dependence to  
39 both core composition and the adiabatic temperature. Comparison with previous thermochemical  
40 models of the cores of Ganymede and the Moon suggest that some studies may have overestimated  
41 the thermal conductivity and hence, the heat flux along the adiabat in these planetary cores.

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44 **Keywords:** iron-nickel alloys, metallic cores, electrical resistivity, multi-anvil apparatus,  
45 Ganymede, the Moon.

46

## 47 INTRODUCTION

48 The metallic cores of terrestrial planets and moons are composed of iron-nickel alloy that  
49 contains different amounts of light elements (such as S, Si, and C). Several wt.% of nickel is  
50 thought to be present in these planetary cores, based on mass balance calculations and on iron-rich  
51 meteorites geochemistry (e.g., McDonough and Sun, 1995; Jarosewich, 1990). For instance,  
52 geochemical models have suggested that about 5.5 wt.% Ni is present in the Earth's core (e.g.,  
53 McDonough and Sun, 1995), and an estimate of about 9 wt.% Ni in the Lunar core has been  
54 suggested by Righter et al. (2017), assuming a bulk Moon Ni content of 2200 ppm, a core fraction  
55 of 2 mass % and using calculations of the metal-silicate partition coefficient of Ni. Meteorite  
56 geochemistry has indicated that the Martian core also likely contains several wt.% Ni, with Fe, Ni,  
57 and S possibly representing the major components of the planet's core (e.g., Wänke and Dreibus,  
58 1988; Lodders and Fegley, 1997).

59 Several investigations have been conducted to understand the effect of nickel on the  
60 chemical and physical properties of iron alloys, and therefore to determine the influence of Ni on  
61 the structure and dynamics of metallic cores. The effect of nickel on the phase diagram of iron is  
62 detectable but small (e.g., Lin et al., 2002) and in particular, Ni stabilizes the face-centered cubic  
63 (fcc) structure under high pressure and temperature (e.g., Côté et al., 2012). It was proposed that  
64 Ni does not affect significantly the melting curve of the Fe-rich side of the Fe-S system at the core  
65 conditions in small terrestrial bodies (Stewart et al., 2007), though it might be more significant at  
66 high pressure relevant to the Earth's core (Komabayashi et al., 2019). This suggests that nickel,  
67 contrary to light-alloying components such as sulfur, is unlikely to affect the onset of core  
68 crystallization of small planets and moons. Nickel may have affected the partitioning behavior of  
69 heavy iron isotopes during core formation (Elardo and Shahar, 2017), but no significant effect of

70 nickel concentration on the partitioning of siderophile elements has been measured under Earth's  
71 core conditions (Ni, Cr, V; Huang and Badro, 2017). Experimental studies of Fe-Ni alloys under  
72 pressure have demonstrated that Ni has a very small effect on several material properties, such as  
73 density, sound velocity and compressibility (e.g., Mao et al., 1990; Lin et al., 2003; Kantor et al.,  
74 2007; Martorell et al., 2013, 2015; Kawaguchi et al., 2017; Wakamatsu et al., 2018; Morrison et  
75 al., 2019), justifying the use of Ni-free iron alloys as core analogues in mineral physics  
76 experiments.

77         The investigation of core dynamics requires constraining the superadiabatic heat flux, i.e.,  
78 the heat that is available to drive convection, which depends strongly on the thermal resistivity of  
79 the core materials. Measurements of the thermal resistivity of Fe-Ni alloys under high temperature  
80 are scarce, but at atmospheric pressure and high temperature (>1673 K), experiments have  
81 suggested that Ni does not affect significantly the thermal resistivity of Fe-Ni melts (Watanabe et  
82 al., 2019). Lower bound estimates of thermal resistivity of Fe-Ni alloys can also be obtained from  
83 electrical resistivity measurements (Watanabe et al., 2019). The electrical resistivity of Fe-Ni  
84 alloys has been well documented at atmospheric pressure, with the effect of temperature and  
85 composition being investigated systematically (e.g., Ho et al., 1983 and references therein; Kita  
86 and Morita, 1984). At defined temperature, resistivity increases with the nickel content, and at  
87 high temperature, the electrical resistivity of molten alloys presents a linear temperature  
88 dependence. The effect of nickel content on iron resistivity was found to be significant both in the  
89 solid and the liquid state in these studies, and to be higher at low temperature than at high  
90 temperature. Electrical resistivity is also a subtle probe of the Invar region, of the martensitic and  
91 austenitic metastable states, and of the magnetic transition across the Curie temperature in Fe-Ni  
92 alloys (e.g., Ho et al., 1983 and references therein). Under high pressure (up to 70 GPa) and at

93 room temperature, electrical resistivity measurements in the iron-rich portion of the Fe-Ni system  
94 (up to 15 wt.% Ni) have highlighted a non-negligible effect of Ni on the electrical properties of  
95 iron alloys, with nickel increasing the alloy resistivity (Gomi and Hirose, 2015). With increasing  
96 pressure, an increase in resistivity by a factor of  $\sim 2$  followed by a drop in resistivity upon the body-  
97 centered cubic (bcc) to hexagonal-close-packed (hcp) structure transition was observed, which was  
98 also reported by Kuznetsov et al. (2007) at 425 K and  $\sim 8$  GPa. Based on the extrapolation of their  
99 data at room temperature to high temperatures, Gomi and Hirose (2015) suggested that the effect  
100 of Ni on resistivity might become negligible under Earth's core conditions, due to resistivity  
101 "saturation" (Gomi et al., 2013; Pozzo and Alfè, 2016; Wagle et al., 2019). This saturation depends  
102 on the mean free path of the electrons, which directly depends on temperature (thermal saturation)  
103 and can be enhanced by the presence of impurities (chemical saturation).

104         These previous thermal and electrical resistivity studies of Fe-Ni alloys have been  
105 conducted at pressure and temperature conditions different from the ones of the cores in small  
106 planets and moons (Figure 1), raising the question of the effect of nickel on the core cooling  
107 processes of Mars-sized or smaller bodies. Here the effect of Ni (up to 10 wt.%) on the electrical  
108 resistivity of Fe-Ni(+/-)S samples has been investigated experimentally up to 8 GPa and  $\sim 1900$  K.  
109 Based on the electrical resistivity results, thermal conductivity is calculated and used to compute  
110 adiabatic heat fluxes in the cores of Ganymede and the Moon.

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### 113 **ELECTRICAL EXPERIMENTS AND ANALYSES**

114         Electrical experiments were performed on three compositions: Fe - 5 wt.% Ni, Fe - 10 wt.%  
115 Ni, and Fe - 5 wt.% Ni - 3wt.% S (referred to as Fe-5Ni, Fe-10Ni, and Fe-10Ni-5S, respectively).

116 Electrical results for Fe and Fe – 5 wt.% S from Pommier (2018) were added to the dataset for  
117 comparison. The detailed compositions are listed in Table 1. The starting materials were  
118 homogeneous powder mixtures of high-purity (> 99%) Fe, Ni and FeS powders (Alfa Aesar) and  
119 stored either in a sealed container placed in a desiccator or in a vacuum oven at 110°C until use.

120 Electrical experiments were conducted at 4.5 and 8 GPa in the multi-anvil apparatus at the  
121 Planetary and Experimental Petrology Laboratory at UCSD-SIO, using tungsten carbide cubes  
122 with a corner-truncation edge length of 8 mm and MgO octahedral pressure media with an edge  
123 length of 14 mm. The COMPRES electrical cell used as part of the experiments consists of a  
124 sample sleeve (MgO) that contains three alumina rings isolated by metal disks made of high-purity  
125 iron (Pommier et al., 2019). As illustrated in Figure 2, the sample is contained in the middle ring  
126 (ID of 1.45 mm, thickness of 1.25 mm). A rhenium heater was used and two 4-bore MgO beads  
127 each contain two W-Re wires. The temperature was monitored by a Type-C (W<sub>95</sub> Re<sub>5</sub>-W<sub>74</sub> Re<sub>26</sub>)  
128 thermocouple. The cell contains two thermocouples that are in contact with the Fe metal disks, the  
129 four wires serving as electrodes. All MgO parts were fired at > 1,000°C and stored in a desiccator.

130 Electrical experiments were performed under quasi-hydrostatic conditions in the multi-  
131 anvil press during cooling and heating cycles using the four-electrode method (e.g., Pommier,  
132 2018) (Table 2). A dwell was first applied to the sample at a temperature below the eutectic  
133 temperature (~600°C). All experiments were quenched at the highest temperature by shutting off  
134 the power to the heating system. The electrode system consists of two wires for voltage drop  
135 measurement and two wires for current measurement. A current with a controlled voltage (DC  
136 potential of 1 V and AC amplitude between 500 and 1000 mV) was imposed during the  
137 measurement. The bulk electrical resistance was measured using an impedancemeter (1260  
138 Solartron Impedance/Gain-Phase Analyzer). A manual shift was used to either read temperature

139 or the electrical response. The measured electrical resistance  $R$  and sample geometric factor  $G$  are  
140 used to calculate the electrical resistivity  $\rho$

$$141 \quad \rho = R \times G \text{ with } G = A/l \quad (1)$$

142 with  $A$  is the area of one electrode (metal disk with a diameter of 2.5 mm), and  $l$  is the thickness  
143 of the sample (determined using SEM images of the recovered sample). Relative errors on values  
144 of  $\rho$  were calculated on the basis of errors on  $A$  and  $l$  as well as propagated errors on each measured  
145 value of resistance  $R$  (Table 2). The sample resistance was obtained from the measured (bulk)  
146 resistance value by removing the contribution from the iron disks and the alumina ring (Pommier  
147 et al., 2019).

148 Longitudinal sections of the experimental charges were mounted and polished for  
149 analytical and chemical characterization using scanning electron microscope (SEM) imaging and  
150 an electron probe microanalyzer (EPMA) at the University of Lille, France. Backscattered electron  
151 images and quantitative chemical analyses of Fe-Ni-S samples were obtained with a Cameca SX  
152 100 electron microprobe using wavelength dispersive spectrometry (WDS). Analyses were  
153 performed with a 15 kV accelerating voltage and a beam current of 40 nA. The phases were  
154 analyzed with a focused beam. Natural and synthetic standards were used to quantify the amount  
155 of different elements ( $\text{Fe}_2\text{O}_3$  for Fe and O, Ni metal for Ni, ZnS for S, orthose for Al, and W metal  
156 for W).

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## 162 **RESULTS**

### 163 **Chemical and textural analyses**

164 Chemical analyses of retrieved samples using the electron microprobe and EDS-SEM  
165 techniques are presented in Table 3 and Figure 3. No significant contamination of the samples by  
166 the alumina or the magnesia rings is observed. A few isolated FeO grains are observed in some  
167 samples but no significant amount of oxygen is measured using electron microprobe analyses  
168 (Table 3, Figure 3). Some samples present a small amount of W in the dendritic phase, which is  
169 consistent with unavoidable interactions with the thermocouple wires at high temperature. No  
170 strong compositional heterogeneity is observed across the recovered samples; an EPMA profile  
171 performed in the Fe-10Ni sample at 8 GPa illustrates this compositional homogeneity (Figure 3).

172 SEM images of recovered samples are available in Supplementary Figure 1. The retrieved  
173 samples present textures of fully molten alloys following rapid solidification. The electrical cell  
174 geometry was well-preserved during the experiment, minimizing uncertainty on the geometric  
175 factor calculations. Although dendritic patterns are visible in all samples, the texture of quenched  
176 Fe-Ni-S samples presents a higher contrast between dendritic structures (light color) and the  
177 surrounding phase (dark color) than the Fe-Ni samples. This can be explained by the presence of  
178 S in the quenched phase surrounding the dendrites, that increases the color contrast between the  
179 two phases (the dark region is a S-rich quenched melt, due to a lower Fe content).

180

### 181 **Electrical resistivity results**

182 The electrical resistivity results as a function of temperature are presented in Figures 4 and  
183 5 and the electrical resistivity as a function of the amount of alloying agent(s) is illustrated in  
184 Figure 6. Reproducibility of the electrical measurements was checked by reproducing two



185 experiments on Fe-5Ni and Fe-10Ni samples at 4.5 GPa (Table 2). The dependence of resistivity  
186 to temperature is comparable for all samples: at 4.5 GPa and at temperature higher than the alpha-  
187 gamma (or bcc-fcc) transition, the electrical resistivity increases by a factor of about 1.6-1.7 for  
188 pure iron, and of about 1.3-1.4 for Fe-5Ni, Fe-10Ni, and Fe-Ni-S. This increase in resistivity with  
189 temperature is linear at high temperature for the Fe-Ni samples. The bcc-fcc transition in the solid  
190 state is clearly identified (especially at 4.5 GPa), marked by a change in slope around 900 K. The  
191 onset of melting, which is characterized by a small increase in resistivity for Fe and Fe-5S, is not  
192 clearly identified for Ni-bearing samples, both at 4.5 and 8 GPa (Figures 4 and 5).

193         The experimental results suggest that the presence of Ni increases the resistivity of both S-  
194 free and S-bearing alloys; at defined pressure and temperature, Fe-Ni alloys are more resistive than  
195 pure iron and the Fe-Ni-S alloy is more resistive than Fe-S (Figures 4 and 5). In particular, the  
196 addition of 5 wt.% Ni to molten iron increases electrical resistivity by a factor of 4, both at 4.5 and  
197 8 GPa (Figure 6; at 8 GPa, the resistivity of pure iron is from Deng et al., 2013 and Silber et al.,  
198 2018). However, the difference in resistivity between Fe-5Ni and Fe-10Ni is negligible, as the Fe-  
199 10Ni samples present a similar or, at high temperature, a slightly higher resistivity than the Fe-5Ni  
200 samples (Figures 5 and 6). At 4.5 GPa and over 900-1700 K, a difference in resistivity of a factor  
201 of about 3 is observed between pure iron and Fe-Ni samples, and may increase slightly at pressure  
202 >4.5 GPa. The Fe-Ni-S sample is slightly more resistive than the Fe-Ni samples at 4.5 GPa, but no  
203 significant difference is observed between the resistivity of the Fe-Ni samples and that of Fe-Ni-S  
204 at 8 GPa.

205         At 4.5 GPa and over the investigated temperature range, the nickel impurity resistivity is  
206 significant and comparable to the effect of sulfur: the resistivity of an iron alloy containing 5 wt.%  
207 or 10 wt.% Ni (4.77 and 9.56 mol.% Ni, respectively) is comparable to the resistivity of an iron

208 alloy containing 5 wt.% S (8.4 mol.% S) (Figures 5 and 6). At 8 GPa, the difference in resistivity  
209 between Ni-bearing and Ni-free alloys is higher than at 4.5 GPa, with Ni-bearing samples being  
210 significantly more resistive than Fe and Fe-5S (Figures 5 and 6).

211 The pressure effect is negligible for the investigated Fe-Ni samples, which contrasts with  
212 the Fe-S system. For instance, at 1300 K and between 4.5 and 8 GPa, electrical resistivity decreases  
213 by a factor of 1.8 for pure Fe and of 2.9 for Fe-5S (8.4 mol.% S) (and this factor increases for  
214 higher S contents up to 50 mol.%S, Pommier (2018)) whereas this factor is only of 1.03 and 1.06  
215 for Fe-5Ni (4.77 mol.%Ni) and Fe-10Ni (9.56 mol.% Ni), respectively (Figure 4). For the Fe-Ni-  
216 S composition, this factor increases to a value of 1.13, which may suggest that the presence of  
217 sulfur increases the pressure dependence of iron alloys resistivity. As discussed below, the  
218 difference in pressure dependence of the resistivity between S-bearing and Ni-bearing alloys might  
219 be explained by the combination of at least two different factors: the alloy compressibility and the  
220 phase assemblage of the starting materials.

221

222

## 223 **IMPLICATIONS**

### 224 **Effect of nickel on electrical resistivity and comparison with previous works**

225 Previous electrical experiments on Fe-Ni alloys under pressure (up to 70 GPa) have been  
226 conducted at 300 K by Gomi and Hirose (2015). As highlighted in Figure 6, their resistivity values  
227 at room temperature are lower than the ones measured in this study, which is consistent with the  
228 fact that temperature increases electrical resistivity. Over the 4.5-8 GPa pressure range, Gomi and  
229 Hirose (2015) measured a negligible effect of Ni content on the resistivity of Fe-Ni samples  
230 containing 5 and 10 wt.% Ni at room temperature, and the present study suggests that this

231 negligible effect persists under temperature (Figures 4 and 5). At atmospheric pressure, electrical  
232 experiments also observed the increase in resistivity with increasing Ni content and/or temperature  
233 (Ho et al., 1983 and references therein). Resistivity values for molten Fe-Ni alloys containing up  
234 to 15 wt.% Ni at 1 atm are in broad agreement with the ones collected under pressure up to 8 GPa:  
235 in the molten state, electrical resistivity values are about 130-160 microhm-cm at 1 atm vs. ~150-  
236 200 microhm-cm at 4.5 and 8 GPa. However, at 300K, a factor of up to 2.5 difference in resistivity  
237 is observed between different studies, as Ho et al. reported electrical resistivity values of 10-25  
238 microhm-cm vs. ~8-10 microhm-cm from Gomi and Hirose (2015).

239         The present dataset allows comparing the effect of Ni and S impurity on resistivity. As an  
240 alloying agent, the addition of sulfur increases the electrical resistivity of iron (Argyriades et al.,  
241 1959; Pommier, 2018). At 4.5 GPa, the resistivity of Fe-Ni and Fe-S with a comparable amount  
242 of alloying component (9.56 mol. % Ni and 8.4 mol.% S; Table 1) is similar over the investigated  
243 temperature range (Figure 5). At this pressure, the Fe-10Ni-5S sample, containing 16.09 mol.%  
244 alloying agents (Table 1), presents only slightly higher resistivity values than the Fe-Ni and Fe-S  
245 samples, whereas at 8 GPa, the difference in electrical resistivity corresponds to a factor of ~2.5  
246 between Fe-S and Fe-Ni-S samples and is negligible between Fe-Ni and Fe-Ni-S samples. This  
247 suggests that the effect of pressure on resistivity depends on the alloy chemistry, and in the Fe-  
248 10Ni-5S sample, the dependence of electrical resistivity to pressure is controlled by Ni impurity,  
249 not by S impurity. Different factors might explain these two observations; in particular, differences  
250 in compressibility and the phase assemblage could contribute to the contrasting pressure effect on  
251 resistivity. First, the Fe-S alloy is less dense than Fe and Ni-bearing iron alloys (e.g., Sanloup et  
252 al., 2000; Lin et al., 2004; Kawaguchi et al., 2017), and Fe-Ni alloys present a slightly higher  
253 density than pure Fe (e.g., Martorell et al., 2015; Watanabe et al., 2016). For instance, Fe-10S at 5

254 GPa and 1770K has a density of about 5.65 g/cm<sup>3</sup> (Sanloup et al., 2000) whereas the density of  
255 Fe-7.6Ni and Fe-7.6Ni-10S at a similar temperature and extrapolated to the same pressure is about  
256 7.8 and 6.8 g/cm<sup>3</sup>, respectively (Kawaguchi et al., 2017). The Fe-S alloy is thus more compressible  
257 than pure Fe and Fe-Ni alloys, which can explain -at least partly- the higher pressure effect on the  
258 resistivity of Fe-5S than on the one of the other investigated alloys. Second, in the Fe-Ni-S sample,  
259 the small pressure-dependence of resistivity suggests a control of the electrical properties by Ni  
260 rather than S, and this might be explained by the multi-phase assemblage of the starting materials.  
261 The Fe-Ni-S sample is likely a mixture of Fe-Ni(-S) alloy with a small volume fraction of Fe<sub>1-x</sub>S,  
262 as the solubility of sulfur is low in solid fcc iron (e.g., Li et al., 2001; Hayashi et al., 2009). The  
263 low S solubility in fcc Fe could result in a minor role of sulfur in controlling the bulk resistivity,  
264 compared to the effect of nickel that substitutes for Fe. The presence of two phases in the solid Fe-  
265 Ni-S sample may account for why Fe-Ni-S and Fe-Ni resistivity present a similar P dependence.  
266 Further work is required to demonstrate whether or not these observations about the relative effect  
267 of nickel and sulfur on electrical resistivity also apply to pressures higher than 8 GPa.

268

### 269 **Thermal conductivity estimates of Fe-Ni alloys**

270 Watanabe et al. (2019) demonstrated that experimentally-measured thermal conductivities  
271 of Fe-Ni melts at atmospheric pressure and high temperature (1700-2000 K) are larger than those  
272 calculated using the Wiedemann-Franz law, due to the contribution of the thermal vibration of  
273 atoms to the thermal conductivity of Fe-Ni alloys. The empirical Wiedemann-Franz law relates  
274 thermal conductivity and electrical resistivity as follows

$$275 \quad L_0 \times T = k \times \rho \quad (2)$$

276 with  $k$  the thermal conductivity,  $\rho$  the electrical resistivity,  $L_0$  the Sommerfeld value of the Lorenz  
277 number ( $2.445 \times 10^{-8} / \text{W } \Omega \text{ K}^{-2}$ ), and  $T$  the temperature (Wiedemann and Franz, 1853). In the study  
278 by Watanabe et al. (2019), the measured thermal conductivities  $k$  are on average about 29.5%  
279 higher than the ones computed using the Wiedemann-Franz law. This implies that 1) the empirical  
280 Wiedemann-Franz law provides lower bound estimates of  $k$  for Fe-Ni alloys, and 2) at first  
281 approximation, an upper bound of  $k$  can be calculated assuming a 30% increase in thermal  
282 conductivity values computed using Eq. (2).

283 Upper and lower bounds of thermal conductivity  $k$  of Fe, Fe-5S, and Fe-Ni(-S) alloys were  
284 computed using the experimentally-determined electrical resistivity values at 4.5 and 8 GPa. These  
285 estimates are presented in Figure 7 for temperatures  $>1000$  K. The computed values of  $k$  show that  
286 Fe-Ni and Fe-Ni-S samples present a narrow range of low thermal conductivity values at 4.5 and  
287 8 GPa (between  $\sim 12$  and  $20$  W/m.K), which is consistent with their high electrical resistivity. A  
288 small temperature dependence is observed, with  $k$  slightly increasing with increasing temperature.  
289 These computed  $k$  values are comparable to the ones for Fe-5S at 4.5 GPa and to Fe-15.6P obtained  
290 from electrical measurements at a slightly lower pressure (3.2 GPa; Yin et al., 2019), but are  
291 significantly lower than the ones obtained for pure iron and Fe-5S at 8 GPa (Figure 7). This would  
292 be consistent with the hypothesis that the pressure dependence of both electrical resistivity and  
293 thermal conductivity of Fe and Fe-5S alloys is more important than the one of Fe-Ni(-S) alloys  
294 (and possibly, Fe-P alloys).

295

### 296 **Application to the core of small terrestrial bodies**

297 The pressure and temperature conditions considered in this study are directly relevant to  
298 the cores of the Moon and of Ganymede (Figure 1). Although there is a large uncertainty on the

299 composition of these terrestrial cores, it has been suggested that S might be present as a major  
300 alloying component (e.g., Breuer et al., 2015; Rückriemen et al., 2015). Assuming a Fe-S core  
301 chemistry, numerical studies have suggested that the thermal conductivity of the Lunar core ranges  
302 from about 15 to 65 W/m.K (Laneuville et al., 2013), and a comparable range of 20-60 W/m.K has  
303 been used to model the cooling of Ganymede's core (Rückriemen et al., 2015). These thermal  
304 conductivity values come from computations (de Koker et al., 2012), not from experiments  
305 conducted at relevant pressure and temperature conditions. As shown in Figure 7a, only the lower  
306 bound of these assumed  $k$  estimates overlaps with the thermal conductivity estimates of Fe-Ni(-S)  
307 alloys. This suggests that these core models may have assumed  $k$  values that are too high. It could  
308 be a significant issue because the thermal conductivity determines the heat flow down the adiabat,  
309 and thus, the presence and structure of any thermally stratified layer at the top of the core and the  
310 power available to generate a dynamo.

311 The adiabatic heat flux ( $q_a$ ) can be estimated using the following equation

312 
$$q_a = -k \times \frac{dT_a}{dr} \quad \text{with} \quad \frac{dT_a}{dr} = -\alpha \times g \times \frac{T_a}{c_p} \quad (3)$$

313 with  $k$  the thermal conductivity,  $dT_a/dr$  the adiabatic temperature gradient across the core, and  
314  $-\alpha \times g \times \frac{T_a}{c_p}$  being evaluated at radius  $r$ . The following values were considered:  $C_p = 830$   
315  $\text{J.kg}^{-1}.\text{K}^{-1}$ ,  $g = 1 \text{ m/s}^2$  (Ganymede) or  $0.6 \text{ m/s}^2$  (Moon). Adiabatic temperatures  $T_a$  come from  
316 Breuer et al. (2015), who proposed two present-day adiabats for each of the two cores using the  
317 same values of  $C_p$  and  $g$  as above and a value of  $\alpha = 9 \times 10^{-5} \text{ K}^{-1}$ .  $T_a$  ranges from about 1280 to  
318 1380 K or from 1640-1760 K for Ganymede's core and from about 1260 to 1600 K or from 1280-  
319 1630 K for the Lunar core, and the  $T_a$  profiles correspond to straight lines. It has been suggested  
320 that core chemistry can affect modestly the slope of the adiabats (Williams, 2009), but this

321 steepening occurs at pressure higher than the core of the Moon or Ganymede. As mentioned in  
322 Williams (2009), there are two sets of values available for the thermal expansion of iron-rich nickel  
323 alloys coming from previous studies (Nasch and Manghnani, 1998; Seifert et al., 1998),  
324 corresponding to  $8 \times 10^{-5}$  and  $1.3 \times 10^{-4} \text{ K}^{-1}$ , or  $1.05 \times 10^{-4} (+/-2.5 \times 10^{-5}) \text{ K}^{-1}$ , for the  
325 compositions considered in the present study. Calculations of  $q_a$  have therefore considered three  
326 different values of  $\alpha$  ( $8 \times 10^{-5}$ ,  $9 \times 10^{-5}$ , and  $1.3 \times 10^{-4} \text{ K}^{-1}$ ). The radius  $r$  of Ganymede's core  
327 is considered to be  $\sim 840$  km and the one of the Lunar core to be  $\sim 330$  km. In the case of a  
328 homogenous Fe-Ni(-S) core, assuming a constant  $k$  value across the core (taken to be 17 or 20  
329 W/m.K, for the lower and higher estimates of the adiabats, respectively) leads to  $q_a$  values of 2.36  
330 or 3.82 mW/m<sup>2</sup> for Ganymede and 1.39 or 2.12 mW/m<sup>2</sup> for the Moon, depending on the adiabat  
331 considered and considering  $\alpha = 9 \times 10^{-5} \text{ K}^{-1}$ . Considering another  $\alpha$  value ( $8 \times 10^{-5}$  or  $1.3 \times 10^{-4}$   
332  $\text{K}^{-1}$ ) can lead to significant differences in heat flux estimates (shaded areas in Figure 7b span  
333 variations of about 60% in  $q_a$  values), illustrating therefore that adiabatic heat flux estimates are  
334 only as good as the experimental constraints on each parameter. The thermal conductivity of Fe-  
335 Ni(-S) being almost constant over the considered pressure range (Figure 7a), calculating  $q_a$  using  
336  $k(r)$  (instead of a constant  $k$  value) results in very small ranges, as illustrated in Figure 7b: 2.36-  
337 2.69 mW/m<sup>2</sup> or 3.47-3.82 mW/m<sup>2</sup> from across Ganymede's core and 1.39-1.67 mW/m<sup>2</sup> or 1.77-  
338 2.12 mW/m<sup>2</sup> across the Lunar core, depending on the adiabat and considering the parameter values  
339 used in Breuer et al. (2015). In the case of a homogenous Ni-free, Fe-S core, the important  
340 pressure-dependence of  $k$  (Figure 7a) leads to the following  $q_a$  ranges across the core: 2.36-6.28  
341 mW/m<sup>2</sup> or 3.47-7.63 mW/m<sup>2</sup> from across Ganymede's core and 1.39-3.50 mW/m<sup>2</sup> or 2.03-4.24

342  $\text{mW/m}^2$  across the Lunar core, depending on the adiabat (Figure 7b). These  $q_a$  values correspond  
343 to a factor of about 2-2.7 across the core. This factor could be higher than 2-2.7 in terrestrial bodies  
344 that have metallic cores spanning higher depth ranges than Ganymede and the Moon. In  
345 comparison, higher constant  $q_a$  values than the ones listed above were considered by Rückriemen  
346 et al. (2015) and Laneuville et al. (2013) (8 and  $10 \text{ mW/m}^2$  for Ganymede and the Moon,  
347 respectively; Figure 7b) and low  $q_a$  values ( $< 4 \text{ mW/m}^2$ ) were considered in earlier core models  
348 (e.g., Hauck et al., 2006; Kimura et al., 2009). These  $q_a$  calculations have two major implications:  
349 1) At first approximation, a constant  $k$  value can be assumed as part of Ni-bearing core models,  
350 whereas modeling Fe or Fe-S cores requires accounting for  $k(r)$ ; 2) A similar amount of heat is  
351 conducted at any depth along the adiabat gradient of a Fe-Ni(-S) core, whereas less heat is  
352 conducted down this gradient at shallow depth in a Ni-free core. Because variation in heat  
353 conduction is critical to drive convection, this implies that it is possibly easier to drive convection  
354 in a Ni-free core than in a Ni-bearing core.

355 Another parameter to account for as part of  $k$  and  $q_a$  calculations across a metallic core is  
356 the possible heterogeneity of core composition as a function of depth that results from core cooling  
357 processes. Core fractional crystallization, which is expected in top-down or bottom-up cooling  
358 regimes, likely results in the heterogenous distribution of light elements (such as S) across the  
359 core, characterized by the enrichment in light elements in the outer portion of the core (e.g., Breuer  
360 et al., 2015; Dumberry and Rivoldini, 2015; Rückriemen et al., 2015; Davies and Pommier, 2018;  
361 Pommier, 2018). New modeling studies considering the distribution and amount of nickel and light  
362 elements across the core are required to assess the importance of thermal conductivity gradients  
363 with depth on core convection.



364

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545 **Figure captions**

546 **Figure 1:** Phase relationships in Fe, Fe-Ni, and Fe-Ni-S systems and summary of experimental  
547 conditions in previous electrical works on Fe-Ni alloys and in this study. Phase diagrams for solid  
548 phases are from Huang et al. (1988). Fe-Ni alloys melting point (MP) temperature at 1atm from  
549 Hansen (1958) and Fe melting curve from Ma et al. (2004) and Anzellini et al. (2013). Fe-S and  
550 Fe-Ni-S eutectic curves are from Fei et al. (2000); Li et al. (2001); Stewart et al. (2007); Zhang  
551 and Fei (2008); Morard et al. (2007). Fe-Ni-S eutectic melting temperature at 20 GPa comes from  
552 Zhang and Fei (2008) for a Ni/(Ni+Fe) ratio of 0.09. GH2015: Gomi and Hirose, 2015; H et al.  
553 1983: Ho et al. (1983). Comparison with the expected pressure and temperature conditions for the  
554 core of the Earth's moon and Ganymede are also shown (green rectangles) (after Breuer et al.,  
555 2015).

556

557 **Figure 2:** COMPRES electrical cell developed for the 4-electrode experiments using 14/8  
558 assemblies. Thermocouple wires are also used as electrodes and are in contact with Fe disks.

559

560 **Figure 3:** EMPA profile in recovered sample (Fe-10Ni, at 8 GPa) showing the concentration of  
561 iron (blue), nickel (green) and oxygen (orange) from the Fe electrode disk to most of the sample's  
562 thickness. The variations in the iron and nickel contents are consistent with crossing the quenched  
563 phases. The average value of Fe and Ni contents show a homogenous composition (90 wt.% Fe,  
564 10 wt.% Ni) in the retrieved molten sample.

565

566 **Figure 4:** Electrical resistivity results as a function of temperature for the different compositions.

567 A) Resistivity of Fe and Fe-5S samples at 4.5 and 8 GPa, and comparison with previous works on

568 pure iron at 7, 5 and 4.5-8 GPa. (D. et al.2013: Deng et al. (2013), GH2015: Gomi and Hirose  
569 (2015), P. et al.2018: Pommier (2018)). B) Resistivity of Fe-5Ni samples at 4.5 and 8 GPa. The  
570 experiment at 4.5 GPa was repeated to check reproducibility. C) Resistivity of Fe-10Ni samples at  
571 4.5 and 8 GPa. The experiment at 4.5 GPa was also repeated to check reproducibility. D)  
572 Resistivity of Fe-10Ni-5S samples at 4.5 and 8 GPa. The alpha-gamma transition and the onset of  
573 melting are indicated with arrows on all plots.

574

575 **Figure 5:** Electrical resistivity results at 4.5 GPa (left) and 8 GPa (right). A slight decrease in  
576 resistivity as pressure is increased is observed for Fe-Ni(-S) samples, whereas the pressure effect  
577 on resistivity is higher for Fe and Fe-S samples. Iron alloys are more resistivity than pure iron and  
578 Fe-Ni(-S) samples are more resistive than Fe-5S at 8 GPa. Data at 7 GPa for pure Fe from Deng  
579 et al. (2013).

580

581 **Figure 6:** Effect of alloy chemistry on resistivity. Left side: data at 4.5 GPa. Right side: data at 8  
582 GPa. A comparison with data on Fe-Ni at 300K (Gomi and Hirose, 2015), Fe-5S (Pommier, 2018),  
583 and Fe (Deng et al., 2013; Silber et al., 2018) is provided.

584

585 **Figure 7:** A) Thermal conductivity ( $k$ ) estimated from electrical measurements at 4.5 and 8 GPa  
586 as a function of temperature (LB=lower bound, UB=upper bound). All the estimates for Fe-Ni(-S)  
587 samples at 4.5 and 8 GPa and Fe-5S at 4.5 GPa fit into a narrow range of  $k$  values (grey lines in  
588 purple area). Comparison with estimates for Fe (blue) from Deng et al. (2013) (7 GPa), Secco and  
589 Schloessin, 1989 (5.3 GPa) and Silber et al. (2018) (3-8 GPa), and for Fe-P from Yin et al. (2019)  
590 (3.2 GPa; orange line). Thermal conductivity decreases significantly when Ni is added to the core.

591 The yellow and orange areas correspond to thermal conductivity estimates used as part of previous  
592 thermochemical models for the core of Ganymede (Rückriemen et al., 2015) and the Moon  
593 (Laneuville et al., 2013), respectively. B) Adiabatic heat fluxes across Ganymede (yellow lines)  
594 and the Moon (red lines) cores, calculated using Eq. 3 and a of thermal expansion value used in  
595 Bruer et al. (2015) ( $9 \cdot 10^{-5} \text{ K}^{-1}$ ). The red and yellow shaded areas correspond to adiabatic heat fluxes  
596 obtained with a thermal expansion value of  $1.05 \cdot 10^{-5} (+/- 2.5 \cdot 10^{-5}) \text{ K}^{-1}$  (Williams, 2009). Dashed  
597 lines correspond to the adiabatic heat flux values used in core modeling studies (Rückriemen et  
598 al., 2015 for Ganymede, Laneuville et al., 2013 for the Moon). The grey areas correspond to the  
599 low and high adiabats for both moons, from Breuer et al. (2015). See text for details.

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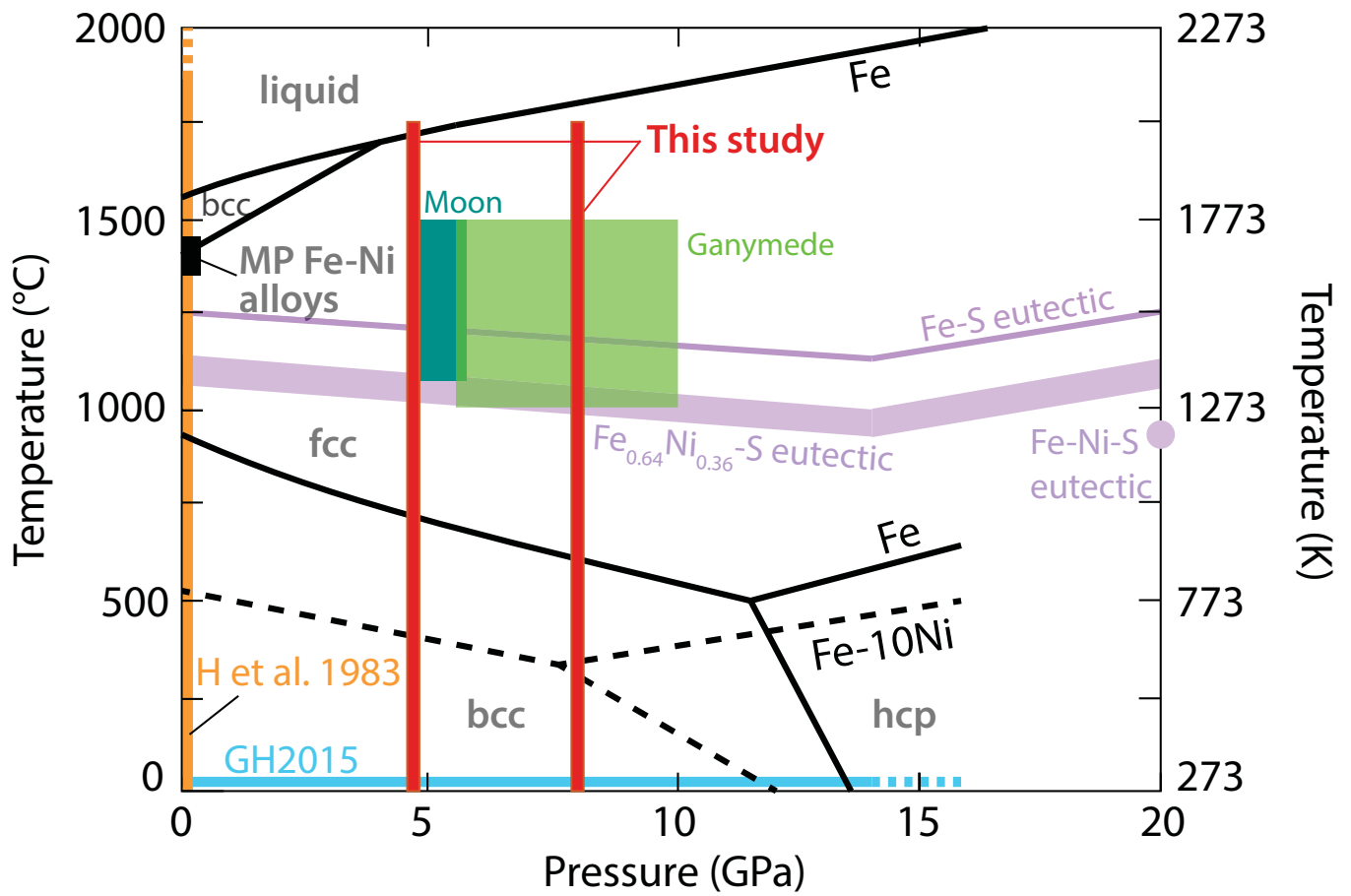
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603 **Supplementary figure captions:**

604 **Supplementary Figure 1:** Backscattered SEM images of molten samples recovered after  
605 electrical experiments. A) Whole electrical cell containing a recovered Fe-10Ni sample. B) Fe-5Ni  
606 sample at 4.5 GPa quenched at 1894 K. C) Fe-5Ni-3S at 4.5 GPa quenched at 1844 K. D) Fe-5Ni-  
607 3S at 8 GPa quenched at 1903 K. The darker region corresponds to the S-rich quenched melt.

Figure 1



**Figure 2**

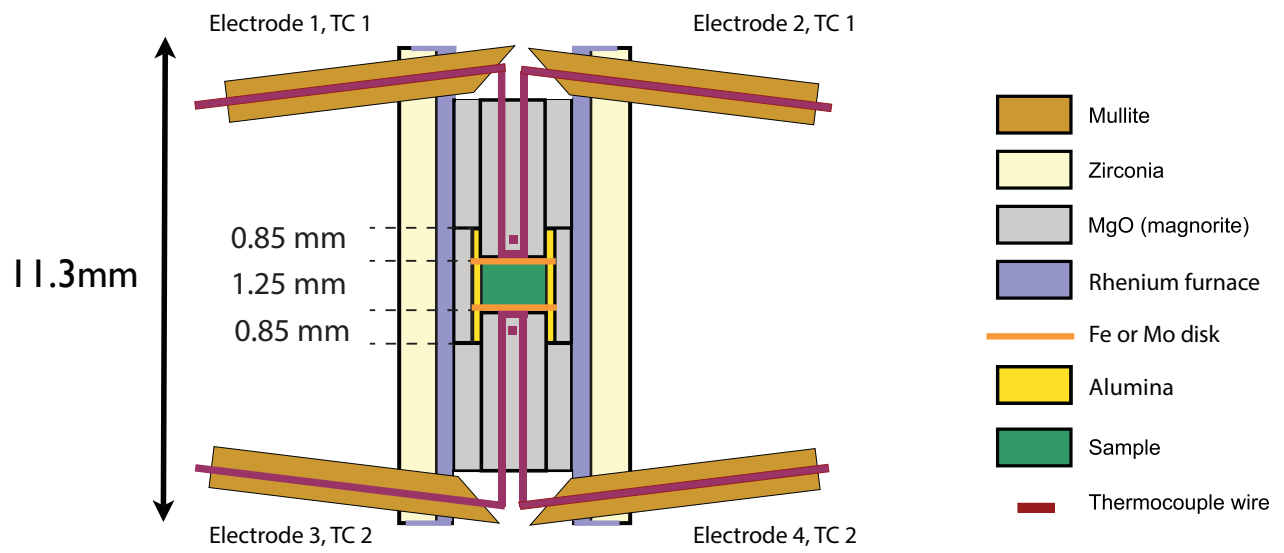


Figure 3

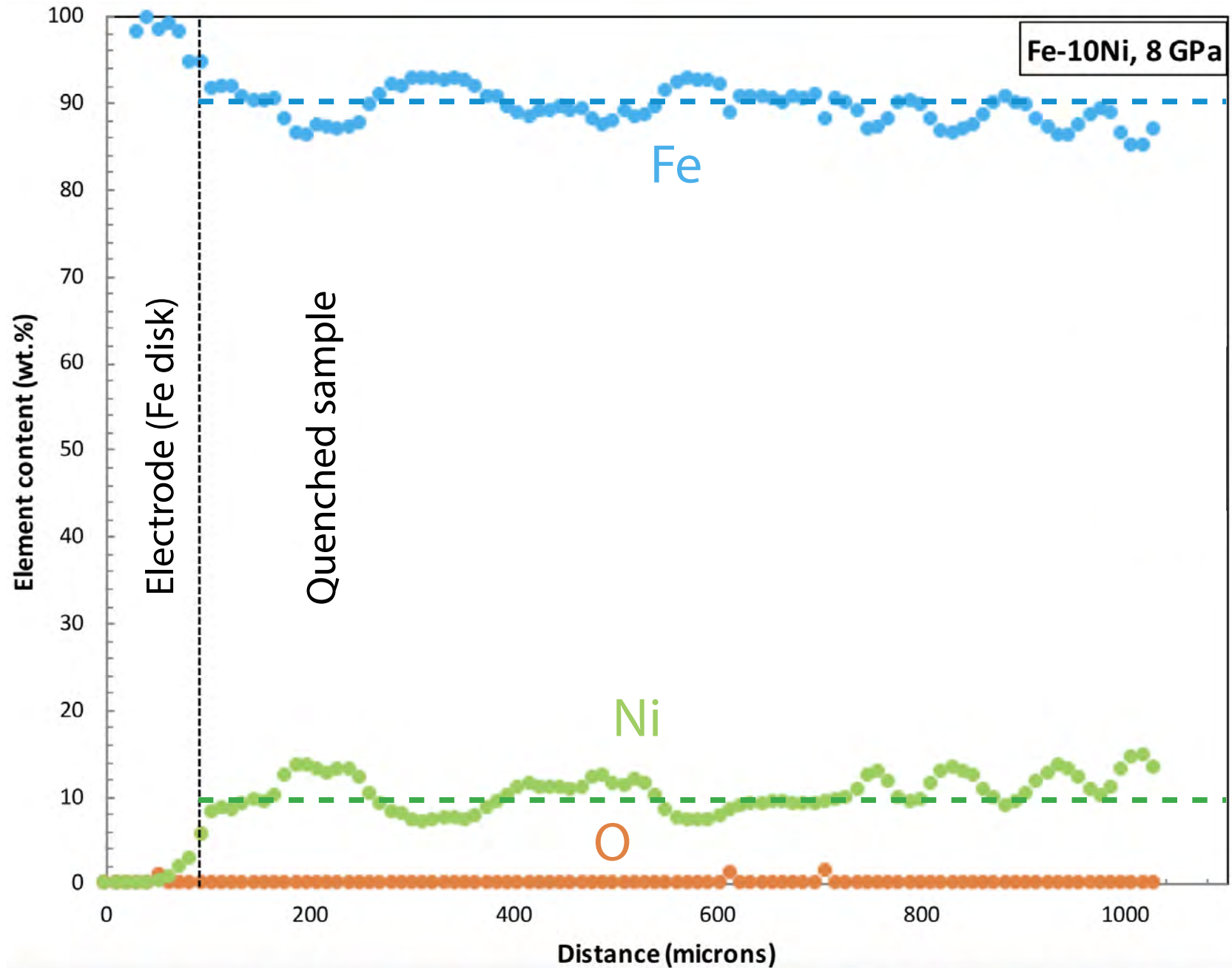


Figure 4

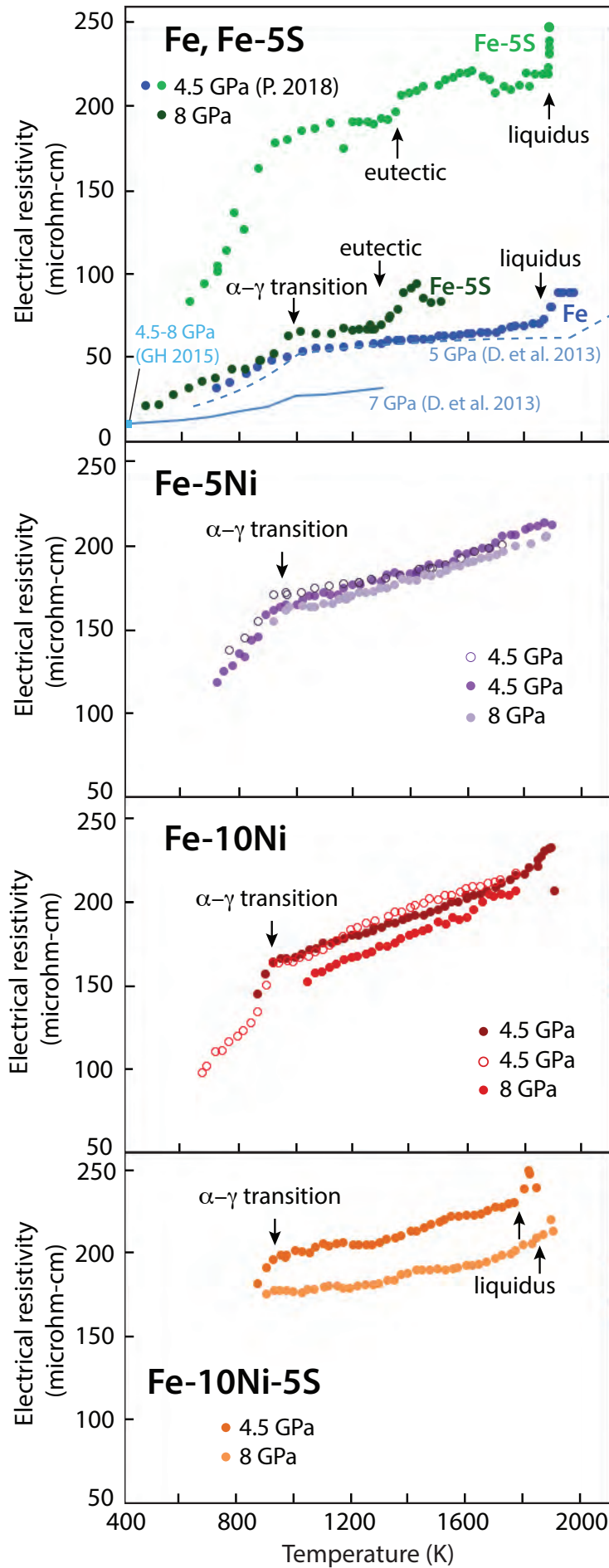


Figure 5

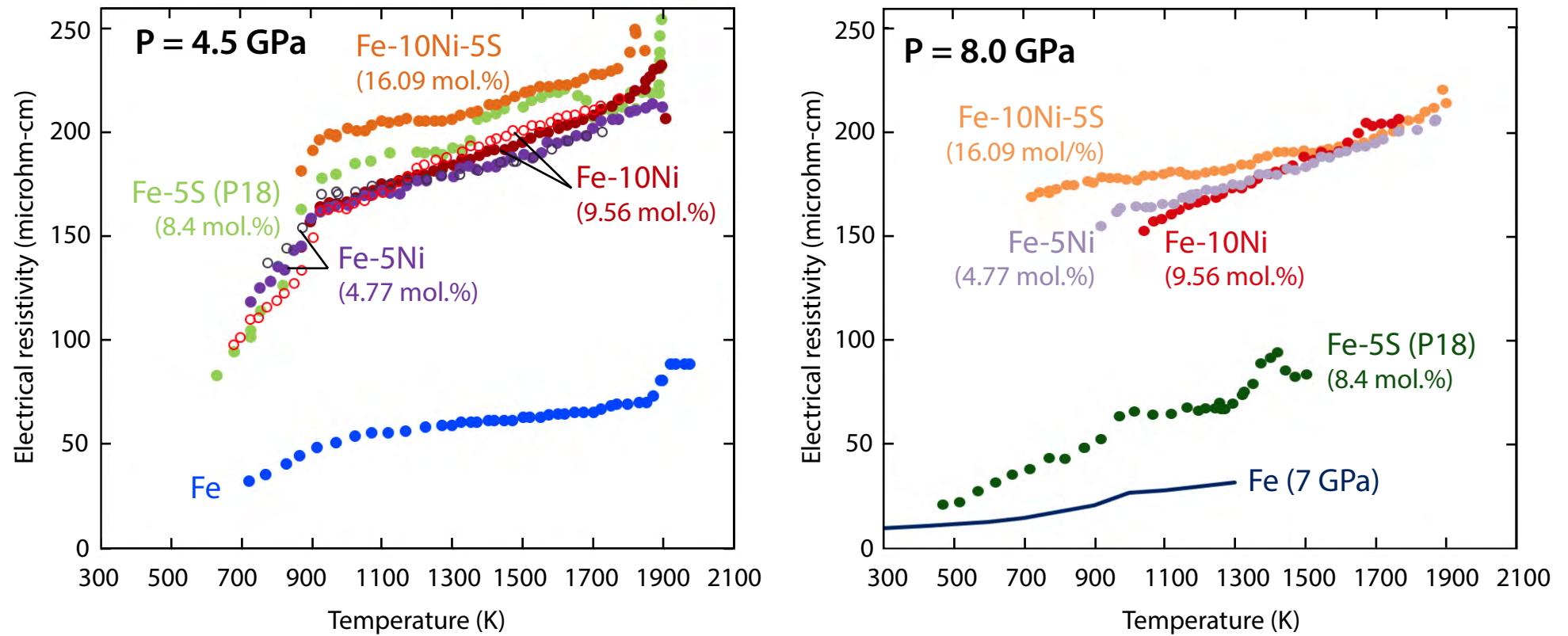
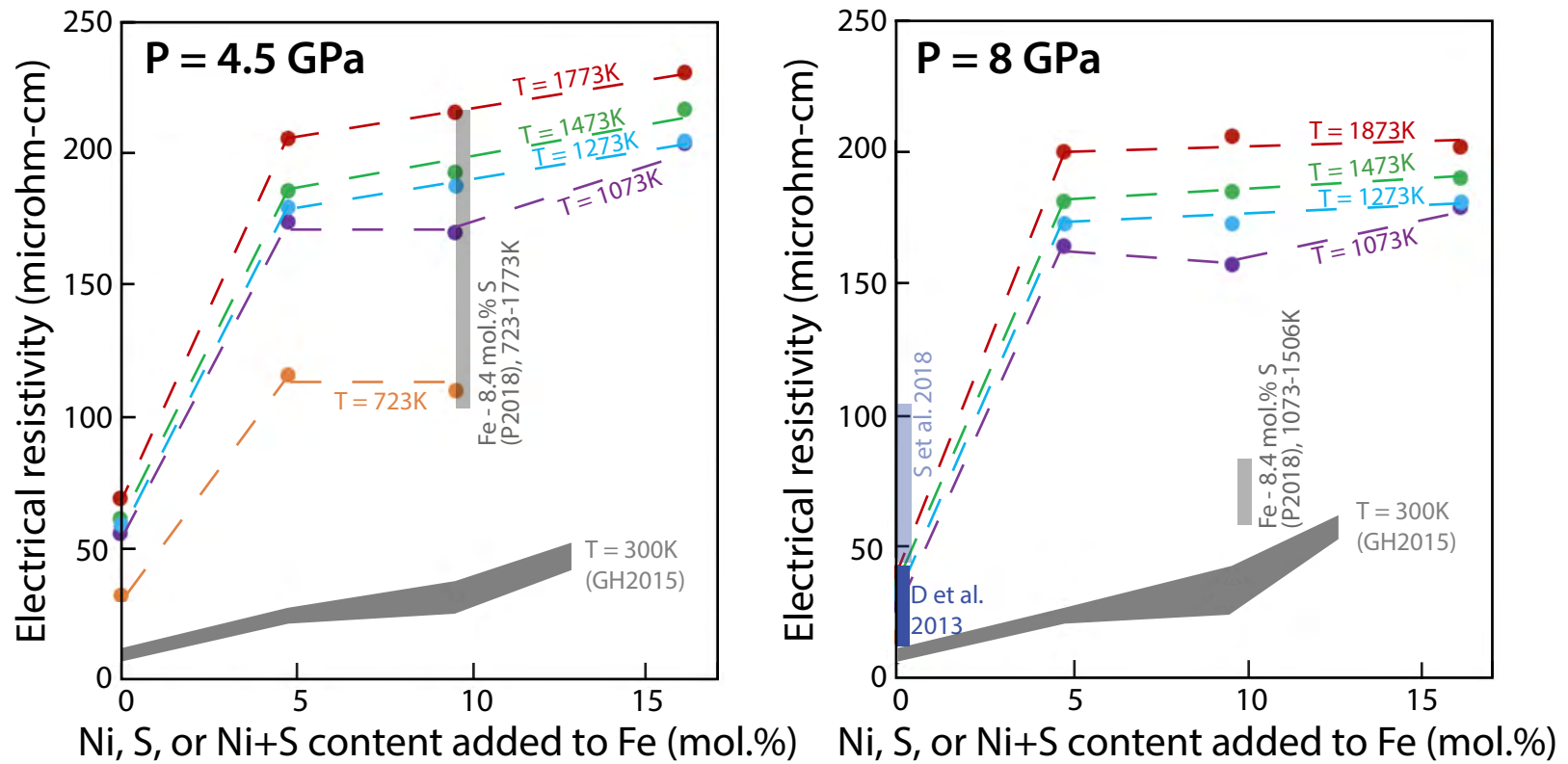
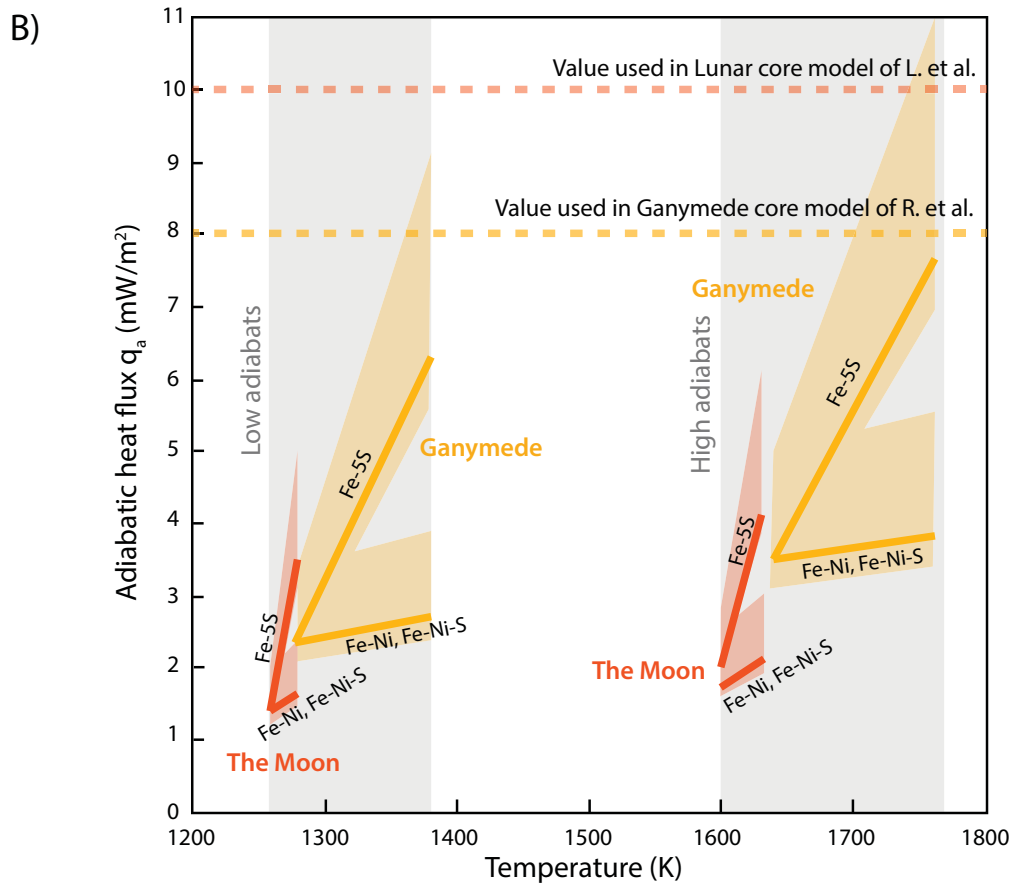
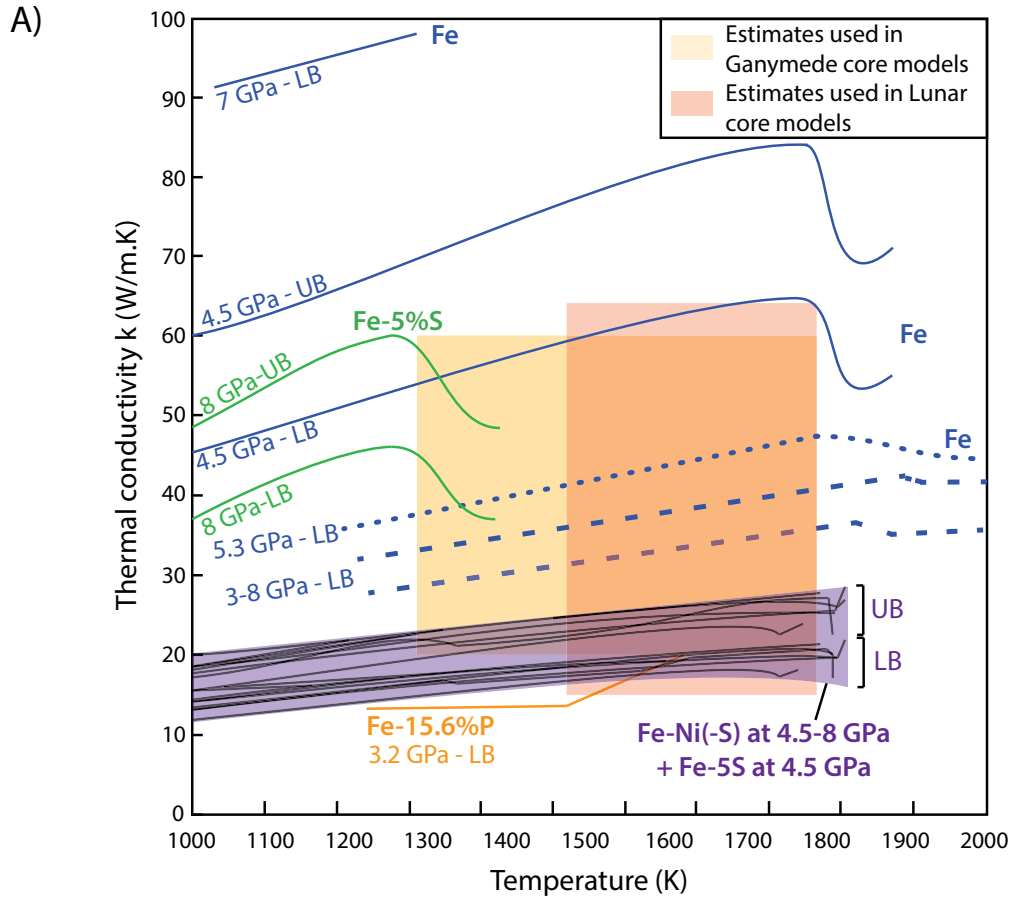




Figure 6



**Figure 7**



**Table 1. Composition of starting materials.**

| <b>System</b> | <b>wt.% Ni</b> | <b>mol.% Ni</b> | <b>wt.% S</b> | <b>mol.% S</b> |
|---------------|----------------|-----------------|---------------|----------------|
| Fe*           | -              | -               | -             | -              |
| Fe-5S*        | -              | -               | 5.0           | 8.40           |
| Fe-5Ni        | 5.0            | 4.77            | -             | -              |
| Fe-10Ni       | 10.0           | 9.56            | -             | -              |
| Fe-10Ni-5S    | 10.0           | 8.40            | 5.0           | 7.69           |

\* From Pommier (2018).

**Table 2. Summary of electrical experiments.**

| <b>Composition</b> | <b>Pressure (GPa)</b> | <b>T range (K)</b> | <b>Initial dwell duration (hr)</b> | <b>Uncertainty on resistivity (%)<sup>◇</sup></b> |
|--------------------|-----------------------|--------------------|------------------------------------|---|
| Fe*                | 4.5                   | 720-1973           | -                                  | 3.7-4.9   |
| Fe-5S*             | 4.5                   | 628-1891           | 3.5                                | 1.8-2.5   |
| Fe-5S*             | 8.0                   | 471-1506           | 2                                  | 4.7-9.9   |
| Fe-5Ni             | 4.5                   | 724-1894           | 2                                  | 4.5-5.1   |
| Fe-5Ni             | 4.5                   | 771-1722           | 1                                  | 4.5-4.8   |
| Fe-5Ni             | 8.0                   | 921-1874           | 2                                  | 6.6-6.9   |
| Fe-10Ni            | 4.5                   | 867-1904           | 2.5                                | 3.4-3.9   |
| Fe-10Ni            | 4.5                   | 676-1771           | 1                                  | 4.9-5.8   |
| Fe-10Ni            | 8.0                   | 1045-1769          | 2.5                                | 8.1-8.4   |
| Fe-10Ni-5S         | 4.5                   | 868-1844           | 1.5                                | 3.5-3.8   |
| Fe-10Ni-5S         | 8.0                   | 898-1903           | 2.5                                | 3.9-4.2   |

\* From Pommier (2018).

$$\diamond \Delta \rho = |\Pi r^2 / l| \times \Delta R + |2\Pi R r / l| \times \Delta r + |-\Pi R r^2 / l^2| \times \Delta l$$

with  $\rho$  the resistivity,  $l$  the sample thickness,  $R$  the electrical resistance of the sample, and  $r$  the radius of the electrode disk.

**Table 3. Chemical analyses on recovered samples (in wt.% norm. to 100; bdl: below detection limit).**

| Starting material                        | Quenched T (K) | P (GPa) | Phase*               | Fe    | Ni    | S     | W     | O     | Al  |
|--|----------------|---------|----------------------|-------|-------|-------|-------|-------|-----|
| <i>Electron microprobe spot analyses</i> |                |         |                      |       |       |       |       |       |     |
| Fe-10Ni-5S                               | 1844           | 4.5     | "Oak leaf" phase (5) | 93.88 | 4.567 | 0.076 | 1.276 | bdl   | bdl |
|  |                |         | Joints (3)           | 64.31 | 2.281 | 31.88 | 0.078 | 0.326 | bdl |
| Fe-10Ni-5S                               | 1903           | 8.0     | "Oak leaf" phase (5) | 90.25 | 4.163 | 0.093 | 4.604 | bdl   | bdl |
|  |                |         | Joints (3)           | 66.49 | 2.816 | 28.60 | 0.898 | bdl   | bdl |
| <i>EDS-SEM analyses</i>                  |                |         |                      |       |       |       |       |       |     |
| Fe-5Ni                                   | 1894           | 4.5     | Bulk sample          | 95.35 | 4.654 | -     | -     | -     | -   |
| Fe-10Ni                                  | 1904           | 4.5     | Bulk sample          | 93.64 | 6.360 | -     | -     | -     | -   |
| Fe-10Ni                                  | 1769           | 8.0     | Bulk sample          | 92.88 | 7.118 | -     | -     | -     | -   |
|  |                |         | Electrode disk       | 99.54 | 0.460 | -     | -     | -     | -   |

\*Numbers in parentheses correspond to the number of analyses.