



Influence of sulfur on the electrical resistivity of a crystallizing core in small terrestrial bodies

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ABSTRACT

Electrical experiments were performed on core analogues in the Fe–S system and on FeSi₂ up to 8 GPa and 1850 °C in the multi-anvil apparatus. Electrical resistivity was measured using the four-electrode method. For all samples, resistivity increases with increasing temperature. The higher the S content, the higher the resistivity and the resistivity increase upon melting. At 4.5 GPa, liquid FeS is up to >10 times more resistive than Fe-5 wt.% S and twice more resistive than FeSi₂, suggesting a stronger influence of S than Si on liquid resistivity. Electrical results are used to develop crystallization-resistivity paths considering both equilibrium and fractional crystallization in the Fe–S system. At 4.5 GPa, equilibrium crystallization, as expected locally in thin snow zones during top-down core crystallization, presents electrical resistivity variations from about 300 to 190 microhm-cm for a core analogue made of Fe-5 wt.%S, depending on temperature. Fractional crystallization, which is relevant to core-scale cooling, leads to more important electrical resistivity variations, depending on S distribution across the core, temperature, and pressure. Estimates of the lower bound of thermal resistivity are calculated using the Wiedemann–Franz law. Comparison with previous works indicates that the thermal conductivity of a metallic core in small terrestrial bodies is more sensitive to the abundance of alloying agents than that of the Earth's core. Application to Ganymede using core adiabat estimates from previous studies suggests important thermal resistivity variations with depth during cooling, with a lower bound value at the top of the core that can be as low as 3 W/mK. It is speculated that the generation and sustainability of a magnetic field in small terrestrial bodies might be favored in light element-depleted cores.

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1. Introduction

In terrestrial bodies (e.g., Earth, Mars, Mercury, Ganymede), the generation of a global intrinsic magnetic field likely results from convection in a fully or partially liquid metallic core (Breuer et al., 2015 and references therein). The presence and intensity of this field highly varies among cores and this diversity possibly arises from different crystallization mechanisms that take place during planetary cooling. The cooling rate of a planetary body is affected by temperature and core chemistry. Light elements (such as sulfur, silicon, oxygen, or hydrogen) could have been added in significant amount to the metallic iron core of terrestrial bodies during differentiation (e.g., Li and Agee, 1996). In particular, meteorite geochemistry (e.g., Dreibus and Wänke, 1985) and solubility experiments (e.g., Tsuno et al., 2011) suggest that the presence of sulfur in metallic cores is possibly a general feature of terrestrial bodies due to their iron-loving properties observed over a wide pressure range. Sulfur is expected to be a major element in the core of

small terrestrial bodies such as Mars, Mercury, and Ganymede (e.g., Dreibus and Wänke, 1985; Hauck et al., 2006; Stewart et al., 2007; Kimura et al., 2009). This contrasts with the Earth's core, where sulfur might be less abundant than previously thought since a combination of light and/or other elements is required to explain the core's density deficit (Alfè et al., 2002; Badro et al., 2015; O'Rourke and Stevenson, 2016; Hirose et al., 2017). The presence of sulfur – like any light element – in a cooling metallic core affects the onset of crystallization by lowering the liquidus temperature (e.g., Fei et al., 2000; Chen et al., 2008a; Stewart et al., 2007) and influences transport properties such as electrical resistivity (or its inverse, electrical conductivity) (Vostryakov et al., 1964; Suehiro et al., 2017), thermal resistivity (Suehiro et al., 2017), and density (Sanloup et al., 2000). Investigating how transport properties relate to core crystallization is required to increase our understanding of planetary evolution, as variations in mass and heat transport in the crystallizing fluid likely impact the convective and diffusive processes that govern core cooling and might contribute to generate a magnetic field (Schubert et al., 1996; Hauck et al.,

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2006; Dumberry and Rivoldini, 2015; Rückriemen et al., 2015; Davies and Pommier, 2018).

Core crystallization in terrestrial bodies is initiated at the depth where adiabat and melting curve intersect (Breuer et al., 2015). Because the melting temperature does not evolve linearly with pressure in the Fe–S system, crystallization may take place at different depth and existing magnetic observations of terrestrial planets and satellites are compatible with top-down, middle, and/or bottom-up crystallization regimes (e.g., Hauck et al., 2006; Chen et al., 2008b; Rückriemen et al., 2015; Davies and Pommier, 2018). In particular, a top-down crystallization regime is thought to be relevant in the core of small terrestrial bodies (Breuer et al., 2015). Electrical resistivity being particularly sensitive to melting, temperature, and chemistry, it is a relevant probe of core crystallization processes. However, the effect of sulfur abundance and crystallization-induced distribution of S across the core on electrical resistivity is not presently understood. The current experimental database of the electrical properties of iron and iron alloys comes mostly from experiments conducted at very high pressure to mimic Earth's core conditions (e.g., Seagle et al., 2013; Gomi and Hirose, 2015; Ohta et al., 2016), or at very low temperature (e.g., Kobayashi et al., 2005). The existing electrical experimental or computed data on Fe and its alloys at pressure and temperature conditions relevant to small bodies remain meager (Deng et al., 2013; Kiarasi and Secco, 2015; Suehiro et al., 2017) and did not investigate systematically the effect of the abundance of the alloying agent on the bulk electrical resistivity. Electrical resistivity can be related to thermal conductivity and magnetic field evolution, though these relationships are debated at planetary core conditions (e.g., Christensen, 2010; Secco, 2017).

Here the results of laboratory experiments at pressures up to 8 GPa and temperatures up to 1850 °C are reported for iron and iron-sulfur samples. One additional experiment was performed on iron disilicide to compare the effect of S and Si on bulk electrical resistivity. These experiments were designed to investigate the electrical properties of core analogues, in order to develop an electrical model of core cooling in small terrestrial bodies (such as Ganymede) containing different amounts of alloying agents and to estimate the effect of core chemistry on electrical and thermal conductivity upon cooling. The effect of alloying agents on the generation of a magnetic field is also discussed.

2. Experimental and analytical methods

2.1. Starting materials

Five starting compositions were considered: pure iron, Fe-5 wt.%S (5.69 mol.%), Fe-20 wt.%S (22.8 mol.%), Fe-36.5 wt.%S (FeS, 41.5 mol.%), and Fe-50.1 wt.%Si (FeSi₂, 44.7 mol.%) (Table 1). These samples were made from high purity Fe rod or of mixtures of high purity (>99%) Fe, FeS, and FeSi₂ powders. To avoid oxidation and contamination, all starting materials were stored in sealed glass containers within a glass desiccator. Nickel was not added to the core analogues as it was observed that its effect on the melting properties of Fe–S is insignificant (e.g., Stewart et al., 2007; Martorell et al., 2013) or only minor when observed (Zhang and Fei, 2008).

Among the starting compositions, Fe, FeS and FeSi₂ correspond to a single phase. Fe-5 wt.%S and Fe-20 wt.%S starting samples are not alloys, meaning that these two materials below the melting temperature correspond to Fe coexisting with S, instead of a Fe–S single phase. As shown below, comparison of the electrical data for all five materials suggests that the difference in bulk resistivity between these two materials (Fe-5 wt.%S and Fe-20 wt.%S) and single-phase samples (Fe, FeS and FeSi₂) is not significant at the considered experimental conditions. However, the interpretation of the electrical results will focus on data collected at temperatures above the eutectic temperature, i.e. when the samples are partially or fully molten and correspond to Fe–S alloys. Experiments on pure Fe were conducted on Fe powder and on an Fe rod in order to estimate the potential effect of electron scattering due to a granular (powder) sample on the bulk electrical resistivity.

2.2. Multi-anvil cell assembly

All electrical experiments were performed up to 8 GPa in a multi-anvil apparatus in the Planetary and Experimental Petrology Laboratory at UCSD-SIO, using tungsten carbide cubes with a corner-truncation edge length of 8 mm and mullite octahedral pressure media with an edge length of 14 mm. Rhenium heaters were used, placed inside an outer zirconia sleeve that provided thermal insulation. Experimental samples were 2 mm in diameter and 0.8–1.5 mm in length and were placed at the center of the cylindrical heater inside an MgO sleeve (Fig. 1). Two molybdenum squares (1.5 mm in edge) or two iron disks (outer diameter 2 mm)

Table 1
Summary of electrical experiments.

Run #	Starting composition			Electrode composition	Pressure (GPa)	Temp. range (K)	Dwell time (hr)	Relative error on resistivity (%) ^b
	System	wt.% S or Si	mol.% S or Si					
BB86	Fe ^a	–	–	Mo	4.5	1420–2129	–	4.8–5.7
BB129	Fe	–	–	Fe	4.5	720–1973	–	3.7–4.9
BB62	Fe–S	5.00	8.4	Mo	3.2	867–1773	2.5	9.1–12.6
BB83	Fe–S	5.00	8.4	Mo	4.5	1123–1970	2	8.1–11.9
BB135	Fe–S	5.00	8.4	Fe	4.5	628–1891	3.5	1.8–2.5
BB119	Fe–S	5.00	8.4	Mo	8.0	471–1506	2	4.7–9.9
BB120	Fe–S	20.0	30.3	Mo	4.5	478–1743	2	3.8–10.3
BB133	Fe–S	20.0	30.3	Fe	4.5	728–1558	6	3.5–4.6
BB60	Fe–S (FeS)	36.5	50	Mo	3.2	670–1385	2	1.5–6.3
BB76	Fe–S (FeS)	36.5	50	Mo	4.5	980–1767	2.5	5.6–10.8
BB131	Fe–S (FeS)	36.5	50	Fe	4.5	1253–1403	3.5	3.7–6.4
BB97	Fe–S (FeS)	36.5	50	Mo	8.0	923–1628	2	2.7–5.3
BB122	Fe–S (FeS)	36.5	50	Mo	8.0	1120–1590	3	3.7–7.3
BB140	Fe–Si (FeSi ₂)	50.1	66	Fe	4.5	1500–1803	3.5	5.6–6.0

^a Starting material is an iron disk. Samples in all other experiments are powders.

^b Error on resistivity ($\Delta\rho$) derived from Eq. (1).

In the case of Fe disk electrodes, $\Delta\rho = |\pi r^2/l| \times \Delta R + |2\pi Rr/l| \times \Delta r + |-\pi Rr^2/l^2| \times \Delta l$.

In the case of Mo square electrodes, $\Delta\rho = |r^2/l| \times \Delta R + |2Rr/l| \times \Delta r + |-\pi Rr^2/l^2| \times \Delta l$ with R the electrical resistance (corrected from the electrode foils), r the radius of the electrode disk in contact with the sample, l the thickness of the sample.

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