



Oxygen and silicon contents of Earth's core from high pressure metal–silicate partitioning experiments

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ABSTRACT

Oxygen and silicon partitioning between molten metal and silicate melts was measured in samples synthesised in piston–cylinder and multi-anvil presses between 2 and 21 GPa, 2273 and 2873 K, and at oxygen fugacities of 1.5–3.6 log units below the iron–wüstite buffer. Our partitioning data are used together with published data to parameterize the individual effects of pressure, temperature and composition on the partitioning of oxygen and silicon. Results show that the oxygen metal–silicate partition coefficient increases with increasing oxygen fugacity, temperature and pressure, whereas the silicon metal–silicate partition coefficient increases with decreasing oxygen fugacity, increasing temperature and pressure. Silicon and oxygen contents of Earth's core were derived for different core formation models. Considering single-stage core formation at 40 GPa, 3200 K, IW-2, the core would contain 1 to 3.5 wt.% silicon and 0.5 to 2.5 wt.% oxygen. In a continuous core-formation scenario, and depending on the oxidation path, Si core content varies from 1 to 11 wt.%, whereas oxygen content ranges from 0 to 2.5 wt.%. These models show that the oxygen content in the core cannot be significantly higher than 2.5 wt.%. In these compositional models, a range of combined silicon and oxygen concentrations in the core could satisfy the seismologically observed range of outer core density deficits.

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1. Introduction

The molten outer core density deficit (CDD) compared to pure iron at the same conditions was first estimated to be around 10% (Birch, 1952). More recently, Anderson and Isaak (2002) updated the value of the CDD between 2.9 and 7% depending on the temperature at the inner–outer core boundary (ICB). The CDD is accommodated by incorporating one or several light elements in the core, the amount of which varies depending on the light element(s) in question. The favoured candidates are silicon, sulphur, oxygen, hydrogen and carbon (e.g., Poirier, 1994). Geochemical and cosmochemical constraints impose less than 2 wt.% sulphur and about 0.2 wt.% carbon in the core (Dasgupta and Walker, 2008; Dreibus and Palme, 1996; McDonough, 2003). Because of limited interactions between the core and mantle since core formation (e.g., McDonough, 2003), the nature and concentration of the light element(s) are directly linked to metal–silicate partitioning of these elements during core–mantle differentiation. How the core came to its present configuration depends on the available material to make it, and the conditions

of its differentiation. Commonly, the bulk Earth's composition is considered similar to that of the CI chondrites composition corrected for volatile depletion, CI chondrites being primitive meteorites with a composition resembling that of the solar photosphere (e.g., Palme and Jones, 2003). Comparison of the siderophile element concentration in meteorites and in the Earth's mantle with experimental partitioning between molten metal and silicate melts provides an insight into the pressure and temperature conditions of core segregation (see for example the review by Righter, 2003). However, the conditions of core differentiation remain unclear and several models have been proposed in recent years. Most of these models consider that core formation took place through metal segregation in a deep silicate magma ocean, as a consequence of the large amount of heat available in early Earth's history. The simplest model, known as the single-stage model, considers that core materials equilibrated at the base of the magma ocean in a single event, namely at fixed conditions (e.g., Li and Agee, 1996; Righter et al., 1997). It has also been proposed that the single equilibration conditions may more plausibly represent an average of a wider range of conditions during a more dynamic process (Li and Agee, 1996). In agreement with dynamical models of Earth's accretion (Kaula, 1995; O'Brien et al., 2006), several studies have recently proposed that core formation occurred as a largely continuous process, with core segregation starting well before

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the end of Earth's accretion and under progressively more oxidising conditions as the Earth grew (Galimov, 2005; O'Neill, 1991; Wade and Wood, 2005).

Silicon is currently seen as the most probable light element in the core. Recent sound velocity measurements carried out at high pressure on solid Fe-alloy and extrapolated to conditions of the inner core concluded that a few percent of Si was likely present in the bulk core (Antonangeli et al., 2010; Badro et al., 2007). Metal–silicate partitioning data, which reveal that Si becomes more siderophile at reduced conditions and high temperatures, also support the presence of Si in the core (e.g., Wade and Wood, 2005). Partitioning studies have not provided a consistent value for the amount of Si in the core, mainly because the pressure dependence of Si partition coefficient has remained controversial, with negative correlations derived in Mann et al. (2009) and Corgne et al. (2008) but positive correlations found in Wade and Wood (2005) and Gessmann et al. (2001).

Oxygen has also been put forward as a possible major light element in Earth's core based on sound velocity measurements on iron alloys (Badro et al., 2007) and on required element partitioning between inner and outer core to account for seismic measurements (Alfè et al., 2002). At present, there is no systematic study of the partitioning of oxygen between metal and silicate melts. With the exception of the study of Li and Agee (2001), who explored a large range of metal and silicate compositions to study the behaviour of sulphur, silicon and oxygen, partition coefficients for oxygen have only been measured between molten metal and mantle minerals (ferroperricite or perovskite) and used as proxy for molten metal–molten silicate partitioning (Asahara et al., 2007; Frost et al., 2010; Kawazoe and Ohtani, 2006; O'Neill et al., 1998; Ozawa et al., 2008; Rubie et al., 2004; Takafuji et al., 2005). All these studies agree on a significant increase of oxygen solubility with temperature. Early studies concluded that pressure has a negative effect on the solubility of oxygen in metal (Li and Agee, 2001; O'Neill et al., 1998; Rubie et al., 2004). However, more recent studies suggested the existence of two pressure regimes, with a decrease of oxygen concentration in the metal up to 10 GPa and subsequently a moderate increase at higher pressures (Asahara et al., 2007; Frost et al., 2010). Overall, these studies suggest that the effect of pressure remains moderate in comparison to that of temperature.

As first mentioned by Birch (1964), the core density deficit could be accounted for by a combination of several light elements, rather than just one. Thus, it is important to understand the interactions between proposed light elements during core formation. Silicon and oxygen are major elements in the bulk Earth, and were readily available in large quantities during all stages of terrestrial accretion. It is therefore fundamental to study their partitioning between molten metal and silicate melts at pressure, temperature and redox conditions relevant to core formation. To date, the coupled behaviour and solubility of silicon and oxygen in molten metal have only been studied in association with a single crystallised mineral phase, perovskite, whereas realistic conditions of core–mantle differentiation in a magma ocean are that of a molten silicate with peridotitic composition. Available solubility data (Ozawa et al., 2009; Sakai et al., 2006; Takafuji et al., 2005) suggest that silicon and oxygen could be the main light elements, but uncertainties regarding equilibrium and oxygen fugacity (f_{O_2}) conditions in these extremely high-pressure experiments prevent the drawing of definitive conclusions. In order to constrain better the Si and O contents of the Earth's core, we need to measure their partition coefficients between molten metal and molten silicate as a function of composition, P, T, f_{O_2} conditions, since core formation probably happened continuously, with changing metal–silicate equilibration depths, temperatures, and redox conditions (e.g., Corgne et al., 2008; Wade and Wood, 2005).

The objective of this experimental study was to derive partition coefficients for oxygen and silicon at conditions that prevailed during core formation in a primordial magma ocean. High-pressure high-

temperature melting experiments were performed to collect the required data, which were integrated in a thermodynamic model to parameterize partitioning and chemical interactions as a function of key variables. Our data are used to answer ambiguities emerging from previous studies and to derive plausible ranges of Si and O contents in the core for proposed core formation models.

2. Methods

2.1. Experimental procedure

Melting and equilibration experiments were performed using two synthetic compositions prepared from high purity powders of oxides and iron metal. The first starting material (SM1) is a 1:1 mixture (weight ratio) of pure iron metal and an $MgSiO_3$ composition prepared by mixing MgO with SiO_2 . The second starting material (SM2) is a mixture (1:1 weight ratio) of pure iron metal and oxides (MgO, Fe_2O_3 and SiO_2 in 0.70:0.15:1 relative molar proportions). The starting powders were pre-homogenised under ethanol and stored in an oven at 100 °C.

Melting experiments were carried out between 2273 and 2873 K and 2 and 21 GPa, using an end-loaded piston-cylinder apparatus and a 1500-ton multi-anvil press at the Geophysical Laboratory. In all runs, starting mixtures were contained in MgO capsules prepared from polycrystalline magnesia rods. For experiments performed at 2 GPa and 2273 K, we used a graphite furnace surrounded by a Pyrex glass and talc as pressure medium. For experiments at 2573 K and 2873 K, we used a piston-cylinder cell assembly similar to the one described in Cottrell and Walker (2006) with $LaCrO_3$ as thermal insulator. For experiments at 7 GPa and 2273 K, we used the 18/11 assembly as described in Corgne et al. (2007). For experiments at 2573 K and 2873 K, we used the same 18/11 assembly but the outer zirconia sleeve was replaced by a thicker $LaCrO_3$ sleeve for better thermal insulation. For experiments at 14 GPa and 21 GPa, we used the 10/5 and 8/3 assemblies respectively, as described by Bertka and Fei (1997).

Temperatures in all experiments were measured using a W5%Re/W26%Re thermocouple placed directly above the MgO capsule. The samples were heated at a rate of 100 K/min to the target temperature, held there for 2 min and quenched by turning off the power to the furnace. Run duration of 2 min ensured approach of equilibrium at such high temperatures (as evidenced from flat chemical profiles across the run products; see Fig. S2), while limiting interaction of the sample with the MgO capsule. Runs showing W or Re contamination of the metallic phase were discarded and redone.

Retrieved samples were mounted in epoxy resin and carefully polished using a suite of diamond powder grit from 150 μm to 0.25 μm to avoid SiC contamination. Polishing using regular SiC sheets and an alumina suspension was observed to lead to significant implantation of Si-rich particles in the metallic phase.

2.2. Analytical procedure

Chemical analyses were performed on carbon-coated samples with a JEOL 8900 electron microprobe at the Geophysical Laboratory using a 15 kV acceleration voltage and a 30 nA probe current. Standards used for analysis of the metallic phase were pure Fe metal, pure Si metal and magnetite (Fe_3O_4) for Fe, Si and O, respectively; for analysis of molten silicate and ferroperricite, standards were enstatite for Si, Mg and O, and magnetite for Fe. When analysing O in the metal phase, we applied a background correction determined from pure iron standard measurement before and after each unknown. The magnitude of the correction was $0.15 \text{ wt}\% \pm 0.02$ (1sd). Microprobe analyses of the silicate melts were performed using a scan window of $10 \times 10 \mu m$ to average the heterogeneous quench texture. As done in previous studies (Gessmann and Rubie, 1998; O'Neill et al., 1998), we used image analysis to obtain the bulk composition of the molten metal (prior to quench), which integrates the individual

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