



Melting in the FeO–SiO₂ system to deep lower-mantle pressures: Implications for subducted Banded Iron Formations

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ABSTRACT

Banded iron formations (BIFs), consisting of layers of iron oxide and silica, are far denser than normal mantle material and should have been subducted and sunk into the deep lower mantle. We performed melting experiments on Fe₂SiO₄ from 26 to 131 GPa in a laser-heated diamond-anvil cell (DAC). The textural and chemical characterization of a sample recovered from the DAC revealed that SiO₂ is the liquidus phase for the whole pressure range examined in this study. The chemical compositions of partial melts are very rich in FeO, indicating that the eutectic melt compositions in the FeO–SiO₂ binary system are very close to the FeO end-member. The eutectic temperature is estimated to be 3540 ± 150 K at the core–mantle boundary (CMB), which is likely to be lower than the temperature at the top of the core at least in the Archean and Paleoproterozoic eons, suggesting that subducted BIFs underwent partial melting in a thermal boundary layer above the CMB. The FeO-rich melts formed by partial melting of the BIFs were exceedingly dense and therefore migrated downward. We infer that such partial melts have caused iron enrichment in the bottom part of the mantle, which may have contributed to the formation of ultralow velocity zones (ULVZs) observed today. On the other hand, solid residues left after the segregation of the FeO-rich partial melts have been almost pure SiO₂, and therefore buoyant in the deep lower mantle to be entrained in mantle upwellings. They have likely been stretched and folded repeatedly by mantle flow, forming SiO₂ streaks within the mantle “marble cake”. Mantle packages enhanced by SiO₂ streaks may be the origin of seismic scatterers in the mid-lower mantle.

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1. Introduction

BIFs are distinctive sedimentary rocks characterized by alternating layers of iron oxides and silica. They occur widely in Archean to Paleoproterozoic terrains all over the world with exposures reaching 1200 m in thickness and several hundred kilometers in extent (see Klein, 2005 for a review). The oldest BIF is found in the 3.8-Ga Isua supracrustal belt, West Greenland. According to Klein (2005), BIFs have been one of the major crustal components of Archean to Paleoproterozoic cratons formed at ~3.5 to ~1.8 Ga. BIF deposition rates have peaked at ~2.5 Ga when the Hamersley Group in Western Australia was formed. Although the detailed mecha-

nism of BIF formation remains unclear, it is robust that they have been formed as a consequence of oxidation of ferrous iron dissolved in seawater. The formation of BIFs stopped at 1.8 Ga, and afterwards has been limited to occasional appearances between 0.8 and 0.6 Ga. The amount of preserved BIFs on the present-day Earth is estimated to be about 10^{14–15} tons (Isley, 1995). However, these preservations on cratons are very likely only a small part of the total BIFs ever generated, because at least any BIF generated on oceanic crust should have been conveyed into the deep interior of the Earth through at subduction (Hopkins et al., 2008) or non-plate-tectonic delamination processes (Harris and Bédard, 2014).

Dobson and Brodholt (2005) argued that these subducted BIFs accumulate above the CMB and form the ULVZs. The BIFs are likely to be a mixture of FeO and SiO₂ near the base of the mantle where the oxidation state is near the iron-wüstite buffer. Dobson and Brodholt (2005) approximated the BIFs to be pure FeO, whose

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Table 1

Experimental conditions of melted/not melted samples.

Melted				Not melted			
Run #	P (GPa)	T (K)	Insulator	Run #	P (GPa)	T (K)	Insulator
1	49	not measured	Ar	3	80	2270	SiO ₂
2	26	2430	none	4	67	2050	SiO ₂
5	33	2680	Ar	6	81	2730	SiO ₂
11	123	3730	SiO ₂	7	33	2200	SiO ₂
				8	116	2760	SiO ₂
				9	131	3350	SiO ₂
				10	101	2180	SiO ₂

melting temperature was originally reported to be >5000 K at the CMB (Knittle and Jeanloz, 1991), and discussed the possibility that the subducted BIFs remain in a solid state in the mantle. However, the more recent work by Fischer and Campbell (2010), based on measurements up to 77 GPa and the Lindemann's prediction, found that FeO undergoes melting at temperatures above 3690 K at the CMB. This estimate is much lower than the estimates by Knittle and Jeanloz (1991) and Seagle et al. (2008) but consistent with Shen et al. (1993). More importantly, the subducted BIFs are indeed a mixture of FeO and SiO₂, and thus melting occurs at the eutectic temperature in the FeO–SiO₂ binary system, which must be lower than the melting temperature of the FeO end-member.

A melting phase diagram in the FeO–SiO₂ binary system so far has only been investigated at pressures below 17 GPa, using a multi-anvil apparatus (Ohtani, 1979; Kato et al., 1984). These previous studies show that FeO wüstite and SiO₂ stishovite form a eutectic system above 17 GPa.

In this study, we performed melting experiments on Fe₂SiO₄ between 26 and 131 GPa in a laser-heated DAC. The solidus temperature of Fe₂SiO₄, corresponding to the eutectic temperature in the FeO–SiO₂ binary system, was determined from the textural and compositional analyses of DAC samples recovered after pressure release. Our results suggest that the subducted BIFs underwent partial melting near the base of the mantle, which is likely to have contributed to the enrichment in iron above the CMB.

2. Experimental procedures

Melting experiments were carried out at high pressure by using laser-heated DAC techniques. Synthetic Fe₂SiO₄ fayalite was used as a starting material. This composition was chosen because the eutectic melt composition is expected to be FeO-rich rather than SiO₂-rich from the melting temperatures of FeO and SiO₂ end-members. The chemical composition and the homogeneity of the starting material were confirmed by electron microprobe analysis. Either Ar or SiO₂ glass was used both as a thermal insulator and as a pressure medium. They were loaded into a sample chamber, a hole drilled on a pre-indented Re gasket. The culet size of diamond anvils was 120 to 300 μm depending on a target pressure. After loading, the whole DAC was dried in a vacuum oven at 423 K for >12 h. We then introduced Ar gas and quickly started compression.

After compression to a pressure of interest, the sample was heated from both sides with a couple of single-mode Yb fiber lasers. Temperature was obtained by a spectroradiometric method (Ohishi et al., 2008; Nomura et al., 2014). The highest temperature measured during heating represents an experimental temperature in each run. Temperature within a melt pocket was relatively homogeneous, but the temperature at melt/solid interface was <150 K lower than the maximum temperature of a sample. It means that temperatures only of 'melted' samples are overestimated by that degree. Pressure in the sample chamber was determined from the Raman spectrum of a diamond anvil at room

temperature after heating (Akahama and Kawamura, 2006). We consider a contribution of thermal pressure of +11.6% per 1000 K for a sample with no pressure medium and samples using SiO₂ as pressure medium on the basis of *in-situ* X-ray diffraction measurements on a mixture of Fe₂SiO₄ and gold (Fei et al., 2007) at BL10XU, SPring-8. For a couple of runs with Ar pressure medium, we added thermal pressure of 1 GPa per 1000 K according to Dewaele et al. (2007).

After heating at high pressure, samples were recovered from the DAC and processed for textural and chemical characterizations. Cross sections of the sample were obtained with an Ion Slicer (JEOL EM-09100IS) (Tateno et al., 2009) or Focused Ion Beam (FIB, FEI Versa 3D at Tokyo Tech or FEI Quanta 200 3DS at Kyoto University) and subsequently examined by field-emission-type electron microprobe (FE-EPMA, JEOL JXA-8530F).

3. Results

We have carried out a total of eleven separate runs in a pressure range from 26 to 131 GPa (Table 1). In experiments performed at relatively high temperatures (runs #1, 2, 5, 11), an FeO-rich homogeneous area was observed at the center of a heated spot (Fig. 1a, b). Such an FeO-rich part was round and exhibited a non-stoichiometric chemical composition. We therefore interpreted it to be a quenched partial melt. Note that the enrichment in FeO in a hot area is not produced by the Soret effect (Sinmyo and Hirose, 2010), which induces the migration of FeO toward a low-temperature region. In addition, coarse SiO₂ crystals were found coexisting with the FeO-rich quenched partial melt (Fig. 1a, b). Such an SiO₂ phase should be the liquidus (the first crystallizing) phase of coexisting partial melts.

On the other hand, the FeO-dominant region was not observed when the sample was heated to relatively low temperatures. As an alternative, a heated portion occurred as a mixture of fine-grain FeO and SiO₂ (Fig. 1c), demonstrating a decomposition of Fe₂SiO₄ fayalite starting material into mixed oxides at subsolidus conditions. We did not find FeSiO₃ phase in recovered samples.

With the existence/non-existence of an FeO-rich non-stoichiometric round portion (a melt pocket) and coexisting coarse SiO₂ grains as melting criteria, the solidus curve of Fe₂SiO₄ composition was obtained (Fig. 2). Since the solidus temperature of Fe₂SiO₄ is equivalent to a eutectic temperature in the FeO–SiO₂ binary system, the upper bound for the solidus curve of Fe₂SiO₄ is given by the melting curve of FeO end-member, determined by Fischer and Campbell (2010) and extrapolated to high pressures on the basis of Simon's equation. The lower bound corresponds to our 'not melted' data. These results show that the eutectic temperature is 3540 ± 150 K at the CMB. In runs using silica as an insulator, the bulk composition may have been slightly SiO₂-rich compared to fayalite. However, as far as the system is eutectic, it does not change the solidus temperature.

The chemical compositions of partial melts were determined by microprobe analyses from four runs (Table 2). It was often difficult to obtain a melt composition without contamination from

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