



# The melting of subducted banded iron formations



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## ABSTRACT

Banded iron formations (BIF) were common shelf and ocean basin sediments 3.5–1.8 Ga ago. To understand the fate of these dense rocks upon subduction, the melting relations of carbonated BIF were determined in Fe–Ca–(Mg)–Si–C–O<sub>2</sub> at 950–1400 °C, 6 and 10 GPa, oxidizing ( $f_{O_2}$  = hematite–magnetite, HM) and moderately reducing ( $f_{O_2}$  ~ CO<sub>2</sub>–graphite/diamond, CCO) conditions. Solidus temperatures under oxidizing conditions are 950–1025 °C with H<sub>2</sub>O, and 1050–1150 °C anhydrous, but 250–175 °C higher at graphite saturation (values at 6–10 GPa). The combination of Fe<sup>3+</sup> and carbonate leads to a strong melting depression. Solidus curves are steep with 17–20 °C/GPa. Near-solidus melts are ferro-carbonatites with ~22 wt.% FeO<sub>tot</sub>, ~48 wt.% CO<sub>2</sub> and 1–5 wt.% SiO<sub>2</sub> at  $f_{O_2}$  ~ HM and ~49 wt.% FeO<sub>tot</sub>, ~20 wt.% CO<sub>2</sub> and 19–25 wt.% SiO<sub>2</sub> at  $f_{O_2}$  ~ CCO.

At elevated subduction geotherms, as likely for the Archean, C-bearing BIF could melt out all carbonate around 6 GPa. Fe-rich carbonatites would rise but stagnate gravitationally near the slab/mantle interface until they react with the mantle through Fe–Mg exchange and partial reduction. The latter would precipitate diamond and yield Fe- and C-rich mantle domains, yet, Fe–Mg is expected to diffusively re-equilibrate over Ga time scales. We propose that the oldest subduction derived diamonds stem from BIF derived melts.

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## 1. Introduction

Banded iron formations (BIF) are composed of alternating bands of Fe-oxides and cherts. These peculiar sediments precipitated primarily in the Archean and Paleoproterozoic 3.5–1.8 Ga ago (Konhauser et al., 2007). Before the rise of oxygen (2.3–2.1 Ga ago), oceans were reducing and contained large amounts of soluble ferrous (Fe<sup>2+</sup>) iron, episodic seawater oxidation led then to Fe(III)-precipitation and BIF sedimentation. The rise of oxygen ended BIF formation, except for the glaciation related Neoproterozoic Rapitan type BIF (0.8–0.6 Ga ago). BIF constitute the world's most important high grade iron ores, bulk iron oxide contents reaching up to 85 wt.%.

Most of the preserved BIF are shallow water marine deposits up to several hundred meters thick (Klein, 2005). They are believed to form through interaction between oxidized surface and upwelling Fe-rich, reduced deep ocean waters, nevertheless, a permanent redox front may have existed across entire ocean basins, not just on continental shelves (Beukes and Gutzmer, 2008). BIFs were also deposited in deep basins beyond the range of epiclastic

offshore influx, resulting in very low Al-contents (Trendall, 2002). At the time, BIFs constituted a major basin sediment, typically intercalated with anoxic C-rich black shales and Fe-rich carbonates. The thickness of deep-ocean basin BIFs may amount to several 100 m as is the case in the Hamersley province, Western Australia (Pickard et al., 2004). As most ocean basin sediments, they were recycled into the Earth's interior by subduction (Dilek and Polat, 2008), thought to occur since the Archean (Kusky et al., 2001).

The reducing oceans have produced the arguably most oxidized sediment in Earth history, yet carbon in these sediments was originally mostly organic (Beukes et al., 1989). Upon low grade metamorphism (~0.3 GPa, 200 °C), organic carbon reacts with Fe(III) to form ankerite or siderite. Due to their high iron content, BIF are denser than any other part of the slab or mantle, hence would rarely re-surface after subduction. BIF may in fact only gravitationally stabilize at the core-mantle boundary where they have been postulated to form the ultralow-velocity zones (Dobson and Brodholt, 2005).

Subducted BIF form an extreme geochemical anomaly in the Earth's mantle, they have an  $X_{Mg} < 0.22$  (Klein, 2005), are oxidized to the magnetite–hematite (HM) buffer and are quartz-saturated. They are essentially composed of magnetite + hematite + quartz ± siderite and may contain minor ankerite, kerogene or graphite, stilpnomelane. Carbon may stem from carbonate precipitation or

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**Table 1**  
Bulk compositions.

	Oxidized ( $fO_2 \sim \text{HM}$ )				Reduced ( $fO_2 \sim \text{CCO}$ )		
	BH1	BH2	BH3	BD	BG1	BG2	BG3
SiO <sub>2</sub> (wt.%)	11.54	10.96	25.80	9.62	16.65	19.58	33.42
FeO	18.63	19.36	16.14	15.53	26.87	35.12	29.07
Fe <sub>2</sub> O <sub>3</sub>	49.07	46.61	38.84	40.92	26.55	31.22	25.85
MgO	–	–	–	3.63	–	–	–
CaO	8.08	8.98	7.48	11.79	11.65	5.48	4.54
CO <sub>2</sub>	12.68	14.09	11.74	18.50	18.29	8.60	7.12
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Molar ratios							
Fe <sup>3+</sup> /(Fe <sup>3+</sup> + Fe <sup>2+</sup> )	0.703	0.684	0.684	0.703	0.471	0.444	0.444
Mg/(Mg + Fe <sup>2+</sup> )	–	–	–	0.294	–	–	–
Ca/(Ca + Fe <sup>2+</sup> )	0.357	0.373	0.373	0.493	0.357	0.167	0.167

from organic matter, bulk isotope compositions are frequently  $\delta^{13}\text{C} \sim -10\text{‰}$  suggesting a mixture of both (Beukes et al., 1989; Walker, 1984). Carbonate contents may amount to 1/10 and even up to 1/2 of the rock (Walker, 1984; Webb et al., 2003). The fate of these ancient sediments upon subduction is the object of this study. We determine the mineralogy, phase relations, melting conditions and melt compositions of C-bearing BIF at 6–10 GPa. The complete range of relevant oxygen fugacities is covered, i.e. from  $fO_2 = \text{hematite-magnetite}$ , where all organic carbon would have reacted to siderite or ankerite, to moderately reducing conditions where organic or elemental carbon would dominate over hematite, resulting in an  $fO_2$  near the graphite-CO<sub>2</sub> (CCO) buffer.

### 1.1. Phase relations in FeO–SiO<sub>2</sub>–O<sub>2</sub>, the most simple BIF model system

#### 1.1.1. Melting at atmospheric pressure

At 1 atm, eutectic melt with 18 wt.% SiO<sub>2</sub> occurs on the magnetite–SiO<sub>2</sub> join at 1455 °C (Phillips and Muan, 1959). For hematite–SiO<sub>2</sub>, slightly higher melting temperatures are expected, but still below 1500 °C (Osborn and Muan, 1960). At more reducing conditions, magnetite + fayalite + cristobalite melt at 1140 °C in an eutectic situated towards the Fe<sub>2</sub>SiO<sub>4</sub> composition. BIFs have Fe<sup>3+</sup>/∑Fe ranging from 0.9 to 0.23, i.e. from between hematite–magnetite to magnetite–fayalite (Horstmann and Halbach, 1995).

#### 1.1.2. Subsolidus phase relations and melting at high pressures

With pressure, ortho-ferrosilite stabilizes instead of fayalite + quartz above 1.6 GPa (Akimoto et al., 1964) and melts incongruently to fayalite-rich liquid + quartz. Melting temperatures of 1285–1480 °C (at 1.6–5 GPa, Lindsley, 1965) are near the mantle adiabat (1400 °C at 5 GPa). Ortho-converts to clino-ferrosilite at 5–7.5 GPa and 840–1400 °C (Woodland and Angel, 1997), which decomposes to stishovite + Fe-ringwoodite at  $\geq 8.75$  GPa, 800–1200 °C (Ringwood and Major, 1966). The  $\alpha$ - to  $\gamma$ -Fe<sub>2</sub>SiO<sub>4</sub> transformation occurs at 4–7 GPa, 700–1200 °C (Akimoto et al., 1965). In the Fe<sub>2</sub>SiO<sub>4</sub>–Fe<sub>3</sub>O<sub>4</sub> binary system, three intermediate spineloid phases occur, solid solution is complete above 8 GPa and 1100 °C (Woodland and Angel, 2000). Eventually,  $\gamma$ -Fe<sub>2</sub>SiO<sub>4</sub> decomposes to wustite + stishovite at 17–18.5 GPa (Ohtani, 1979). Melting temperatures in Fe–Si–O<sub>2</sub> at  $>10$  GPa are only known for Fe<sub>2</sub>SiO<sub>4</sub> and amount to 2000–2200 °C (Ohtani, 1979).

### 1.2. BIF + carbon at high pressure

As C-free BIF systems are unlikely to melt in the Earth, Fe-rich carbonates become crucial, in particular as minimum melting above 2.5–5 GPa typically results in carbonate melts. Such a behavior is observed in carbonated pelites (Grassi and Schmidt, 2011), basalts (Dasgupta et al., 2004; Yaxley and Brey, 2004), and peridotites (Dasgupta and Hirschmann, 2006; Brey et al., 2008; Ghosh et al., 2009).

BIFs frequently contain Fe-carbonates, most commonly siderite or dolomite–ankerite (Horstmann and Halbach, 1995; Klein, 2005; Beukes and Gutzmer, 2008). Siderite most probably forms through dissimilatory ferric-oxyhydroxide reduction in the presence of organic carbon (Beukes and Gutzmer, 2008). Subsolidus reactions in Fe–C–O<sub>2</sub> have been studied to 1.5 GPa, 700 °C (French, 1971; Weidner, 1972; Koziol, 2004). Melting of siderite has been studied to 20 GPa (Tao et al., 2013; Shatskiy et al., 2014; Kang et al., 2015), carbonate melting only becoming stable (instead of a subsolidus decarbonation) at 7 GPa, 1460 °C increasing to 1875 °C at 20 GPa. Slightly Mg-bearing siderite ( $X_{\text{Mg}} > 0.2$ ) melts at 1250 °C, 3.5 GPa (Kang et al., 2016), also Ca–Fe-carbonates, at least to an  $X_{\text{Fe}}$  of  $\sim 0.62$  melt at 3.5 GPa, 1120 °C (Franzolin, 2010). Melting in the Ca–Mg carbonate system is peritectic at 3 GPa (Irving and Wyllie, 1975) and includes a minimum at 1350 °C, 6 GPa almost at dolomite composition (Buob et al., 2006).

The system Fe–Si–C–O<sub>2</sub> has been thermodynamically modeled, siderite stability is maximal between the CCO and QFM (quartz–fayalite–magnetite) or FsQM (ferrosilite–quartz–magnetite) buffers where all C is oxidized and Fe present as Fe<sup>2+</sup> (Connolly, 1995; Kang et al., 2015). Higher oxygen fugacities lead to rapid destabilization of siderite, but ternary Fe–Mg–Ca-carbonates are less affected.

## 2. Experimental and analytical methods

### 2.1. Starting material

In our experiments oxygen fugacity was internally buffered by adjusting oxygen contents for saturation with hematite + magnetite ( $fO_2 = \text{HM}$ ) or graphite/diamond + carbonate mineral or melt ( $fO_2$  close to CCO). Bulk compositions (Table 1) were simplified into Fe–Ca–Si–C–O<sub>2</sub> and saturated in CaFe(CO<sub>3</sub>)<sub>2</sub> carbonate, SiO<sub>2</sub> (coesite/stishovite) and Fe<sub>3</sub>O<sub>4</sub> (magnetite) at subsolidus conditions. During the course of this study, bulk compositions had to be multiply adjusted to achieve saturation in five minerals at the then univariant solidus. Only such multiply saturated melts correspond to true minimum melts that could be eutectic or azeotropic as in the CaCO<sub>3</sub>–FeCO<sub>3</sub> binary at 3.5 GPa (Franzolin, 2010). Starting materials were made of powders of SiO<sub>2</sub>, Fe<sup>0</sup> or Fe<sub>3</sub>O<sub>4</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaCO<sub>3</sub> and FeCO<sub>3</sub>. The carbonates introduce the desired amount of CO<sub>2</sub> and were high purity CaCO<sub>3</sub> (99.997%), natural, almost pure siderite from Greenland ( $X_{\text{Fe}} = 0.947$ ), or synthetic FeCO<sub>3</sub>. Magnetite was synthesized in a 1-atm gas-mixing furnace at 1200 °C and  $\log fO_2 = -7$  and drop-quenched into distilled water. Alternatively, a molar 20:80 metal:hematite mixture was used. Components were dried, mixed, homogenized and ground under ethanol/acetone using an agate mortar. The starting materials were then kept in an evacuated glass desiccator and dried under a heat lamp before each use. Siderite turned out to be sensitive to oxidation even

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