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Melting and subsolidus phase relations in peridotite and eclogite systems with reduced C–O–H fluid at 3–16 GPa

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article info abstract

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Melting phase relations of peridotite and eclogite coexisting with reduced C–O–H fluid have been studied at 3-16 GPa and 1200-1600 \degree C. In order to perform these experiments the double-capsule technique with fO_2 control by outer Mo–MoO₂ or Fe–FeO buffer capsule was designed and developed for multianvil experiments at pressures 3–21 GPa. Silicate phase assemblages resemble those in volatilefree lithologies, i.e. olivine/wadsleyite–orthopyroxene–clinopyroxene–garnet in peridotite and garnet– omphacite in eclogite. Melting was detected by the appearance of quenched crystals of pyroxene, feldspar and glassy silica. Estimated solidus temperatures for peridotite $+$ C–O–H fluid with $fO₂$ = Fe–FeO are 1200 °C at 3 GPa and 1700 °C at 16 GPa. The solidus of the system with $fO_2 = Mo-MO_2$ was about 100 °C lower. Estimated solidus temperatures for eclogite + C–O–H fluid with $fO₂$ = Fe–FeO are 1100 °C at 3 GPa and 1600 °C at 16 GPa, and for eclogite at $fO_2 = Mo-MoO_2$ solidus temperatures were 20–50 °C lower. These solidus temperatures are much higher (300–500 ◦C) than those for peridotite and eclogite systems with H₂O and/or CO₂, but are still 300–400 °C lower than the solidi of volatile-free peridotite and eclogite at studied pressures. The compositions of partial melt were estimated from mass-balance calculations: partial melts of peridotite have CaO-poor (6–9 wt.%) basaltic compositions with 44–47 wt.% SiO₂ and 1.1-1.6 wt.% Na₂O. Melts of eclogite contain more SiO₂ (47-49 wt.%) and are enriched in CaO (9–15 wt.%), Na₂O (9–14 wt.%), and K₂O (1.3–2.2 wt.%). All runs contained graphite or diamond crystals along with porous carbon aggregate with micro-inclusions of silicates indicating that reduced fluid may dissolve significant amounts of silicate components. Analyses of carbon aggregates using a defocused electron microprobe beam reveal compositions similar to estimated partial melts. The diamonds formed from reduced C–O–H fluid may have natural analogues as polycrystalline diamonds. The oxygen fugacity in the Earth's mantle decreases with pressure from about fayalite–magnetite–quartz at shallow depths of 20–50 km to about iron–wustite at 250–300 km according to $fO₂$ estimations from cratonic peridotite. We show significant increase of solidus temperatures in peridotite and eclogite coexisting with reduced CH_4-H_2O fluid relative to the systems with oxidized H_2O – CO_2 fluid. We emphasize that redox melting by change of oxidation state across a mantle section, a phase transition, or the lithosphere–asthenosphere boundary can be the dominant melting process in the deep Earth's interior.

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1. Introduction

The major source of mantle heterogeneity is the subduction of oceanic crust, which transports oxidized and volatile-bearing sediments and altered basalts to great depths, at least during Phanerozoic time. A significant part of this material, especially volatile and incompatible elements, is involved in devolatilization and melting in the mantle wedges [\(Poli and Schmidt, 2002;](#page--1-0) [Gorman et al., 2006; Hacker, 2008; van Keken et al., 2011\)](#page--1-0). In turn,

some portion of these materials are erupted as volcanic rocks and attached to subcontinental crust in island arcs, whereas others are returned back into the convecting mantle. Melting beneath ridges and hot spots can also create heterogeneities in the both lithosphere and asthenosphere in the form of depleted domains and deep cumulates of pyroxenite and eclogite [\(Sobolev et al., 2007\)](#page--1-0). The hidden source for heterogeneities is the core, which may particularly emit volatiles into the lower mantle. Low mixing rates of mantle materials indicate that heterogeneities can survive through Earth history on a timescale of billions of years (e.g. [Anderson,](#page--1-0) [2006\)](#page--1-0). Signs of the heterogeneities in the lithospheric mantle are, for example, pyroxenite and eclogite xenoliths in basalts and kimberlites, and those in deep mantle may be seen as prominent seismic reflectors of various scales (e.g. [Stixrude, 2007\)](#page--1-0).

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The role of volatiles, mainly in the C–O–H system, below levels of magma generation ca. 40–200 km, is poorly understood. Redox states of peridotite xenoliths from kimberlite indicate that *f*O2 would gradually decrease with depth due to increasing solubility of $Fe³⁺$ in mantle silicates [\(Woodland and Koch, 2003;](#page--1-0) [Frost and McCammon, 2008\)](#page--1-0). Besides, relatively reduced conditions in the mantle (more reduced than redox state of subsurface volcanic rocks) are dictated by the conditions of separation of the metallic core, i.e. the equilibrium between molten/solid iron and silicates, and by melting processes, in which more oxidized species are partitioned into the melt or fluid phase. Estimations of mantle *f*O2 during core formation are from 2 to 4 log units below iron–wustite (IW) oxygen buffer (hereafter, IW-2–IW-4) [\(Frost et](#page--1-0) [al., 2008\)](#page--1-0). The C–O–H fluid composition at $fO₂$ near IW would be significantly different from oxidized H_2O – CO_2 fluids, and would correspond to CH_4-H_2O or CH_4-H_2 [\(Ballhaus and Frost, 1994;](#page--1-0) [Frost and McCammon, 2008\)](#page--1-0). Thus, it is extremely important to model high-pressure phase equilibria of mantle rocks coexisting with reduced fluid compositions [\(Jakobsson and Holloway, 1986;](#page--1-0) [Taylor and Green, 1987; Taylor, 1990\)](#page--1-0).

The high-pressure experiment is the key instrument to study volatile behavior in the deep mantle. Most experiments were conducted with oxidized volatiles, $H₂O$ and $CO₂$, and covered a pressure range up to 30–32 GPa [\(Wyllie and Huang, 1976; Green](#page--1-0) [and Wallace, 1988; Wallace and Green, 1988; Green and Falloon,](#page--1-0) [1998; Wyllie and Ryabchikov, 2000; Williams and Hemley, 2001;](#page--1-0) [Poli and Schmidt, 2002; Luth, 2003; Schmidt and Poli, 2003;](#page--1-0) [Dasgupta et al., 2004; Dasgupta and Hirschmann, 2006; Foley et](#page--1-0) [al., 2009; Litasov and Ohtani, 2002, 2009b, 2010; Litasov, 2011;](#page--1-0) [Litasov et al., 2011\)](#page--1-0). Also, significant effort has been expended to address hydrogen solubility in nominally anhydrous silicates as a function of *P*–*T* –*X*– *f* O2 parameters [\(Bolfan-Casanova, 2005;](#page--1-0) [Litasov and Ohtani, 2007; Withers and Hirschmann, 2008; Litasov](#page--1-0) [et al., 2009; Sokol et al., 2010\)](#page--1-0).

Experimental studies with reduced C–O–H fluid have been performed in various systems at pressures up to 6–7 GPa using different modifications of a double-capsule method. This method is used to prevent the escape of hydrogen, and may be other reduced species, from the sample capsule, which may cause significant sample oxidation and violation of equilibrium [\(Eugster, 1957;](#page--1-0) [Huebner, 1971; Whitney, 1972; Boettcher et al., 1973; Jakobsson](#page--1-0) [and Holloway, 1986; Taylor and Green, 1987; Foley, 1989; Taylor](#page--1-0) [and Foley, 1989; Taylor, 1990; Sokol et al., 2009, 2010\)](#page--1-0). The double-capsule method allows dynamic control of $fO₂$, $fH₂$, and $fH₂O$ in the experiment. This method can be described as follows. A welded outer capsule contains sufficient amounts of an oxygen buffer, e.g. Fe–FeO, and H_2O , which can be added as talc, brucite, or liquid water. A welded or pressure-sealed inner capsule contains the sample and a fluid source, which produces a fluid of certain composition. The fluid source can consist of different compounds producing $C-O-H$ fluid by thermal decomposition. $H₂O$, graphite, stearic acid and silver oxalate are typical sources for components of the C–O–H fluid. Ideally, fluid in the outer and inner capsule would correspond to the conditions of $aH_2O \approx 1$ and $aH_2O < 1$, respectively. The fO_2 is determined by the equilibrium $2H_2 + O_2$ $= 2H₂O$ at a level close to that of the included oxygen buffer by means of hydrogen exchange between buffer and inner sample capsule. The double capsule method can provide only so-called dynamic equilibrium, since buffer and fluid in the outer capsule and fluid in the inner capsule have limited "lifetime".

The composition of reduced C–O–H fluids in equilibrium with graphite/diamond has been investigated in several different systems. Modeling of C–O–H fluid in equilibrium with graphite and diamond + forsterite was performed at 2-7 GPa [\(Matveev et al.,](#page--1-0) [1997; Sokol et al., 2009, 2010\)](#page--1-0). The system peridotite–C–O–H at fO_2 controlled by WCWO buffer (WC–W–WO₂ = IW + 1) was studied at 0.9–3.6 GPa. In this system the $CH_4/(CH_4 + H_2O)$ ratio in the fluid decreases from 0.8 to 0.3 in the temperature interval from 1050 to 1250 \degree C at 1.5–3.6 GPa [\(Taylor and Green, 1988\)](#page--1-0). The solidus in the system with reduced CH_4-H_2O fluid is located at higher temperatures than in oxidized systems with H_2O and CO_2 . This is due to strong non-ideality of mixed CH_4-H_2O fluid and with low solubility of silicates in methane- and hydrogen-bearing fluid and low solubility of reduced fluid species in silicate melt. The behavior of $H₂O$ in reduced fluid is poorly understood. It was shown that the solidi of silicate–C–O–H systems are high even if the H2O content in the fluid approach 90 mol.% [\(Sokol et al., 2009,](#page--1-0) 2010), i.e. even if $fH₂O$ is higher than $aH₂O$ in hydrous silicate melt. In some cases the fluid composition measured in the experiments [\(Taylor and Green, 1988; Jakobsson and Oskarsson, 1990;](#page--1-0) [Matveev et al., 1997; Sokol et al., 2004\)](#page--1-0), is consistent with that calculated from equations of state for real gases (e.g. [Saxena and Fei,](#page--1-0) [1987; Zhang and Duan, 2009\)](#page--1-0), but in some others not. For instance, [Sokol et al. \(2009\)](#page--1-0) measured high concentrations of hydrogen and low methane in the quenched fluid, which is inconsistent with calculations.

In the work by [Jakobsson and Holloway \(2008\),](#page--1-0) melting of peridotite coexisting with C–O–H fluid was studied at 5–12 GPa and 1200–1500 \degree C. The fluid source $+$ buffer and sample were placed in the graphite capsules separated by Pt disc and then sealed to a single Pt capsule. These authors analyzed silicate melts compositions, but their solidus was located at much lower temperatures (below 1200 \degree C) in comparison to the work by [Taylor and Green](#page--1-0) [\(1988\).](#page--1-0)

In present work we extended studies of the systems with reduced C–O–H fluid to higher pressures and report melting phase relations for the peridotite and eclogite systems at 3–16 GPa and 1200–1600 ◦C using a modified double-capsule technique with *f*O2 controlled by outer Mo-MoO₂ or Fe-FeO buffer capsules. We determined the solidus temperatures and melt compositions and compare the results with those in systems with oxidized H_2O – CO_2 fluid. The results are applied to assess redox melting by changing oxidation state in deep mantle regions.

2. Experimental and analytical techniques

High-pressure experiments were performed using Kawai-type multianvil apparatuses at Tohoku University. The truncated edge length (TEL) of the tungsten carbide anvils was 12.0 (3–6.5 GPa) and 6.0 mm (12–16 GPa). We used modified double capsule methods [\(Sokol et al., 2009, 2010\)](#page--1-0), in which the outer capsule was made of thick-walled buffer material [\(Fig. 1\)](#page--1-0). Semi-sintered zirconia was used as the pressure medium for experiments. A cylinder of graphite (3–6.5 GPa) or LaCrO₃ (12–16 GPa) was used as the heater, and a MgO insulator was placed between the capsule and heater. The outer buffer capsule was not welded before experiments, but became nearly impermeable for $H₂O$ under pressure as confirmed by imaging of recovered samples. The sample was placed into AuPd or Pt capsules and separated from the outer capsule by a hydrogen transmitting medium (HTM) made of talc, which transforms to enstatite and H_2O -fluid or H_2O -bearing silicate melt upon heating.

The size of the cell for TEL 12 mm is ample for placing two 3 mm wide and 2.5 mm tall outer capsules, which are well separated from the inner capsules by HTM. However, experiments with the cell for TEL 6 mm were very difficult due to limited space in the cell [\(Fig. 1\)](#page--1-0). The outer capsule in this cell need to have sufficient wall and cap thicknesses (at least 0.2–0.3 mm) and should not be significantly deformed under compression to prevent rupture. We performed cell checks after each experiment including both inner and outer capsule conditions. First, we checked the separation of inner capsule from outer buffer capsule, which is Download English Version:

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