



Solidus of carbonated peridotite from 10 to 20 GPa and origin of magnesiocarbonatite melt in the Earth's deep mantle

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ABSTRACT

We have experimentally determined the solidus of an alkali-bearing carbonated peridotite (with 5 wt.% CO₂) between 10 and 20 GPa. Based on K-deficit in all low-temperature runs we assumed that some melt could be present in the low temperature runs and the true solidus of an alkali-bearing carbonated peridotite is placed below 1200 °C. However, based on the disappearance of magnesite and the appearance of the visible quenched melt coexisting with silicate phases, the 'apparent' solidus, which may be applicable for peridotite with low alkali contents, was identified. The 'apparent' solidus temperature increases from ~1380 °C at 10 GPa to ~1525 °C at 15 GPa and the 'apparent' solidus curve becomes almost flat from 15 GPa to 20 GPa, where it is located near 1550 °C. At 10 GPa, the 'apparent' solidus of carbonated peridotite is ~550 °C lower than the solidus of CO₂-free natural anhydrous peridotite. The solidus of the present study was also ~120 °C lower than the solidus determined by Dasgupta and Hirschmann [Dasgupta, R., Hirschmann, M.M., 2006. Melting in the Earth's deep upper mantle caused by carbon dioxide. *Nature*, 440, 659–662.] for natural carbonated peridotite. The drop in the solidus temperature is mainly due to the effect of alkalis (Na₂O, K₂O). The melt near the 'apparent' solidus has high CO₂ (>40 wt.%) and contains <6.0 wt.% SiO₂, <0.30 wt.% Al₂O₃ and <0.25 wt.% TiO₂. The composition of near-solidus partial melt is close to that observed at 6–10 GPa in the CMS-CO₂ and CMAS-CO₂ systems, and natural carbonated peridotite, with some variations in Ca/Mg-ratio. High alkali contents in measured and calculated partial melts are consistent with the compositions of deep-seated fluids observed as inclusions in diamonds and may be consistent with the compositions of parental melt, reconstructed for natural magnesiocarbonatite. We have demonstrated that magnesiocarbonatite-like melt can be generated by partial melting of carbonated peridotite at pressure up to at least 20 GPa. The generation of calciocarbonatite and ferrocyanatite is unlikely to be possible during melting of carbonated peridotite in the deep mantle.

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1. Introduction

Carbon dioxide is an important volatile constituent in the Earth's mantle. It drastically lowers the solidus temperature and changes the melting phase relations and melt compositions of mantle peridotite and eclogite (e.g. Wyllie, 1977; Wendlandt and Mysen, 1980; Wallace and Green, 1988; Falloon and Green, 1989; Yaxley and Green, 1994; Dalton and Presnall, 1998; Dasgupta et al., 2004; Dasgupta and Hirschmann, 2006; Brey et al., 2008). Recent experimental studies suggest that CO₂ plays an important role in the formation and dynamics of the Earth's mantle (e.g. Dasgupta and Hirschmann, 2006; Dasgupta et al., 2007; Ghosh et al., 2007).

It has been estimated that Earth's mantle contains very small amount of CO₂ in the range from 50 to 200 ppm (Marty and Tolstikhin, 1998; Saal et al., 2002). However, this value is quite uncertain due to very obscure understanding of carbon fluxes and carbon storage in the Earth's mantle and core (e.g. Jambon, 1994). CO₂ or carbonate may not be the dominant carbon-bearing phases in the mantle. According to the estimations of mantle oxidation state (e.g. Wood et al., 1996; Luth, 1999; Frost and McCammon, 2008) methane, diamond, and even some metal carbides could be preferentially stabilized in the Earth's mantle at pressures above 6 GPa. Nevertheless, CO₂ and carbonate can be important even in the deep mantle, for example, in oxidized subduction-related environments.

Thermal modeling of the subducting slabs indicates that a significant amount of carbonates can be transported to the deep mantle without decarbonation during melting beneath the island arcs (e.g. Bebout, 1995; Kerrick and Connolly, 2001). After decomposition of dolomite at about 4.5 GPa and 1000 °C, magnesite is a major carbonate phase, which stores oxidized carbon in mantle-related assemblies of peridotite and eclogite (e.g. Falloon and Green, 1989; Martinez et al.,

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1996; Dalton and Presnall, 1998; Hammouda, 2003; Yaxley and Brey, 2004; Dasgupta et al., 2004; Gudfinnsson and Presnall, 2005; Dasgupta and Hirschmann, 2006). Experimental study shows that magnesite itself is stable at 15 GPa and 2050 °C (Katsura and Ito, 1990). In the lower mantle, magnesite is stable at pressures over 100 GPa (magnesite transforms to high-pressure polymorph, magnesite II at pressures above ~115 GPa) and temperatures to 2700 °C (e.g. Katsura et al., 1991; Biellmann et al., 1993; Isshiki et al., 2004).

Carbonates have been observed rarely among mineral inclusions in diamonds (calcite by Meyer and McCallum, 1986, magnesite by Phillips and Harris, 1995 and Wang et al., 1996, and dolomite by Stachel et al., 1998). They are present in different fluid-bearing diamonds, which contain also abundant alkalis, chlorine and water and formed in the mantle at pressure above 4 GPa (e.g. Izraeli et al., 2004; Klein-BenDavid et al., 2007). Recently, Brenker et al. (2007) have found carbonate inclusions in ultra-deep diamonds from Juina (Brazil), which have presumably been transported from the transition zone and even from the lower mantle. Similar to mineral inclusions in diamond, carbonate minerals rarely occur in ultramafic mantle-derived xenoliths (e.g. Berg, 1986; Smith, 1987; Ionov, 1998) and the absence of carbonates in xenoliths is probably due to the decarbonation reaction with mantle peridotite minerals, such as dolomite + orthopyroxene = clinopyroxene + olivine + CO₂, responsible for the formation of wehrlite (e.g. Wyllie and Huang, 1975; Boyd and Gurney, 1986; Wallace and Green, 1988; Canil, 1990; Dalton and Wood, 1993).

Carbonatite and kimberlite magmas, which originate at pressures of at least 4–8 GPa, undoubtedly provide a proof of the significant amount of CO₂ in the Earth's mantle. Due to their low density and viscosity and highly reactive nature, carbonatite melts and fluids are considered to be one of the most important metasomatic agents in the lithospheric mantle (e.g. Navon et al., 1988; Bell, 1989).

Carbonatite is an igneous rock which contains more than 50 vol. % of carbonated minerals (e.g. dolomite, calcite, magnesite, etc.). These magmas generally contain low silica (0–10 wt.%) and high incompatible trace elements (e.g. Rb, Sr, U, Th and REE; average Sr and Nd contents are 7000 and 250 ppm, respectively), which make them unique among other igneous rocks. Carbonatites have been found at every continent and ranges in age from Archean to the present (e.g. Bell, 1989). Carbonatite magmas have been believed to be originated by low degree partial melting of garnet peridotite in the presence of CO₂ and/or H₂O in the upper mantle (e.g. Wyllie and Huang, 1975; Egger, 1976) or being separated from original carbonated silicate melt at subsurface by crystal fractionation or liquid immiscibility. Primary carbonatite magmas, can be separated from their source regions and migrate by infiltration even at melt fraction of 1% or less (e.g., McKenzie, 1989). Although naturally observed carbonatite are alkali-poor magma, with the exception of rare modern natrocarbonatite of Oldoinyo Lengai volcano (Tanzania), their parental magma is expected to be extremely alkali-rich. Therefore, alkali may play an important role in carbonatite petrogenesis.

Numerous experimental studies have been conducted on carbonated peridotite systems to understand the petrogenesis of carbonatites (e.g. Wyllie and Huang, 1975; Egger, 1976; Green and Wallace, 1988; Dalton and Presnall, 1998; Gudfinnsson and Presnall, 2005; Dasgupta and Hirschmann, 2006). It has been demonstrated that melting of carbonated lherzolite at 2–3 GPa produces CO₂- (>40 wt.%) and CaO-rich (>20 wt.%) and SiO₂- (<10 wt.%) and MgO poor (<20 wt.%) calcicarbonatitic melt (Green and Wallace, 1988; Wallace and Green, 1988; Thibault et al., 1992; Dalton and Wood, 1993; Dalton and Presnall, 1998). At pressures above 5 GPa near solidus melt becomes MgO-rich (>20 wt.%) and changes to magnesiocarbonatite in composition (e.g. Dasgupta and Hirschmann, 2006). Recently, Dasgupta and Hirschmann (2006) reported the solidus and melting phase relations of natural carbonated peridotite (2.5 wt.% CO₂) from 3 to 10 GPa and observed steep slope of the solidus, which may exceed 1500 °C at 10 GPa. Extrapolation of this solidus line to the greater depth indicates that magnesite can be stable at very high temperatures in the deep mantle.

In this study we present the melting phase relations of an alkali-bearing carbonated peridotite (5 wt.% CO₂) from 10 to 20 GPa and temperature between 1200 °C and 1630 °C to clarify the solidus of carbonated peridotite, melt composition, and alkali behavior during partial melting of carbonated peridotite to transition zone depths and discuss near solidus melt compositions with implication for the origin of carbonatite-like magma in the deep mantle.

2. Experimental procedures

2.1. Starting material

The starting material was a synthetic carbonated fertile peridotite, which is close to an average mantle pyrolite composition after Ringwood (1975) except the CO₂ and alkali contents (Table 1). We added 0.5 wt.% Na₂O and 0.4 wt.% K₂O in the starting material to monitor the behavior of alkalis during partial melting of peridotite. The starting composition was synthesized from high purity oxides and carbonates. All the oxides and carbonates compounds were dried in a one-atmosphere furnace at 900 °C and 300 °C for 12 h to remove water from them before mixing them under ethanol. Dasgupta et al. (2005) have shown that the addition of different carbonates in the base silicate rock has modest effect whereas the addition of large amount of calcite in the starting material may produce unrealistic results. Therefore, carbon dioxide was added as CaCO₃, Na₂CO₃, K₂CO₃ (reagent grade carbonates) and natural magnesite from Bahia, Brazil (mineral collection of Museum of Natural History, Tohoku University) with a composition of Mg_{0.975}Fe_{0.015}Mn_{0.006}Ca_{0.004}CO₃. Oxides were ground in an agate mortar under ethanol and then fired at 1400 °C in a CO₂/H₂ gas mixing furnace under controlled oxygen fugacity at log fO₂ = QFM – 1 to remove Fe⁺³. The quenched peridotite glass and minerals mixture were crushed and ground under ethanol, after that carbonates were added and finally, the entire mixture was ground again under ethanol to homogenize the mixture.

2.2. Multianvil apparatus

All experiments were carried out in a Kawai-type multi-anvil high-pressure apparatus (1000 ton press) at Tohoku University. Tungsten carbide cubes with 3.5 mm truncated edges and 26 mm edge length were used as the second-stage anvils for all the experiments. The cross section of a cell assembly is shown in Fig. 1. Zirconia was used as a pressure medium and cylindrical lanthanum chromite heater was used

Table 1

Compositions of the starting material (wt.%) used in the present study compared with those used in other high pressure experiments in carbonated peridotite systems.

Elements	1	2	3	4	5	Present study
SiO ₂	45.2	45.56	45.07	42.26	42.29	42.4
TiO ₂	0.7	1.14	0.29	0.15	0.20	0.3
Al ₂ O ₃	3.5	6.02	4.05	3.41	3.52	4.2
Cr ₂ O ₃	0.4	0.62	0.69	0.29	0.21	0.4
FeO*	8.0	7.44	6.44	7.70	8.07	7.6
MnO	0.1	0.12	0.11	0.11	0.13	–
MgO	37.5	29.49	33.92	39.65	39.26	35.6
NiO	0.2	0.12	0.11	–	–	–
CaO	3.1	6.41	6.09	3.27	3.52	3.6
Na ₂ O	0.6	0.82	0.60	0.29	0.29	0.5
K ₂ O	0.1	0.15	0.58	0.02	0.01	0.4
H ₂ O	–	–	0.22	–	–	–
CO ₂	–	2.29	1.83	2.61	2.51	5.0
Mg#	89.31	87.60	90.37	90.18	89.66	89.30

1. Pyrolite model composition (Ringwood, 1975).

2. Wallace and Green (1988).

3. Thibault et al. (1992).

4. Hirose (1997).

5. Dasgupta and Hirschmann (2006).

*Total Fe as FeO.

Mg# = 100 X molar Mg / (Mg + Fe).

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