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Phase transition and thermal equations of state of (Fe, Al)-bridgmanite and post-perovskite: Implication for the chemical heterogeneity at the lowermost mantle

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ABSTRACT

In this study, we have determined the phase boundary between $Mg_{0.735}Fe_{0.21}Al_{0.07}Si_{0.965}O_3$-Bm and PPv and the thermal equations of state of both phases up to 202 GPa and 2600 K using synchrotron X-ray diffraction in laser heated diamond anvil cells. Our experimental results have shown that the combined effect of Fe and Al produces a wide two-phase coexistence region with a thickness of 26 GPa (410 km) at 2200 K, and addition of Fe lowers the onset transition pressure to 98 GPa at 2000 K, consistent with previous experimental results. Furthermore, addition of Fe was noted to reduce the density (ρ) and bulk sound velocity (V_{Φ}) contrasts across the Bm-PPv phase transition, which is in contrast to the effect of Al. Using the obtained phase diagram and thermal equations of state of Bm and PPv, we have also examined the effect of composition variations on the ρ and V_{Φ} profiles of the lowermost mantle. Our modeling results have shown that the pyrolitic lowermost mantle should be highly heterogeneous in composition and temperature laterally to match the observed variations in the depth and seismic signatures of the D″ discontinuity. Normal mantle in a pyrolitic composition with ∼10% Fe and Al in Bm and PPv will lack clear seismic signature of the D″ discontinuity because the broad phase boundary could smooth the velocity contrast between Bm and PPv. On the other hand, Fe-enriched regions close to the cold slabs may show a seismic signature with a change in the velocity slope of the D″ discontinuity, consistent with recent seismic observations beneath the eastern Alaska. Only regions depleted in Fe and Al near the cold slabs would show a sharp change in velocity. Fe in such regions could be removed to the outer core by strong core-mantle interactions or partitions together with Al to the high-pressure phases in the subduction mid ocean ridge basalts. Our results thus have profound implication for the composition of the lowermost mantle.

1. Introduction

The bottom 100–300 km of the lowermost mantle, called D″ layer, is one of the most enigmatic regions in the planet (Cobden and Thomas, 2013; Lay, 2015; Wysession et al., 1998). It exhibits various anomalous seismic features, including strong seismic anisotropies, anti-correlation between the shear and bulk sound velocities, etc., and has been regarded to act as the thermal boundary and a chemical distinct layer above the core-mantle boundary (CMB) (e.g. Dziewonski et al., 1977; Garnero and Helmberger, 1996; Lay and Helmberger, 1983). In particular, a velocity discontinuity with strong lateral variations in depth was observed at the top of the D″ layer, referred to as the D″ discontinuity (Lay and Helmberger, 1983; Wysession et al., 1998). Many locations have a clear D″ discontinuity with a 0.5–3% increase in the compressional and shear-wave velocities, whereas a few locations only show a change in the velocity gradient at the top of the D″ layer (e.g. Hutko et al., 2008; Li et al., 1998; Sun et al., 2016a). Some locations even lack clear seismic signatures of the D″ discontinuity (Lay and Garnero, 2004; Sun et al., 2016a). In addition, recent seismic studies have identified a complicated lens-structure of the D″ discontinuity beneath Central Pacific regions and Central America (Hutko et al., 2008; Kawai et al., 2007a, 2007b; van der Hilst et al., 2007). Significant variations in the depth, seismic signature, and structure of the D″ discontinuity are expected to be caused by strong lateral chemical

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and/or temperature variations, yet lacking direct evidence and necessary constraints (e.g. Garnero and McNamara, 2008; Lay, 2015; Sun et al., 2016b; van der Hilst et al., 2007).

The discovery of the phase transition from lower-mantle $MgSiO_3$-bridgmanite (Bm) to post-perovskite (PPv) at relevant pressure–temperature conditions of the lowermost mantle has shed lights on deciphering the origin of the D″ discontinuity (Murakami et al., 2004; Oganov and Ono, 2004; Tsuchiya et al., 2004). The Bm-PPv phase transition in the $MgSiO_3$-system is sharp and can produce a sudden increase in the shear-wave velocity, consistent with seismic observations of the D″ discontinuity (Hirose, 2006; Murakami et al., 2007a; Ono and Oganov, 2005). The lateral variations in the depth of the D″ discontinuity have been interpreted by the occurrence of the Bm-PPv phase transition at different temperatures (Hernlund et al., 2005; Hirose et al., 2006). However, subsequent experimental and theoretical studies have shown that the phase transition between (Fe, Al)-bearing Bm and PPv is much more complex than that in the $MgSiO_3$-system (e.g. Catalli et al., 2009; Dorfman et al., 2013; Grocholski et al., 2012; Ono and Oganov, 2005; Tateno et al., 2007). Since the lower-mantle Bm in a pyrolitic composition contains 8–11% Fe and 7–11% Al, the presence of Fe and Al can broaden the Bm-PPv transition thickness up to ∼400–600 km, in contrast to the observed sharp D″ discontinuity with a thickness of 50–75 km (Akber-Knutson et al., 2005; Caracas and Cohen, 2005; Catalli et al., 2009; Dorfman et al., 2013; Dziewonski et al., 1977; Lay and Helmberger, 1983; Mao et al., 2004; Tateno et al., 2007; Tsuchiya et al., 2004). The onset transition pressure of (Fe, Al)-bearing Bm is 500–600 km above the CMB, which is at a shallower depth than the D″ discontinuity, although a few studies also argued that Fe can stabilize Bm instead of PPv (Akber-Knutson et al., 2005; Caracas and Cohen, 2005; Catalli et al., 2009; Dorfman et al., 2013; Ismailova et al., 2016; Mao et al., 2004; Murakami et al., 2005; Stackhouse et al., 2006).

The Bm-PPv phase transition has also been studied in various multi-component systems, including KLB-1 peridotite, mid-ocean ridge basalt (MORB), San Carlos olivine, and harzburgite (Grocholski et al., 2012; Murakami et al., 2005; Ohta et al., 2008; Ono and Oganov, 2005; Sinmyo et al., 2011). In MORB composition, Bm starts to transform to PPv at 108–112 GPa and 2500 K using the Au pressure scale of Tsuchiya (2003) (Grocholski et al., 2012; Ohta et al., 2008). Yet there is a large uncertainty in the transition thickness in the MORB composition, ranging from 5 GPa in Ohta et al. (2008) to 12 GPa in Grocholski et al. (2012) at 2500 K. The transition thickness in the MORB composition is much narrower than that in the single component system potentially due to the partition of Fe to the calcium–ferrite-type aluminous (CF) phase and Al to the high-pressure phase of SiO_2 and/or the CF phase (Grocholski et al., 2012; Hirose et al., 2015). In the peridotite composition, existing experimental studies provided conflicting results about the phase boundaries between Bm and PPv (Grocholski et al., 2012; Murakami et al., 2005; Ohta et al., 2008; Ono and Oganov, 2005). Although the Bm-PPv phase transition in the peridotite composition was reported to occur at 113 GPa and 2500 K, later studies observed this phase transition taking place at 124–169 GPa and 2500 K using the same Au pressure scale of Tsuchiya (2003) (Grocholski et al., 2012; Ohta et al., 2008; Ono and Oganov, 2005). The difference in the transition pressure is as large as 20–30 GPa at 2500 K in the peridotite composition (Grocholski et al., 2012; Murakami et al., 2005; Ohta et al., 2008; Ono and Oganov, 2005). The conflicting results in various studies were interpreted to be caused by the thermal (Soret) diffusion during laser heating and/or kinetic hindering for fully transformation of Bm to PPv by repeatedly heating the same sample spot (Hirose et al., 2015).

Besides the Bm-PPv phase boundary, thermoelastic parameters of Bm and PPv are also needed to model the density and velocity profiles of the lowermost mantle in order to understand the origin of the D″ discontinuity. However, most experiments for the equations of state (EoS) of Bm and PPv were performed at high pressures but 300 K (e.g. Ballaran et al., 2012; Dorfman et al., 2013; Lundin et al., 2008; Mao et al., 2014, 2015, 2017; Nishio-Hamane and Yagi, 2009; Shieh et al., 2006). The thermal EoS was studied up to 108 GPa and ∼2900 K for $MgSiO_3$-Bm and up to 170 GPa and 2700 K for $MgSiO_3$-PPv (Fiquet et al., 2000; Sakai et al., 2016; Tange et al., 2012). Wolf et al. (2015) has determined the thermal EoS of $(Mg_{0.87}, Fe_{0.13})SiO_3$-Bm up to 133 GPa and ∼2400 K. Yet the combined effect of Fe and Al on the thermal EoS of both Bm and PPv is unknown. The density and velocity profiles of the lowermost mantle with (Fe, Al)-Bm and PPv as the dominant phases are thus poorly constrained.

Here we have determined the phase boundary between $Mg_{0.735}Fe_{0.21}Al_{0.07}Si_{0.965}O_3$-Bm and PPv as well as the thermal elastic parameters of both phases using synchrotron X-ray diffraction in laser-heated diamond anvil cells (DACs). Our experimental results yield crucial constraints on the combined effect of Fe and Al on the phase boundary between Bm and PPv and the thermal elastic parameters of both phases. The experimental results obtained here together with literature data have been used to model the density and velocity profiles of the lowermost mantle, which are important to decipher the origin of the D″ discontinuity.

2. Experiment

Natural enstatite, purchased from Alfa Aesar Corp., was used as the starting material. It was examined by X-ray diffraction (XRD) at ambient conditions to confirm the crystal structure and has a composition of $Mg_{0.735}Fe_{0.21}Al_{0.07}Si_{0.965}O_3$ determined by the electron microprobe at the Material Center, University of Science and Technology of China. The starting enstatite was ground into the fine powder and mixed with 5 wt.% Au which was used as the pressure standard and laser absorber (Fei et al., 2007). Re was used as the gasket material. NaCl, used as the pressure medium and thermal insulator, was pre-dried for 5 h to avoid the potential contamination of water in the air. The sample mixture was sandwiched between two NaCl foils with thickness of ∼5 μm. DACs equipped with a pair of diamonds with 200 μm in culet were used for experiments under 90 GPa to study the phase stability and thermal EoS of Bm. Measurements up to 140 GPa were performed using DACs with 150/300 μm beveled diamonds to investigate the thermal EoS of Bm and the Bm-PPv phase transition. Experiments between 140–200 GPa were conducted using DACs with 75/300 μm beveled diamonds.

The high pressure and temperature XRD experiments up to 202 GPa and 2600 K were performed at the GeoSoilEnviroConsortium (GSECARS) of the Advanced Photon Source (APS), Argonne National Laboratory (ANL). The sample was heated from both sides using a double-sided Nd:YLF laser heating system (Prakapenka et al., 2008). For experiments below 140 GPa, we always heated the sample at a fresh spot at an interval of 100–300 K in each heating cycle. The temperature was determined by fitting the collected thermal radiation spectrum using the Planck radiation function under the Graybody approximation (Prakapenka et al., 2008). A temperature quenched XRD pattern after each heating cycle was also collected to determine the unit cell parameters of Bm or PPv at 300 K and high pressures. We have also quenched the DACs with 200 μm culet diamonds to ambient conditions to constrain the unit cell volume of Bm.

3. Result

The phase stability of Bm and transition between Bm and PPv have been well constrained by our high pressure–temperature re-