Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

SMP044-P05 Room:Convention Hall Time:May 25 14:00-16:30

High-pressure stability field and thermodynamic properties of anhydrous phase B (Mg14Si5O24)

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Anhydrous phase B (Anh-B:$Mg_{14}Si_5O_{24}$) is a high-pressure magnesium silicate possibly stable in magnesium-rich regions in the upper mantle (Ganguly and Frost, 2006). They examined the equilibrium boundary of the reaction, 5forsterite (Fo) + 4periclase(Per) = Anh-B, at 9.0-12.5 GPa and 1173-1873 K. Ottonello et al. (2010) computed thermo-chemical and -physical properties of Anh-B by first principles calculation. However, those of Anh-B have been poorly determined by experiments. In this study, we examined stability field of Anh-B by high pressure experiments, and performed drop-solution calorimetry, heat capacity measurement and high temperature X-ray diffraction measurement on Anh-B. Obtained data were applied to thermodynamic calculation of the phase equilibrium boundaries of the reactions Fo+Per =Anh-B and Anh-B=Wads+Per which were separately determined by high pressure experiments.

High-pressure high-temperature phase relations experiments on Anh-B were performed using a Kawai-type 6-8 multianvil high pressure apparatus. The starting material was a mixture of Per and Fo with 4:5 in molar ratio. The experimental conditions were in a pressure range of 12-23GPa and in a temperature range of 1673 -2073 K. Samples recovered after quenching were analyzed with powder X-ray diffraction method and SEM-EDS. Drop solution calorimetry of Anh-B was performed using a Calvet-type calorimeter with Ar bubbling technique and lead borate solvent. Heat capacity was measured by DSC in a range of 300-770 K. High temperature X-ray diffraction measurements were made in a range of 303-773K every 50 K step. Raman spectroscopy of Anh-B was done using a micro-Raman spectrometer (JASCO NRS-3100).

The results of the high-pressure and high-temperature experiments indicated that the equilibrium boundaries of 5 Fo + 4 Per = Anh-B and of Anh-B = 5 Wadsleyite(Wads)+ 4 Per are located at 13 GPa and at 19 GPa, respectively, at 1873 K. The drop-solution enthalpy of Anh-B was determined to be 844.3 +/- 29.9 kJ/mol. The drop-solution enthalpy of Fo was 168.2 +/- 0.9 kJ/mol, and that of Per was 33.7 +/- 1.0 kJ/mol (H. Kojitani, unpublished data), and that of Wads was 142.2 +/- 2.7 kJ/mol (Akaogi et.al., 2007). The enthalpy changes for the reactions 5 Fo+ 4 Per = Anh-B and Anh-B = 5 Wads + 4 Per were obtained as 130.9 +/- 30.5 kJ/mol and -1.46 +/- 33.08 kJ/mol, respectivery. We obtained a heat capacity equation of $C_{P(Anh-B)}$=$1.531*10^3$-$1.315*10^4T^{-0.5}$-$7.925*10^8T^{-3}$ and thermal expansivity of $2.07*10^{-5}$+$1.75*10^{-8}$T. Raman spectrum of Anh-B were consistent with lattice vibrational mode frequencies calculated by Ottonello et al. (2010). By applying the C_P, thermal expansivity and Raman data to Kieffer model calculation, entropy of Anh-B was estimsted to be 563.37 J/(mol K). Therefore, entropy changes for the reactions 5 Fo+ 4 Per = Anh-B and Anh-B = 5 Wads + 4 Per were obtained as -14.43 kJ/(mol K), and -24.07 kJ/(mol K), respectively. The phase boundaries by thermodynamic calculations were consistent with those obtained by the high pressure experiments. It is concluded that stability field of Anh-B is placed at the range of 13-19 GPa and 1873 K.

Keywords: anhydrous phase B, high pressure high temperature experiment, thermodynamic calculation