

Low-temperature heat capacity measurement of MgSiO_3 majorite

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It is thought that garnet is a major Earth's mantle constituent mineral. High pressure high temperature experiments have cleared that pyroxene component dissolves into the garnet phase with increasing pressure. MgSiO_3 majorite (Mj) which does not contain Al is one of the important endmembers of such the silicate garnet containing pyroxene component. No entropy data of MgSiO_3 Mj based on measured heat capacity existed though it is necessary for thermodynamic treatment of MgSiO_3 Mj. In this study, we performed low-temperature heat capacity (C_p) measurement of synthetic MgSiO_3 Mj using the thermal relaxation method and determined the entropy from the lattice vibrational contribution at standard state. MgSiO_3 Mj sample for calorimetry was synthesized by holding MgSiO_3 glass at 19 GPa and 2173 K for one hour using a high-pressure apparatus at Ehime University, GRC. Since the recovered sample showed the coexistence of MgSiO_3 akimotoite with MgSiO_3 Mj at lower temperature region, MgSiO_3 akimotoite was removed. After the cleaning, it was confirmed that the MgSiO_3 Mj sample was a single phase using a micro-focused X-ray diffractometer and a micro-Raman spectrometer. Low-temperature C_p measurement was made using a Physical Property Measurement System (PPMS) apparatus (Quantum Design) in a temperature range of 2-306 K with a temperature step of 2 K. The weight of the sample was 14.415 mg. Low-temperature C_p data measured in this study are compared with previous ones measured above 150 K using DSC method by Yusa et al. (1993). Our data is consistent within the errors with those of the latter around 300 K. While, in a temperature range of 150-200 K, the present values are about 5% larger than those of the latter. Entropy from the lattice vibrational contribution at standard state ($S_{298.15, \text{vib}}$) was determined to be 65.35(2) J/mol.K by integrating C_p/T in an interval of 0-298.15 K. In addition, configurational entropy (S_{conf}) is calculated as 1.76 J/mol.K by assuming that the degree of Mg-Si disorder at the octahedral sites is 15%. Therefore, the standard entropy of MgSiO_3 Mj is obtained to be 67.11 J/mol.K from the summation of $S_{298.15, \text{vib}}$ and S_{conf} . The present result indicates that the standard entropy of MgSiO_3 Mj is larger than those previously estimated, e.g., 60.3 J/mol.K by Fabrichnaya et al. (2004).

Keywords: majorite, heat capacity, entropy, thermal relaxation method