Neutron diffraction study of delta AlOOD at high pressure and implications for pressure evolution of strong hydrogen bond

Asami Sano[1]; Carine Vanpeteghem[2]; Eiji Ohtani[3]; Akio Suzuki[4]; Kazuki Komatsu[5]

[1] ISSP, Univ. Tokyo; [2] Inst. Mineral., Petr. and Econ. Geol.; [3] Inst. Mineral, Petrol. & Econ. Geol.,Tohoku Univ; [4] Inst.Mineral. Petrol.& Econ. Geol., Faculty of Sci.,Tohoku Univ; [5] Tohoku Univ

Delta-AlOOH is a high-pressure polymorph of diaspore (alpha-AlOOH) and has ability for transporting hydrogen into lower mantle in the subducting slab. The structure is described as a distorted rutile structure; edge-sharing Al-O octahedrons make single-chain along c-axis. This hydrous phase is remarkable for strong hydrogen bond suggested both short O-O length (2.50-2.55 A; Suzuki et al., 2000, Kudoh et al., 2004, Komatsu et al., 2006). First principals calculations also showed hydrogen bond of delta AlOOH to be symmetric. However, the transition pressure of asymmetric to symmetric bond is vary; Tsuchiya et al., 2002 suggest that transition occurs at 28 GPa, Panero and Stixrude 2004 reported that hydrogen bond is symmetric at ambient pressure, and Li et al., 2006 argued that symmetric hydrogen bond is formed above 50 GPa. To investigate the hydrogen position and hydrogen bond behavior at the high pressure, we conducted the neutron diffraction experiments of delta-AlOOD.

Time-of-flight neutron diffraction study was performed at the PEARL high pressure facility of ISIS at Rutherford Appleton Laboratory. Delta-AlOOD was synthesized from deuterated bayerite at 18 GPa and 900 C using multi anvil high pressure apparatus. At ambient condition, delta-AlOOD was loaded into the vanadium can. High pressure diffraction data was corrected using an opposed-anvil Paris-Edinburgh cell. Sample was loaded into the Ti-Zr gasket and deuterated MeOD/EtOD was added as pressure medium. Pressure was calculated from refined unit cell volume using equation of state of delta-AlOOH. Data was corrected up to 9 GPa and any significant broadening was not observed. Rietveld refinement was obtained to refine the structure including hydrogen position.

In refinement of the structure at ambient condition, four models were attempted; two of asymmetric hydrogen bond models (Space group of $P2_1nm$ and $Pn2_1m$), symmetric hydrogen bond model (Pnnm) and hydrogen disordered model (Pnnm). R factor is significant low in $P2_1nm$ model compared to other three models. Moreover, there remained unexplained peak in three model except for $P2_1nm$ model. At high pressure, O1...D bond shortens as pressure increase and O1...O2 bond length decreases, whereas O2-D bond was lengthened. The discrepancy of the Al-O bond lengths that consists AlO6 octahedron become small as pressure increases. Linear extrapolation of O1...D and O2-D evolution as a function of pressure provides the intersection at 24 GPa.