

## Hydrogen bonding of goethite at high pressure -An application of TOF neutron diffraction experiments at high pressure-

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Goethite crystallizes in space group Pbnm. Oxygen atoms occupy two crystallographically independent oxygen sites, O(1) and O(2), in a slightly distorted HCP arrangement. The O(1) is surrounded by three iron atoms to form an almost planar triangle. The O(2) is hydrogenated, and surrounded by three iron atoms and a hydrogen atom, which form a distorted tetrahedral coordination. A non-linear hydrogen bonding connects O(2)-H...O(1) in an adjacent octahedron. Prior to neutron scattering experiments, we performed synchrotron x-ray powder diffraction experiments at high pressure on goethite ( $\alpha$ -FeOOH) (Nagai et al., 2003). The compression behaviour of goethite is anisotropic and the a-axis is almost two times more compressible than the b- and c-axes. Compression of goethite seems to be mainly controlled by shortening of the hydrogen bonded O...O distance. Since not only the O...O distance is important, but also the bond angle of O(2)-H...O(1) plays an additional role in hydrogen bonding, which becomes crucial during compression, it is interesting to determine the hydrogen position in goethite at high pressure and see what happens in hydrogen bonding with increasing pressure.

Actually, since hydrogen itself has large amplitude of incoherent scattering for neutrons, deuterated goethite powder was synthesized as a targeted material. About 100 mg of the deuterated goethite was loaded into the Paris-Edinburgh opposed-anvil cell. Ti/Zr gaskets, which are almost invisible by neutrons, were used and deuterated methanol/ethanol 4:1 mixture was put into the sample chamber as pressure transmitting media in order to generate hydrostatic pressure up to 10 GPa (Marshall and Francis, 2002). TOF neutron diffraction patterns were obtained on the POLARIS beamline in ISIS. The generated pressure was estimated from the compression data of goethite in Nagai et al.(2003).

We obtained TOF neutron diffraction patterns of goethite at 0.1 MPa, 2.3, 5.2 and 7.2 GPa. A typical exposure time was from 3 to 5 hours. No significant peak broadening occurred at all pressure conditions. Rietveld refinement was carried out and we obtained structural parameters at each pressure. On the basis of those refined parameters, selected bond distances were calculated. Hydrogen bonded O...O distance shortens with increasing pressure and these values are quite good agreement with the data by x-ray diffraction experiments (Nagai et al., 2003). In this study, we could refine a hydrogen position. With increasing pressure, O(2)-D is almost constant and O(1)...D hydrogen bonding shortens. However, it is interesting that O(2)-D...O(1) angle slightly decreases with increasing pressure.