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# Estimation of hydrogen position in super hydrous phase B, $Mg_{10}Si_3H_4O_{18}$, at 1.0 GPa using single-crystal diffraction data

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## Introduction

Super hydrous phase B (sup B), $Mg_{10}{}^{VI}Si^{IV}Si_2H_4O_{18}$, is known as one of the dense hydrous silicate minerals (DHMS). According to several previous researches (*ex.* [1], [2]), this phase is stable under high-pressure and high-temperature conditions such as -30 GPa and -1500C. DHMS phases are important in the view of the carriers and reservoirs of water in the Earth′s subduction zone. Sup B has hydrogen bonding in its structure. Hydrogen bonding should be influenced on its thermodynamic properties, especially under high-PT conditions. We try to investigate the hydrogen position under high-perssure condition by ME (Maximum entropy) analysis using single crystal diffraction data set.

High-pressure single-crystal X-ray diffraction measurements of sup B were conducted to collect intensity data for estimation of hydrogen position at 1GPa. In this paper, we reported the results of structural analysis and ME analysis of sup B at 1 GPa.

## Experimental Procedure

The sample used for this study was synthesized at 20 GPa and 1000°C kept for 4 hours using a Kawai type multi anvil apparatus installed in Gakushu-in University. A single crystal of OH end member sup B (0.08 × 0.08 × 0.04 $mm^3$ in size) was mounted on a modified Merrill-Bassett type diamond anvil cell with a small piece of a ruby crystal, which used for the pressure calibration. The 4:1 fluid mixture of methanol and ethanol was used for the pressure medium and a SUS301 stainless plate used for a gasket. Pressure was determined by the ruby fluorescence method. The wavelength of X-ray radiation was calibrated by the unit cell volume of the ruby standard crystal at room temperature.

The X-ray diffraction intensities were measured using an automated four-circle X-ray diffractometer installed at the beam line BL-10A, Photon Factory, High Energy Accelerator Research Organization. The wavelength of synchrotron radiation (λ=0.6493Å). The unit cell parameters of sup B at 1.0 GPa were $a$= 5.095 Å, $b$= 13.974 Å, $c$= 8.707 Å. The X-ray reflection intensity data at 1.0 GPa were collected up to $\sin\theta/\lambda <$ 0.88 $Å^{-1}$ (maximum $2\theta$ is 70.0°). Corrections for background and Lorentz-polarization were applied to all measured reflections. After these corrections, the absorption correction of diamond anvil cell was applied to all intensity data. Internal consistency of equivalent reflections was $R_{int}$=3.6%. The symmetrically equivalent reflections were averaged by the Laue symmetry of *mmm*. 550 reflections (Fo > 4.0σ(Fo)) were used for the structure refinement at 1.0 GPa. In this analysis, space group of sup B structure is *Pnnm*. Final agreement factors were $R$=2.8 %, $wR2$=7.4% and GoF=1.129. All calculations were performed using SHELXL97 [3]. For ME analyses, PRIMA [4] was used. The initial phase for ME analysis was calculated from the result of structure analysis. Unit cell was divided into 48 × 140 × 88 sections for ME analysis. Reflections, which have Fo<1.5σ(Fo), do not use for ME analysis.

## Results

The position and isothermal temperature factor of hydrogen could be refined at 1.0 GPa. H located at $x/a$=0.929(10), $y/b$=0.297(8), $z/c$=0.100(5), $U_{eq}$=0.08(2). O-H distance was 0.86(9) Å, which is in agreement with that of sup B at ambient conditions (0.91(3)Å in our result and 0.89(2)Å in [5]).

The electron distribution map of sup B after ME analysis was shown in Figure 1. In this analysis, although the electron distribution of each atom was distributed widely along the $b$-direction because of decrease of the number of observed reflection intensity data, the electron concentration corresponding to hydrogen was observed.

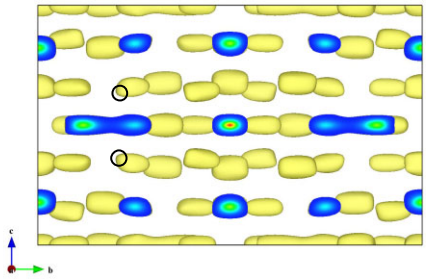

**Figure 1.** Electron distribution map of sup B at 1.0 GPa calculated by PRIMA using single-crystal diffraction data.

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## References

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