Compression of O-H- -O distances of Phase A, $Mg_7Si_2H_6O_{14}$

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Sets of X-ray diffraction intensities up to $\sin\theta/\lambda$ = 0.7~1.0 $Å^{-1}$ were measured with a 60×50×30 μm single crystal of phase A, $Mg_7Si_2H_6O_{14}$ synthesized by Kagi et al. (2000)[1] using a multi-anvil apparatus at conditions of 1000℃ and 10 GPa. X-ray diffraction data at ambient pressure were measured with MoKα radiation (50 kV, 40 mA). X-ray diffraction data at high pressures were measured using synchrotron radiation of wave lengths 0.6965 Å, 0.6993 Å and 0.7026 Å at 4.5 GPa, 9.1 GPa and 9.4 GPa, respectively at the beam line BL-10A, Photon Factory, Tukuba, Japan. Result of electron microprobe analysis yields a chemical composition of $Mg_{6.99}Si_{1.99}H_{6.06}O_{14}$. The difference of the total weight was ascribed to H_2O. The modified Merrill-Bassett type diamond anvil pressure cell [2] was used. A 0.25 mm thick stainless steel (SUS301) plate was used as gasket material. As a fluid pressure medium, a 4:1 mixture of methanol and ethanol was used. The pressure was calibrated using the ruby fluorescence method. The unit cell parameters obtained using 15~24 reflections with 2θ from 10° to 50° are given in Table 1. Out of 2300 reflections in a sphere which were accessible with diamond anvil high pressure cell, a total of 164~299 symmetry independent reflections (Io≥1.5σIo ~ Io≥3σIo) were obtained by averaging the symmetry equivalent intensities in Laue group 6/m. Intensities were corrected for Lorentz factor and no absorption correction was applied because of the sufficiently small value of μr (<0.04). From the study of neutron powder diffraction data of phase A at ambient pressure and 3.2 GPa, Kagi et al. (2000)[1] reported that increased pressure further increases the hydrogen bonding of D1 compared to D2 and O4-D2---O3 showed no significant increase or decrease in hydrogen bonding in the pressure range investigared. Our results in the present study are in general agreement with the observation of Kagi et al. (2000)[1]. The O-O distances around hydrogen atoms in Table 2 indicate that both O2-O3 and O4-O3 distances significantly decrease and O2-O3 distance decrease more than O4-O3 distance with increasing pressure. O2-O3 distance decrease around 10% at 9.4 GPa, while O4-O3 distance decrease around 6% at the same pressure. It should be noted that the trend of O4-O3 is on a line of the O-O decrease in brucite up to 9.3 GPa reported by Parise et al. (1994) [3].

Table 1. Unit cell parameters, number of Fo and R values

P(GPa)	0	4.5	9.1	9.4
a (Å)	7.8604(7)	7.756(1)	7.6445(3)	7.644(1)
c (Å)	9.5702(8)	9.469(3)	9.383(7)	9.371(2)
$2\theta_{max}$ (°)	60.0	90.0	60.0	80.0
Radiation	MoKα	Synchrotron		
#of Fo	363	299	164	255
	Io≥σ3Io	Io≥σ3Io	Io≥σ1.5Io	Io≥σ3Io
R(%)	5.0	9.6	7.4	7.9
Rw(%)	6.0	9.1	8.4	7.3

Table 2. Atomic parameters of O2, O3 and O4 (parameters of all other atoms are not listed)

P(GPa)	0.0	4.5	9.1	9.4
O2 x	0.4759(8)	0.4728(15)	0.4748(13)	0.4700(12)
y	0.0983(8)	0.0985(15)	0.1046(13)	0.0985(12)
z	0.4853(8)	0.4848(20)	0.4836(22)	0.4815(20)
B	0.17(8)	0.4(1)	0.0(2)	0.0(1)
O3 x	0.4510(7)	0.4500(11)	0.4475(10)	0.4486(10)
y	0.2927(7)	0.2891(11)	0.2827(10)	0.2873(11)
z	0.2334(8)	0.2421(29)	0.2472(24)	0.2517(30)
B	0.13(9)	0.0(1)	0.0(2)	0.5(1)
O4 x	0.1660(7)	0.1634(10)	0.1539(11)	0.1604(9)
y	0.4344(7)	0.4344(11)	0.4310(11)	0.4370(10)
z	0.2406(8)	0.2443(27)	0.2628(29)	0.2549(25)
B	0.24(9)	0.0(1)	0.0(2)	0.0(1)

Table 3. O-O distances (Å) around H

P(GPa)	0	4.5	9.1	9.4
O2-H--O3	2.91(1)	2.79(3)	2.66(3)	2.64(2)
O4-H--O3	3.168(9)	3.06(2)	2.92(1)	2.99(1)

References

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