Stress-Induced Proton Disorder in Hydrous Ringwoodite

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The system $(Fe,Mg)_2SiO_4$ with its modifications olivine, wadsleyite and ringwoodite has been the subject of numerous studies in the past to determine the maximum solubility of hydrogen in their structures (e.g., Kudoh et al., 2000). Nowadays experimental work is focused on the effect of the water content on the physical, structural and thermodynamic properties.

Camorro Perez et al. (2006) studied hydrous (1 and 1.7 wt % water) Mg-ringwoodite by in-situ high-pressure IR spectroscopy up to 30 GPa using Neon as pressure medium and reported a second-order phase transition occurring in hydrated ringwoodite at pressures of about 25 GPa. The main feature of this phase transition was the sudden disappearance of the prominent OH band in Mg-ringwoodite centred at 3150 cm^{-1}. To study this phenomenon in more detail we synthesized more Fe-rich ringwoodite of different compositions ranging from x_{Fe} = 1.0 to x_{Fe} = 0.40. The samples were investigated by electron microprobe, secondary ion mass spectrometry (SIMS), conventional and synchrotron FTIR spectroscopy at ambient condition as well as in-situ up to 30 GPa in a diamond anvil cell (DAC).

Water determination using SIMS in combination with IR spectroscopy yielded concentrations ranging from 0.6 wt % for the Fe- endmember to 2.5 wt % for the Mg-richest sample. Several DAC experiments were performed with ringwoodite of compositions x_{Fe} = 0.4, 0.5 and 0.6 in a Megabar diamond anvil cell. We performed the high pressure measurements in three different pressure transmitting environments: (1) CsI powder, (2) cryogenically loaded liquid Argon, (3) cryogenically loaded liquid Argon annealed at 8.6 GPa to 120 °C (by placing the DAC in an oven for one hour) before further increasing the pressure to ensure more hydrostatic conditions (Wittlinger et al. 1997). In the OH – stretching region all samples loaded with method (1) and (2) show a sudden disappearance of the prominent OH feature at pressures between 10 and 12 GPa, independent on composition. This feature was accompanied by discontinuities in the lattice vibration region in the same pressure range. However, applying method (3) the OH stretching vibrations as well as lattice vibrations could be observed up to 30 GPa without any discontinuity and their pressure behaviour ($d\nu/dP$) can well be described by linear fits. Molecule vibrations are very sensitive to non-hydrostatic conditions and we interpret the disappearance of the OH-bands using method (1) and (2) as a stress-induced proton disordering in hydrous ringwoodite Salts as pressure media such as CsI or KBr are easy to load and transparent in the infrared but they are known for producing strongly non-hydrostatic conditions. The same is true for Argon above 10 GPa without thermal annealing.

References

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