

## Stability of Forsterite in the Presence of H<sub>2</sub> Fluid under High Pressure and Temperature

The influence of H<sub>2</sub> fluid on the phase stability of forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) was investigated under high pressure and high temperature using diamond anvil cells. In-situ X-ray diffraction measurements at BL-18C and Raman spectroscopy indicated decomposition of forsterite to periclase (MgO) and stishovite/quartz (SiO<sub>2</sub>) in the presence of H<sub>2</sub> fluid, even though the pressure and temperature conditions were consistent with the stability region of forsterite under the dry condition. The microtexture of the recovered samples suggested that periclase crystallized directly from forsterite under high temperature. On the other hand, SiO<sub>2</sub> composition is considered to dissolve into H<sub>2</sub> under high temperature and then crystallize with decreasing temperature.

Fluids such as H<sub>2</sub>O drastically change the stability phase of coexisting mantle minerals [1], and thus play an important role in the chemical evolution and dynamics of the Earth's mantle. The composition of the fluid phase is significantly influenced by the surrounding oxidation states. In the reduced mantle, fluids contain a certain amount of H<sub>2</sub> in addition to H<sub>2</sub>O [2]. However, the effects of the H<sub>2</sub> fluid on the stability and chemical properties of the coexisting minerals have not been understood. In this study, we performed experiments at high pressure and temperature to clarify the influence of H<sub>2</sub> fluid on the stability of olivine, which is the most abundant mineral in the upper mantle.

The high-pressure and -temperature experiments on the H<sub>2</sub>-Mg<sub>2</sub>SiO<sub>4</sub> system were conducted using diamond anvil cells (DAC) in the range between 2.5 GPa, 1400 K and 15.0 GPa, 1500 K. Powdered forsterite (iron-free composition of olivine) was first loaded into the sample chamber with a ruby ball for pressure measurements [3]. Then, hydrogen gas (99.99999% purity) was loaded into the sample chamber using high-pressure gas-loading apparatus. At first, the sample was compressed to the target pressure at room temperature, and then heated using a CO<sub>2</sub> laser for around 20 min. The temperature was measured by the spectroradiometric method. The samples were quenched to room temperature by turning off the laser beam. XRD measurements were performed under high pressure and room temperature with a monochromatic X-ray beam ( $\lambda = 0.6166 \text{ \AA}$ ) at BL-18C. The X-ray beam was collimated to 25–100  $\mu\text{m}$  in diameter to obtain the XRD pattern of the sample in DAC.

The XRD patterns were detected using imaging plates (IP). Raman spectra were also measured under high pressure. The microtexture of the quenched samples under the ambient condition was investigated using a scanning electron microscope (SEM) and a transmission electron microscope (TEM).

The XRD measurements were performed after heating under high pressure to confirm the stability of forsterite in the presence of H<sub>2</sub> fluid. The distinct diffractions of periclase (MgO) and stishovite (SiO<sub>2</sub>) were observed at the laser heating spot in addition to that of residual forsterite after heating at 10.0 GPa, 1600 K (Fig. 1). The formation of periclase and quartz (polymorph of stishovite) was also observed after heating at 2.5 GPa, 1400 K. The results clearly show decomposition of the forsterite, although the present pressure and temperature range is the stability field of forsterite under the dry condition. From the Raman spectra, H-H stretching mode was observed at around 4200 cm<sup>-1</sup> both before and after heating, implying that the decomposition of forsterite was induced by H<sub>2</sub> fluid.

To investigate the decomposition mechanism of forsterite in the presence of H<sub>2</sub> fluid, the microtexture of the recovered samples was investigated after being heated from 15.0 GPa, 1500 K using TEM. The reacted area consisted of aggregates of fine periclase grains, while larger crystals of residual forsterite were observed surrounding the reacted area. Periclase aggregates maintained the shape of the original forsterite like a pseudomorph. Periclase was considered to be crystallized under high temperature from forsterite directly. On

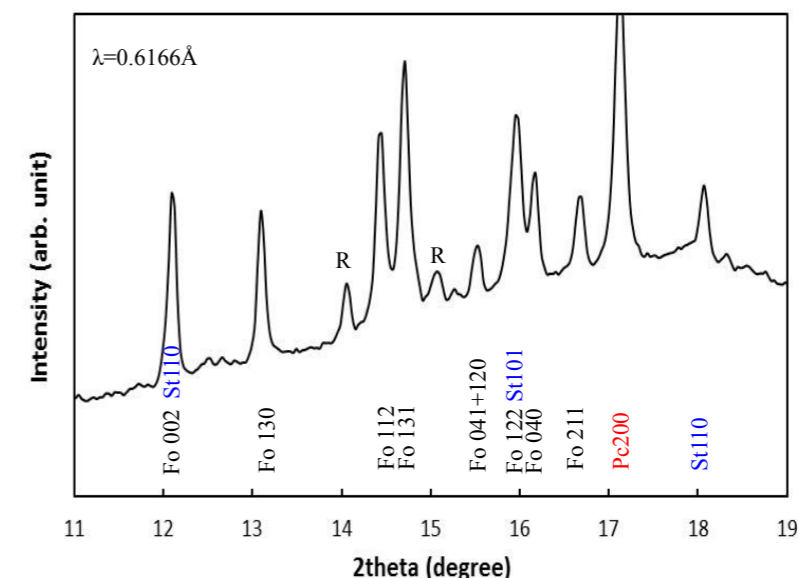


Figure 1: A representative XRD pattern of Mg<sub>2</sub>SiO<sub>4</sub>-H<sub>2</sub> system after heating at around 10 GPa, 1600 K. Fo; forsterite, Pc; periclase St; stishovite R; Ruby.

the other hand, euhedral crystals of stishovite were observed at the boundary between the reacted periclase and the residual forsterite. In addition, quench crystals of quartz were observed after heating at 2.5 GPa, 1400K. These results implied that the SiO<sub>2</sub> composition dissolved into H<sub>2</sub> fluid under high temperature and then quartz/stishovite was re-crystallized as the temperature decreased. See [4] for the details of the SEM and TEM observations. The present experimental results indicate that forsterite is incongruently dissolved in the presence of H<sub>2</sub> fluid to form periclase. Such dissolution of SiO<sub>2</sub> composition was not observed in the H<sub>2</sub>O-forsterite system [1]. The present study suggests that the stability of olivine in the mantle is strongly influenced by the composition of the surrounding fluid.

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### BEAMLINE

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