In Situ X Ray Observation of the Phase Transitions from $\alpha$ to $\gamma$ and from $\gamma$ to Perovskite+Periclase in Mg$_2$SiO$_4$

T. Kato, T. Kubo$^{1}$, H. Morishima$^{2}$, E. Ohtani$^{2}$, A. Suzuki$^{1}$, D. Yamazaki$^{5}$, K. Mibe$^{3}$, T. Kikegawa$^{3}$, and O. Shimomura$^{6}$

Geoscience Institute, University of Tsukuba, Tsukuba, Ibaragi 305, Japan.  
$^{1}$Faculty of Science, Tohoku University, Sendai, Miyagi 980, Japan.  
$^{2}$National Institute for Research in Inorganic Materials, Tsukuba, Ibaragi 305, Japan.  
$^{3}$Geological Institute, University of Tokyo, Tokyo 113, Japan.  
$^{4}$Earthquake Research Institute, University of Tokyo, Tokyo 113, Japan.  
$^{5}$Photon Factory, National Laboratory of High Energy Physics, Tsukuba, Ibaragi 305, Japan.  
$^{6}$SPring-8, Kanaji 1503-1, Kamigori, Ako, Hyogo 678-12, Japan.

In situ X-ray observation has been made on Mg$_2$SiO$_4$ forsterite in the pressure ranges over 25-30 GPa and 1250°C, using double stage multianvil system with sintered diamond anvil window and intense X-rays from synchrotron radiation. Forsterite ($\alpha$-phase) transformed into spinel ($\gamma$-phase) at 28 GPa and 800°C in the first heating cycles of the two runs. Spinel transformed into the assemblage of perovskite and periclase in the second heating cycles at 26.4 GPa and 1200°C and at 28.5 GPa and 950°C. The results show that $\alpha$-$\gamma$ transition can proceed outside the stability field of spinel, and that the blocking temperature, above which the stable assemblage of perovskite and periclase can be formed, has a pressure dependence.

[Earth material, phase relation, synchrotron X-ray, phase transition mechanism, kinetics]

1. Introduction

Understanding of phase transition behaviors of the mantle candidate minerals is important to clarify the structure and evolution of the silicate mantle of the terrestrial planets. Among them, Mg$_2$SiO$_4$ is the major end member of the Earth’s mantle, and its properties are believed to control the present mantle dynamics. We have developed a double stage multianvil system in order to apply in situ X-ray diffraction method to the above problem in the extended pressure temperature conditions [1-3]. Here we report preliminary observations of the transitions from $\alpha$ to $\gamma$ and from $\gamma$ to perovskite+periclase in Mg$_2$SiO$_4$ in the pressure range 24-28 GPa, which corresponds to the depth of the 670 km discontinuity of the Earth’s mantle.

2. Experimental method

In situ X-ray diffraction experiments were made at National Laboratory of High Energy Physics using MAX90 (BL-14C) and MAX80 (NE-5C) installed at Photon Factory. Double stage multianvil system with a sintered diamond anvil window for X-ray was used to generate the high pressure and high temperature conditions. Preformed pyrogyllite gaskets and (Mg,Co)O pressure medium were used for the pressure generation. TiC and diamond composite sheet heater and W3Re/W25Re thermocouple were used for the temperature generation and measurement. The sample container was boron formed by epoxy resin. Details of the experimental procedure is described in [3]. The starting material was the powdered mixture of $\alpha$-Mg$_2$SiO$_4$ and Au, and pressure values were calculated from the equation of state of Au [4, 8]. The error of pressure determination is about 0.5 GPa, due to uncertainties of EOS parameters and of volume and temperature measurements of the in situ experiments.

3. Results and discussions

Phase transition behaviors were investigated in two heating cycles of two runs. In the first heating cycle, gradual relaxation of the deviatomic stress component occurred and the diffraction peaks became sharp at temperatures above 400°C. The pressure and temperature conditions of the in situ measurements are shown in Fig. 1(a) and (b). Anticlockwise behavior of the P-T path is resulted from the pressure decrease associated with the volume reduction of the sample by the high pressure transitions.

The $\alpha$-$\gamma$ transition started at 800°C and 28.7 GPa in both runs, and the run pressures decreased gradually with the progress of the transition, which lasted about 30 min at 850°C and 28-25 GPa. The heating was terminated within 1 hours to prevent the pressure reduction. Fig. 2(a) shows a diffraction profile, which indicates coexistence of spinel and gold.
Fig. 1. Pressure and temperature conditions of the in situ measurements on Mg$_2$SiO$_4$. Open circles, open squares, and open triangles indicate $\alpha$-Mg$_2$SiO$_4$, $\gamma$-Mg$_2$SiO$_4$, and the assemblage of perovskite + periclase, respectively. (a) Run 1, and (b) Run 2.

Fig. 2. Diffraction profiles acquired at high pressures and temperatures. (a) Run 2 heating cycle 1 at 26.8 GPa and 850°C over the counting time 400 sec. Solid and open arrows indicate peaks corresponding to spinel and gold, respectively. (b) Run 2 heating cycle 2 at 27.1 GPa and 950°C over the counting time 600 sec. Solid and open arrows indicate peaks corresponding to perovskite or periclase, and gold, respectively.

In the second heating cycles after the further compression at room temperature, the transition from $\gamma$ to perovskite+periclase occurred within a few minutes at 1200°C and 26.4 GPa in the first run and 950°C and 28.5 GPa in the second run. The heating rate was about 10°C/min, and the temperature was kept constant.
when the transitions had started. The run pressures were found to be again decreased with these transition. After completion of the transitions, the run temperatures were elevated, and this assemblage was stable to higher temperatures.

The critical data points, where the phase transitions were observed to have started and progressed, are plotted in Fig.3, together with the acceptable range of the phase boundary between $\gamma$ and perovskite+periclase in Mg$_2$SiO$_4$ [5-6]. It is noted that $\gamma$ could grow from $\alpha$ in deep inside of the stability field of perovskite+periclase. The similar observation has been reported using quenching method, and its mechanism has been discussed on the basis of the TEM observation of the recovered run products of San Carlos olivine by [7]. Our data suggest that the blocking temperatures, above which the stable assemblage of perovskite+periclase could be formed, show pressure dependence. The accurate determination of the phase boundary, thus, requires further understanding of the kinetic aspects of the phase transition.

![Diagram](image)

Fig.3. Blocking temperatures of the transitions from $\alpha$ to $\gamma$ and from $\gamma$ to the assemblage of perovskite and periclase. Possible range of the stable phase boundary between $\gamma$ and pv+pc [5-6] is also shown.

References