Subsolidus Transition from Wadsleyite (beta phase) to Spinel (gamma phase) in the System Mg_2SiO_4 as a Function of Pressure and Temperature

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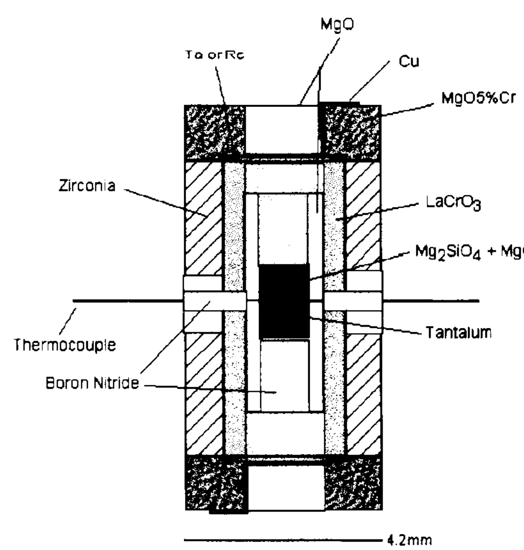
Abstract. Two experiments were made in order to locate the univariant boundary between alpha-phase and beta-phase in the system Mg_2SiO_4. The experiments indicate a phase boundary that is not entirely consistent with a previous in-situ determination or quench determinations. The dP/dT slope of the transition indicated by these results, ~ 0.0074 GPa/K, is steeper than nearly all previous determinations (less temperature dependent). However, ambiguity in the exact location of the transition remain for two reasons: 1) sluggish reaction kinetics, especially at temperatures less than 1400 K and when moving from alpha to beta phase, and 2) rapid grain growth of beta phase at temperatures greater than 1500 K. Further experiments are required to confirm and refine this important result.

Introduction. Olivine, $(0.9Mg, 0.1Fe)_2SiO_4$, is the most abundant mineral in earth's upper mantle. This mineral undergoes three important phase transitions which are critical for understanding upper mantle seismic structure. These are: 1) the Olivine to Wadsleyite transition (alpha to beta phase), 2) the Wadsleyite to Ringwoodite transition (beta to gamma phase), and 3) the dissociation of Ringwoodite to perovskite + magnesiowustite. The alpha-beta transition is widely recognized to be responsible for the seismic discontinuity at ~ 410 km, and the gamma dissociation reaction is thought to be responsible for the seismic discontinuity at ~ 670 km. Our group at Misasa, together with M. Matsui from Kyushu University, have been systematically determining these transitions in the system Mg_2SiO_4 *in situ* at SPring8. Initially, the of my experiment was to determine the beta-gamma transition. However, the initial pressure drop upon heating that occurred in our first experiment made the alpha-beta transition better target, so we decided to combine my beam time with that of M. Matsui in order to further investigate this critical transition.

Experimental. Fig. 1 shows a schematic of the pressure cell.

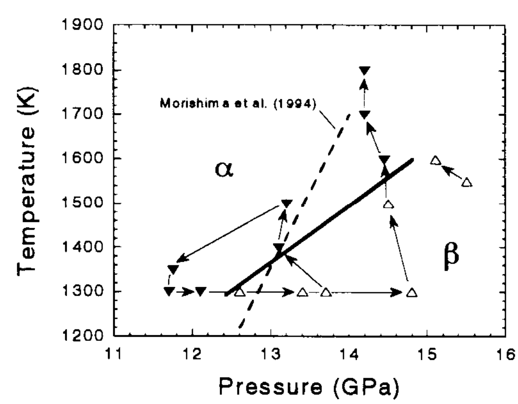
Fig. 1 10/4 Cell Assembly for SPring 8



The sample was contained within an MgO-5%Cr octahedron with 10 mm edge lengths, and the carbide anvils had 4 mm truncated edge lengths. This design can achieve pressures in excess of 20 GPa. Important characteristics of the design are: 1) the sample is a mixture of Mg_2SiO_4, $MgSiO_3$ (to help retard grain growth) and MgO (as pressure calibrant), 2) the sample is centrally located and the X-ray path is coaxial with the heater, 3) the sample is surrounded by highly insulating materials to reduce temperature gradients, 4) the thermocouple (TC) contact is made at the sample container for accurate sample temperature measurement, and 5) the heater is shortened and electrical leads are buried within the cell to avoid deformation of the heater and extrusion into the gasket. We made two successful experiments in which high temperatures were obtained (1000 - 1800 K) for periods of about 6 to 8 hours before TC failure. Both experiments failed at about 1800 K and the reason for this is not yet clear, but reaction of the TC with the tantalum sample container is likely. This design provided an excellent X-ray path with a window typically about 600 microns in diameter (we used a 50 micron collimated beam), and the signal from the sample and calibrant was strong. At each desired P-T condition, a 5 minute acquisition was made and pressure was calculated from the EOS of MgO by calculating unit cell volumes, typically using three MgO diffraction peaks.

Results. Fig. 2 shows the results of our experiments on a P-T diagram. Solid symbols depict alpha-phase stability, the open symbols are for beta phase, and the arrows depict the P-T paths of the experiments. The pressure was initially raised to about 20 GPa, well within the beta stability field, and temperature was increased. At about 1000 K, beta phase began to form, as evidenced by the growth of a strong {141} beta peak and reduction of the {211}, {131} and {120} alpha peaks. Further increase in temperature caused a decrease in pressure due to relaxation of pressure cell and gaskets. At 1400 K and 13.1 GPa, the beta {141} peak diminished and the alpha peaks clearly began to grow. Next, P and T were decreased, and then P was increased at a constant T of 1300 K. At 12.6 GPa, the first indication of the transformation to beta phase occurred as the strong alpha {211} peak began to diminish. Further increase in pressure produced a continued diminishing of alpha {211} and eventually the emergence of the beta {141} peak. However, no other strong beta peaks were observed and some alpha peaks remained, even after about 60 minutes at 1300 K and at higher pressures well within the beta stability field, attesting to the very sluggish growth of beta phase. Next, T was increased, and P decreased somewhat due to relaxation. At 1600 K and about 14.5 GPa the first indication of growth of alpha phase occurred. Again, the primary feature was the diminishing of the {141} beta peak and growth of the {211} alpha peak. In the second experiment, an attempt was made to cross the phase boundary at a higher P and T. However, rapid grain growth of beta phase at 15 GPa and 1600 K precluded good diffraction patterns at higher T, and the run failed at 1800 K.

Fig. 2



Conclusions. Fig. 2 shows that these new results are not totally consistent with the previous in situ determination made by Morishima et al. (1994). In particular, the dT/dP slope of the transition found in this study (~135 K/GPa) is much less temperature dependent than found by Morishima et al. (~330 K/GPa, Science, 265, 1202.). If our data are correct, there are two important geophysical implications: 1) the T assigned to the 410 km seismic discontinuity would be significantly different depending on which determination is used, and 2) the deflection of the seismic discontinuity to shallower depths caused by slab penetration would be more using our determination. However, we believe two improvements on the experiments are needed to confirm these results including: 1) Au as a pressure calibrant instead of MgO, and 2) longer collection times at critical P-T conditions to confirm growth of phases.