Supplementary information for

Evidence that the post-spinel transformation in Mg_2SiO_4 is responsible for the 660-km seismic discontinuity

Sang-Heon Shim, Thomas S. Duffy, and Guoyin Shen

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Due to thermal pressure and relaxation in the laser-heated diamond cell[1–3], the pressure measured using an internal standard changes as a function of temperature, heating duration, stress conditions, etc. during a single heating cycle. If described accurately, this P–T path provides additional information since it provides several different pressure-temperature points for one run. Pressure-temperature paths are shown for two runs near the phase boundary in figure 2 of the accompanying manuscript. Here we provide a detailed description of the paths for two other runs which were carried out near the post-spinel phase boundary.

Run 4 was performed during compression with ringwoodite as the starting material (Fig. 1s). The pressure before heating was 23.1 GPa. The pressure-temperature conditions for the first data point (pt. a in Fig. 1s) are anomalous relative to the others measured during this run. Since this datum also has large pressure and temperature uncertainties relative to the other points, it was not included in our analysis. After 5 minutes of heating the $MgSiO_3$ perovskite + MgO signature appeared in the X-ray diffraction patterns (pt. b), indicating the sample is above the post-spinel boundary. After 20 minutes of heating, we observe no ringwoodite and only the $MgSiO_3$ perovskite + MgO assemblage (pt. c). The disappearance of ringwoodite at later times in the run (despite generally lower pressures later in the run) may indicate that the progressive transformation of ringwoodite to perovskite + periclase is largely complete. However, this cannot explain the appearance of ringwoodite at the last heating point (pt. d) at apparently higher pressures. One possibility is that ringwoodite nucleates at the preceding point and the diffraction line of ringwoodite is observed due to crystal growth by P–T fluctuations between the two points. As we discuss in more detail below and in the text, other important effects (P–T uncertainty, kinetics, and preferred orientation) must be considered when

evaluating phase stability from the P-T path.

Run 9 was performed during decompression (Fig. 2s). The starting material was $MgSiO_3$ perovskite + MgO which were synthesized above 25 GPa and 1500–2000 K. After 6 minutes of heating, the diffraction peaks of ringwoodite appeared and coexisted with the high-pressure assemblage. This coexistence lasted for $\sim$24 minutes as the pressure and temperature conditions varied somewhat randomly. Finally, complete transformation to ringwoodite was observed during an abrupt decrease in pressure from 24.7 GPa to 21.7 GPa. The high-pressure assemblage was observed again upon increase of pressure in the next diffraction pattern. It is notable that large pressure excursions, and especially pressure decreases, were more commonly observed during decompression runs than during compression runs (see Fig. 2 of the paper and Fig. 1s). This may be related to the more effective relaxation of the diamond cell during decompression by heating.

In determining the post-spinel phase boundary, we have not explicitly considered the P–T path and instead assumed the data points near the phase boundary are randomly scattered. There are several reasons for this. First, as discussed in the text, the temperature uncertainties propagate into a pressure uncertainty of 0.3–0.9 GPa. This can explain much of the observed scatter ($\pm$ 1.1 GPa). The kinetics of the phase transformation is such that complete transformation from one assemblage to another is not readily observed in most cases. It is also important to recognize that our diffraction patterns reflect significant crystallographic preferred orientation during the heating process. In order to obtain a homogeneous temperature in the X-ray probed area, a small X-ray beam is crucial. However, this decreases the randomness of the X-ray probed sample powder. Moreover, recrystallization during the phase transformation and as a result of heating further decreases the randomness. Although this problem can be minimized by oscillating the diamond cell about the loading axis during heating, the intensity distribution of our diffraction patterns clearly indicates the existence of strong preferred orientation. Hence, we have adopted a conservative strategy and do not explicitly consider the pressure-temperature path in determining the phase boundary.

References

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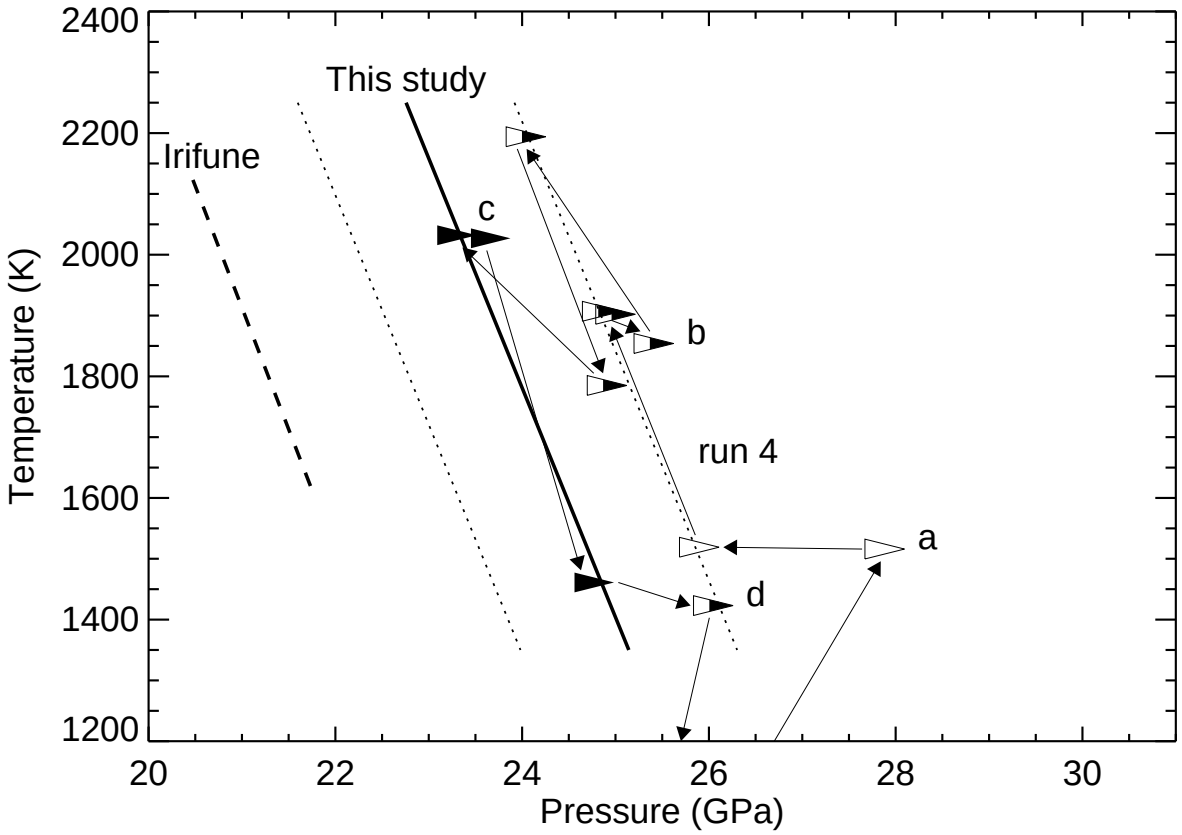

Figure 1s. P–T path for the run 4. This run was performed during compression. The notations for symbols follow those of figure 2 in the manuscript.

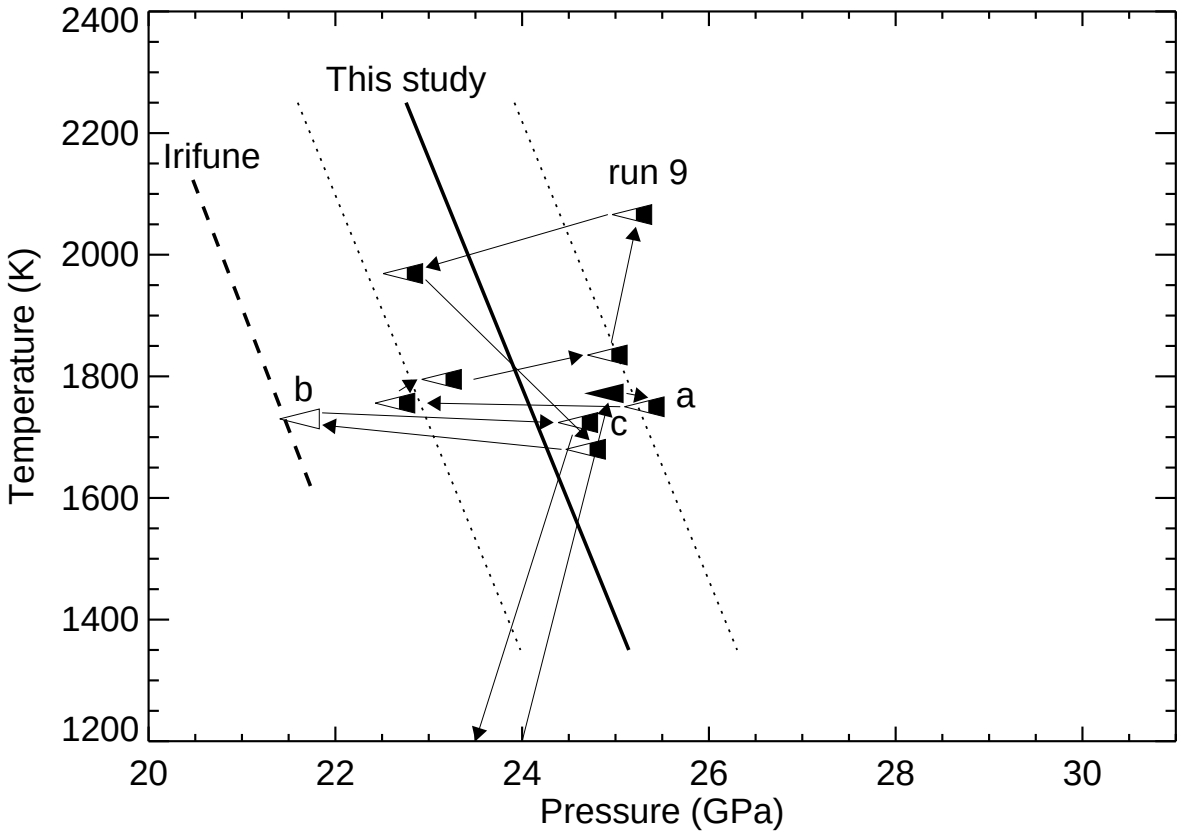

Figure 2s. P–T path for the run 9. This run was performed during decompression. The notations for symbols follow those of figure 2 in the manuscript.