

Supplementary Information for

**Water exchange process and bulk composition regulate slab dynamics and
deep earthquakes**

Jintao Zhu^{1,2,3}, Renbiao Tao^{2*}, Lifei Zhang¹, Takayuki Ishii^{3*}

¹ *SKLab-DeepMinE, MOEKLab-OBCE, School of Earth and Space Sciences, Peking
University, Beijing 100871, China*

² *Center for High Pressure Science and Technology Advanced Research (HPSTAR),
Beijing 100193, China*

³ *Institute for Planetary Materials, Okayama University, Misasa 6820193, Tottori,
Japan*

*Corresponding authors:

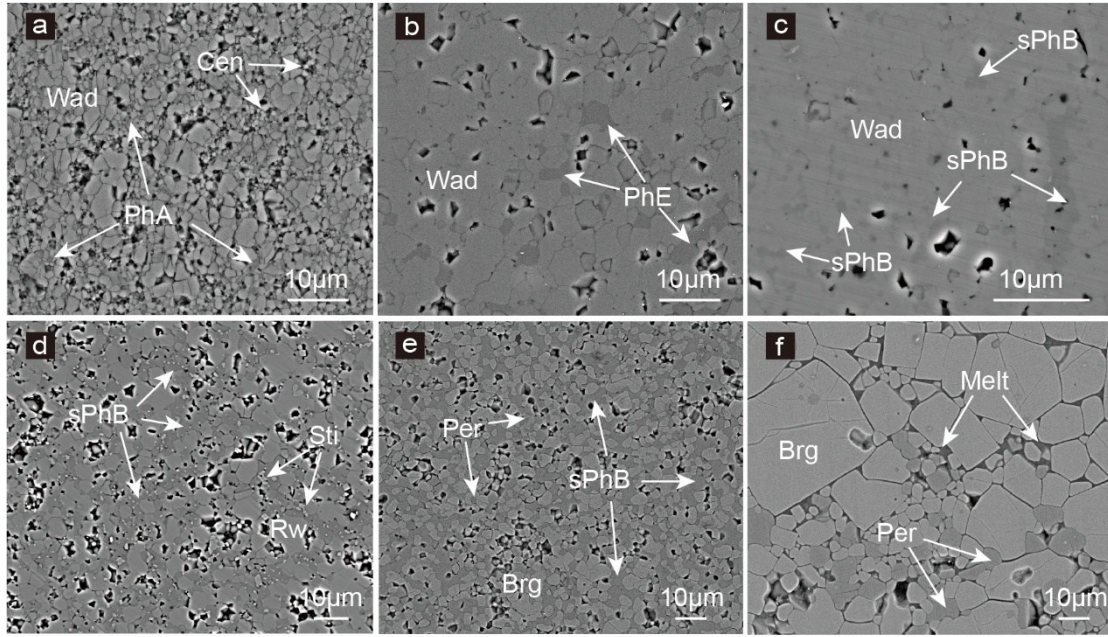
renbiao.tao@hpstar.ac.cn

takayuki.ishii@okayama-u.ac.jp

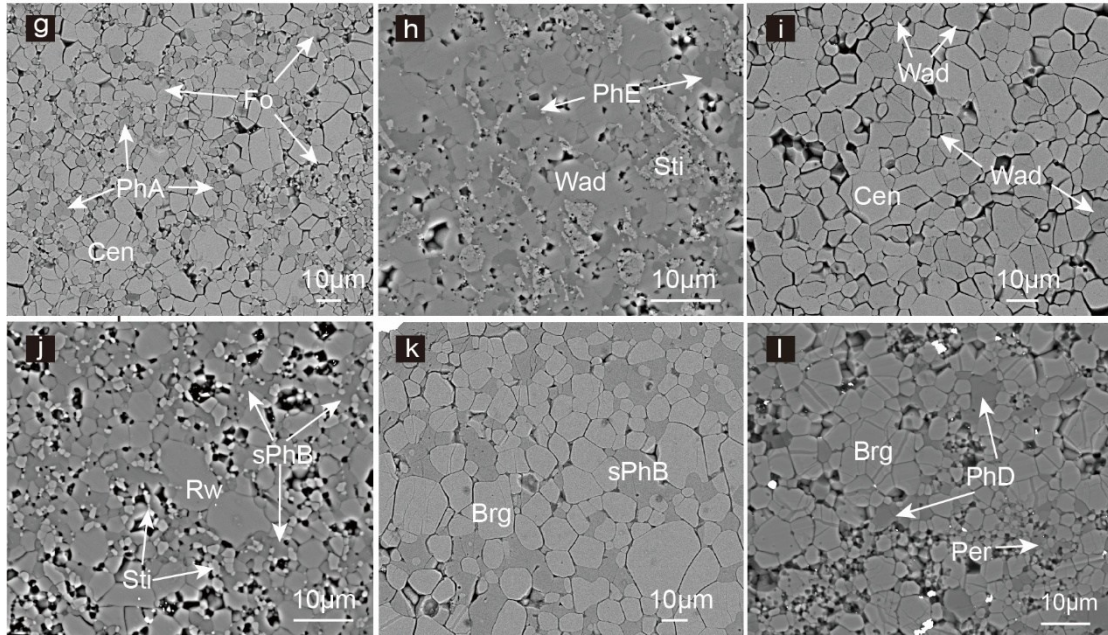
Introduction

This supporting information provides the Supplementary Figure 1–10, Supplementary Notes 1–3, and Supplementary Dataset 1–6 (In Excel format).

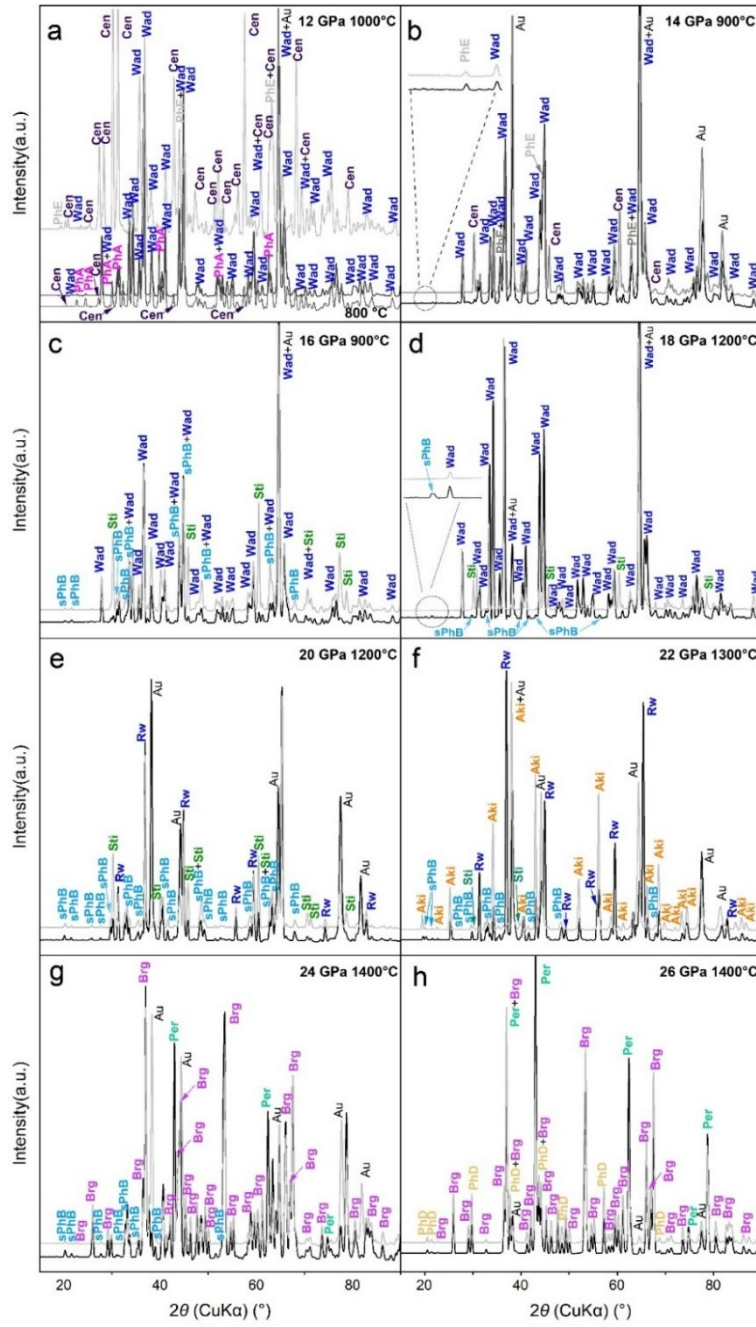
$\text{Mg}_2\text{SiO}_4 + 2\text{wt}\% \text{H}_2\text{O}$, $\text{Mg}/\text{Si}=2$



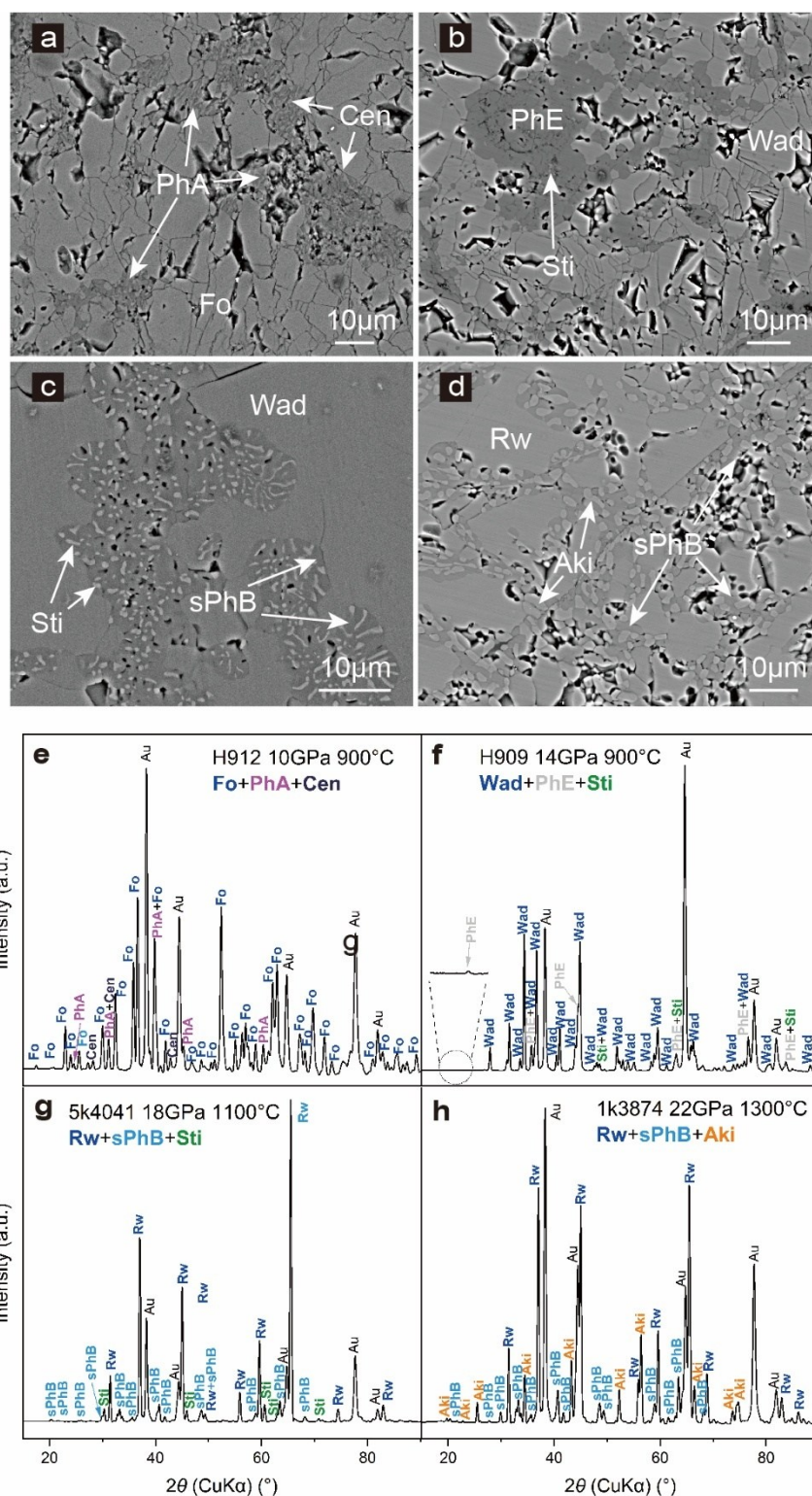
$\text{Mg}_{1.3}\text{SiO}_{3.3} + 2\text{wt}\% \text{H}_2\text{O}$, $\text{Mg}/\text{Si}=1.3$



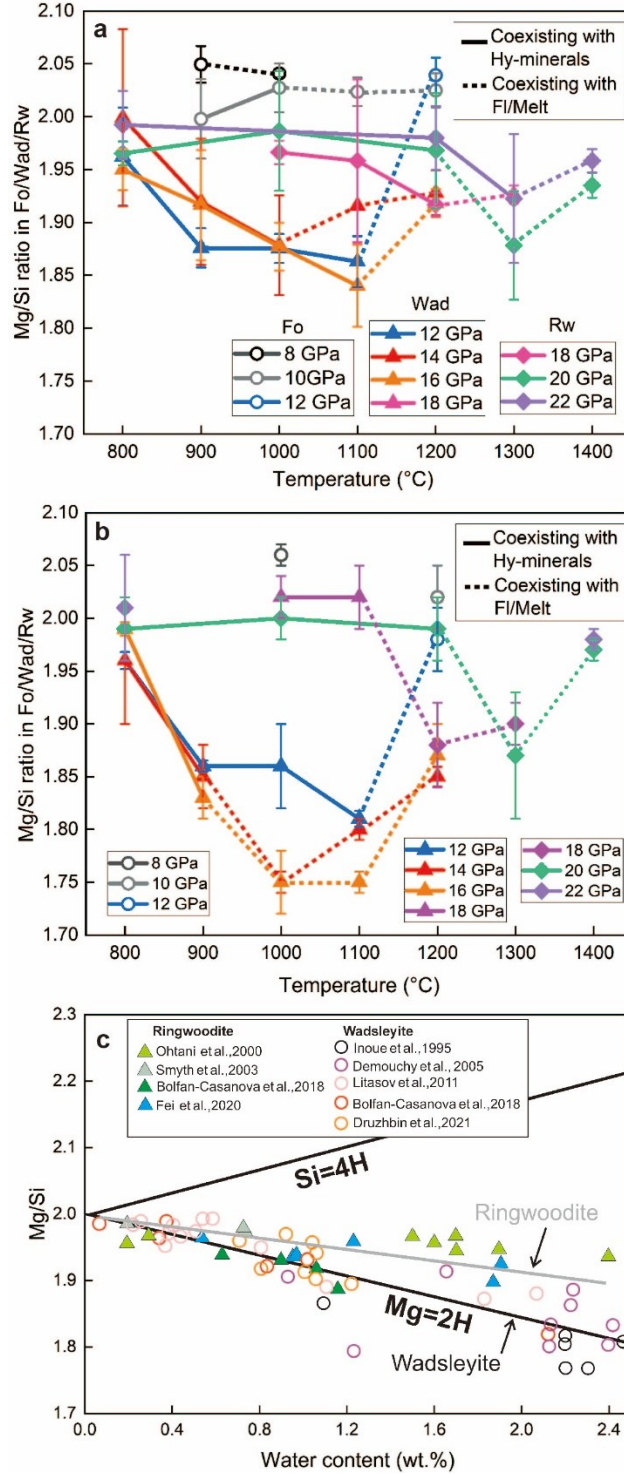
Supplementary Figure 1. Representative back-scattered electronic (BSE) images of recovered samples in $\text{Mg}_2\text{SiO}_4 + 2 \text{ wt}\% \text{H}_2\text{O}$ system (a–f) and $\text{Mg}_{1.3}\text{SiO}_{3.3} + 2 \text{ wt}\% \text{H}_2\text{O}$ system (g–l). (a) H857, 12 GPa and 800 °C; (b) H883, 12 GPa and 900 °C; (c) H849, 16 GPa and 1100 °C; (d) 5k3928, 20 GPa and 1200 °C; (e) 1k3851, 24 GPa and 1400 °C; (f) 5k4056, 26 GPa and 1300 °C; (g) H856, 10 GPa and 1000 °C; (h) H890, 14 GPa and 900 °C; (i) H849, 16 GPa and 1100 °C; (j) 5k3928, 20 GPa and 1200 °C; (k) 1k3851, 24 GPa and 1400 °C; (l) 5k4237, 26 GPa and 1100 °C.



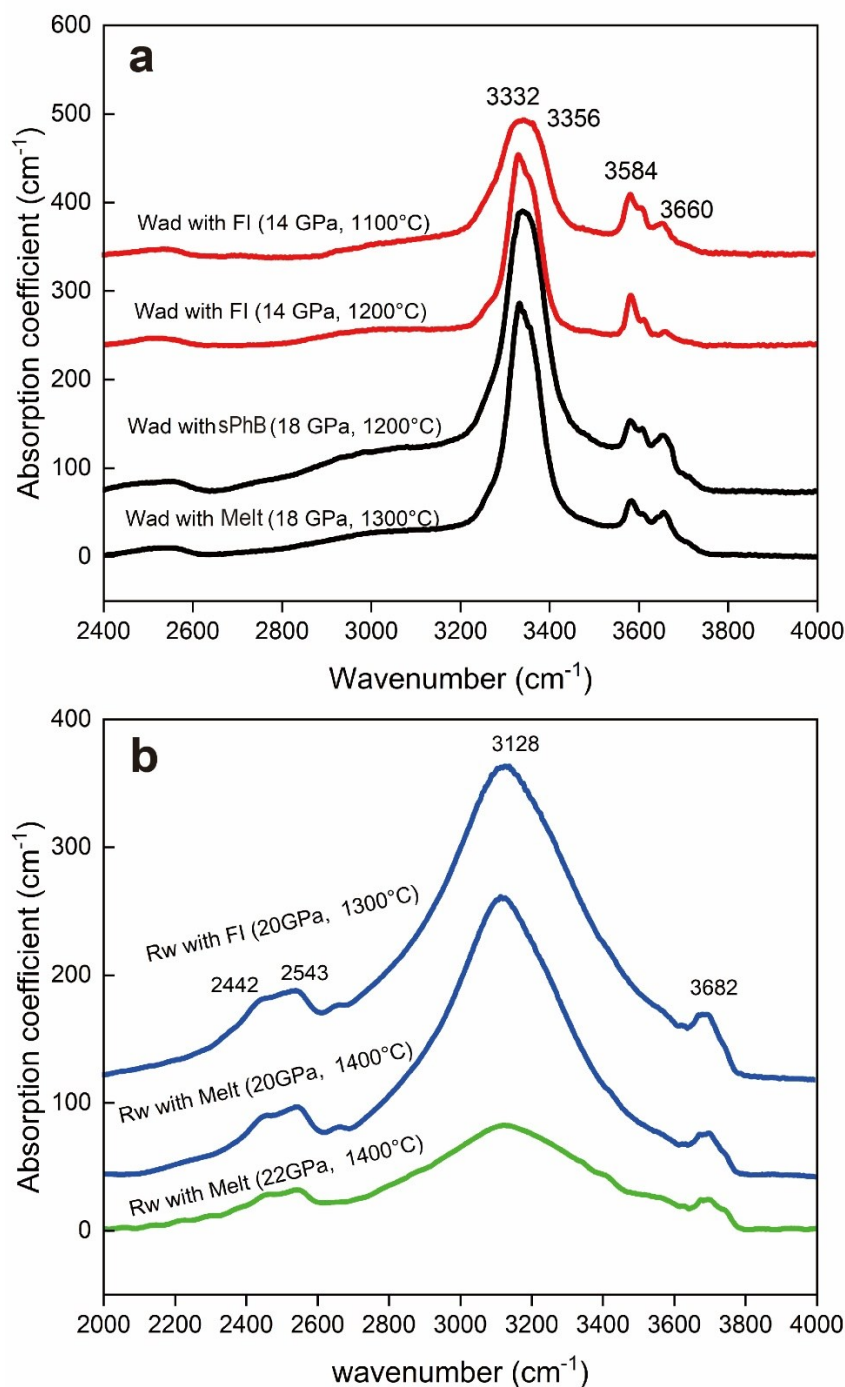
Supplementary Figure 2. Representative micro-focus X-ray diffraction patterns of the recovered samples of experiments in the systems $\text{Mg}_2\text{SiO}_4 + 2 \text{ wt\% H}_2\text{O}$ system (black lines) and $\text{Mg}_{1.3}\text{SiO}_{3.3} + 2 \text{ wt\% H}_2\text{O}$ system (Gray lines) (a) H857, 12 GPa and 800 °C and H854, 12 GPa and 1000 °C; (b) H890, 14 GPa and 900 °C; (c) H898, 16 GPa and 900 °C; (d) 5k3929, 18 GPa and 1200 °C; (e) 5k3928, 20 GPa and 1200 °C; (f) 1k3767, 22 GPa and 1300 °C; (g) 5k4122, 24 GPa and 1400 °C; (h) 5k4255, 26 GPa and 1200 °C.



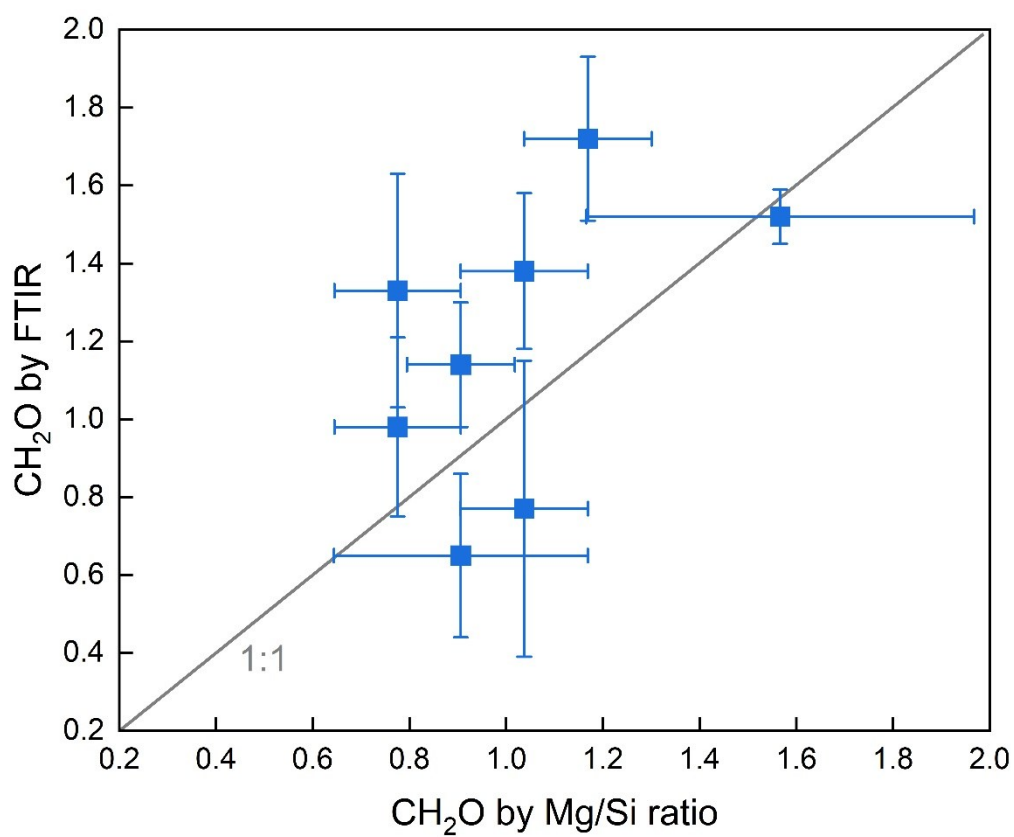
Supplementary Figure 3. Representative BSE images and micro-focus X-ray diffraction patterns of recovered samples of high-pressure experiments using pre-synthesized highly hydrated wadsleyite (a–b, e–f) and ringwoodite (c–d, g–h). (a), (e) H912, 10 GPa and 900 °C; (b), (f) H909, 14 GPa and 900 °C; (c), (g) 5k4041, 18 GPa and 1100 °C; (d), (h) 1k3874, 22 GPa and 1300 °C.



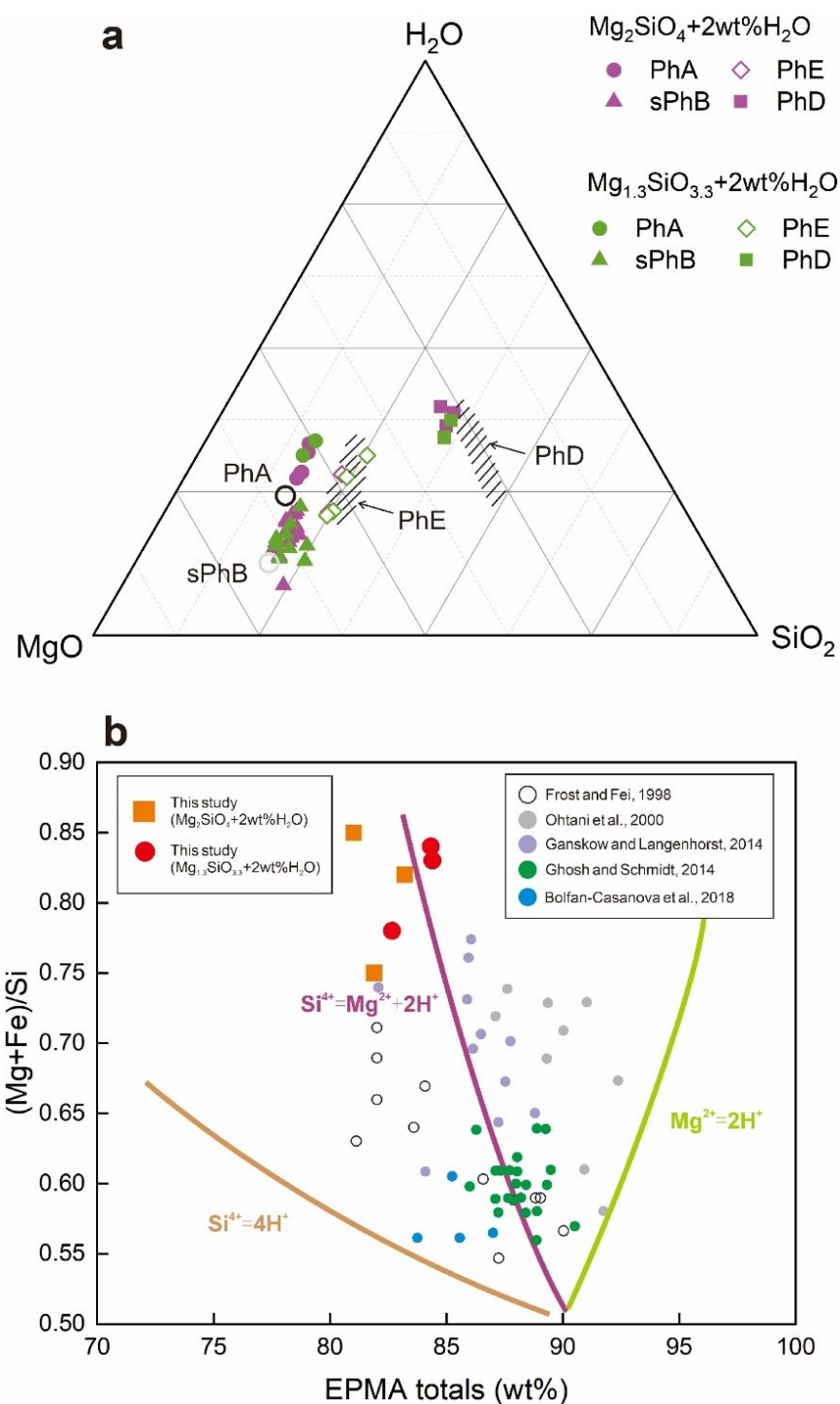
Supplementary Figure 4. Chemical composition of forsterite/wadsleyite/ringwoodite in this study and previous studies. Chemical compositions of the run products with temperature in $\text{Mg}_2\text{SiO}_4 + 2 \text{ wt\% H}_2\text{O}$ system (a) and $\text{Mg}_{1.3}\text{SiO}_{3.3} + 2 \text{ wt\% H}_2\text{O}$ system (b). Chemical composition of wadsleyite/ringwoodite as a function of H_2O in previous studies¹⁻⁸ in $\text{MgO-SiO}_2\text{-H}_2\text{O}$ systems. The hydrogen substitution in hydrous wadsleyite and ringwoodite generally follows the $\text{Mg}^{2+}=2\text{H}^+$ trend.



Supplementary Figure 5. Examples of unpolarized FTIR spectra of wadsleyite and ringwoodite in the $\text{Mg}_2\text{SiO}_4 + 2 \text{ wt\% H}_2\text{O}$ system. (a) FTIR spectra of wadsleyite. Samples from top to bottom: H868 (14 GPa, 1100 °C, 0.81 wt% H_2O); H844 (14 GPa, 1200 °C, 0.68 wt% H_2O); 5k3929 (18 GPa, 1200 °C, 1.77 wt % H_2O); 5k3943 (18 GPa, 1300 °C, 1.20 wt% H_2O); (b) FTIR spectra of ringwoodite. Samples from top to bottom: 5k3949 (20 GPa, 1300 °C, 1.28 wt% H_2O); 5k3931 (20 GPa, 1400 °C, 1.12 wt% H_2O); 1k3750 (22 GPa, 1400 °C, 0.83 wt% H_2O).

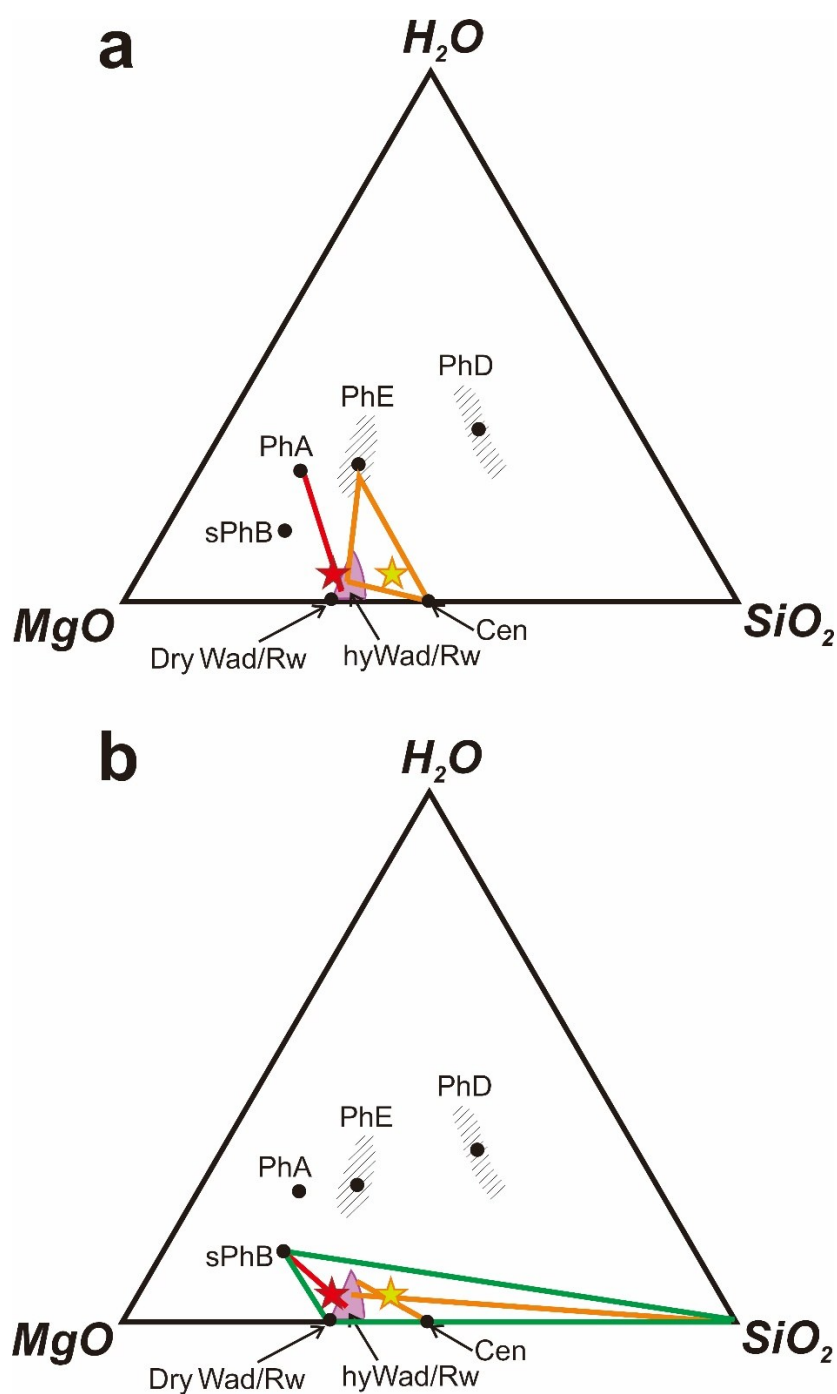


Supplementary Figure 6. Comparison of water contents determined by FTIR (y-axis) and Mg/Si ratios (x-axis) in chemical formula of wadsleyite and ringwoodite.

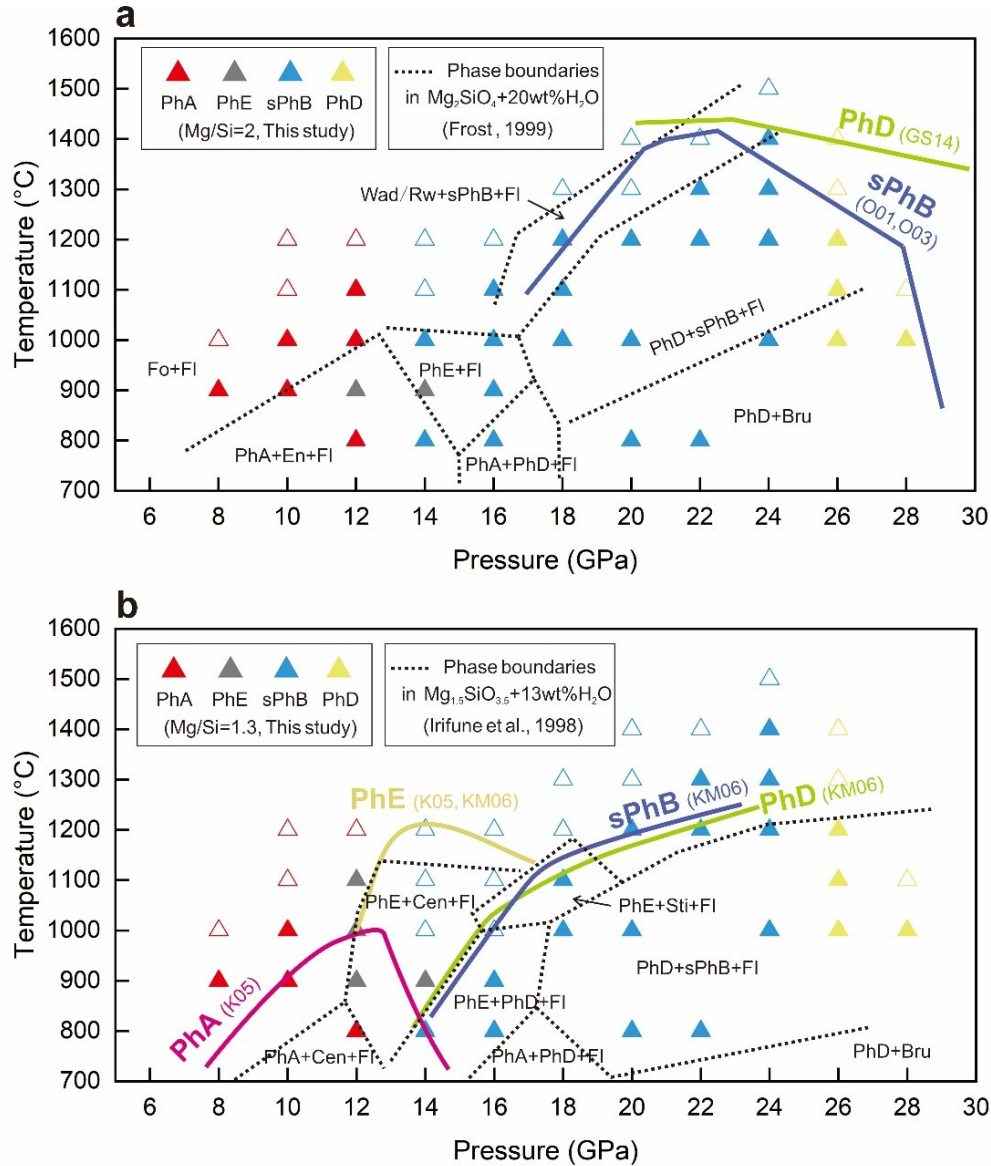


Supplementary Figure 7. Chemical composition of hydrous minerals. (a) Ternary diagram illustrating the chemical composition of hydrous minerals in this study (H_2O contents were calculated by mass deficit of EPMA data) and previous studies⁹. The circles denote the compositions of phase A (PhA) and superhydrous phase B (sPhB), which are derived from their ideal chemical formulas, $\text{Mg}_7\text{Si}_2\text{O}_8(\text{OH})_6$, $\text{Mg}_{10}\text{Si}_3\text{O}_{14}(\text{OH})_4$, respectively. The striped fields represent the broad compositional

variations of phase E (PhE) and phase D (PhD) reported in previous studies on MgO-SiO₂-H₂O systems (See Ref. 9). (b) (Mg+Fe)/Si ratio of PhD in this study and previous studies^{2,6,10-12} with EPMA totals.

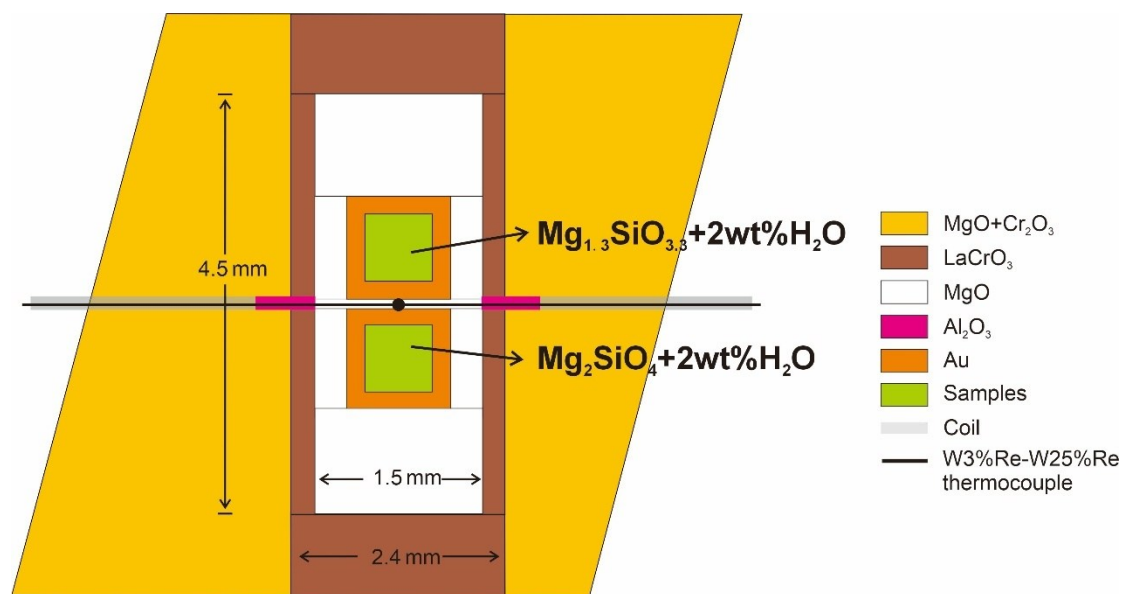


Supplementary Figure 8. Ternary phase diagram in the MgO-SiO₂-H₂O system at 12 GPa, 1000–1100 °C and at 14–18 GPa, 1000–1200 °C. The red and green stars denote the compositions of the starting materials with Mg/Si of 2.0 and 1.3, respectively. The pink region denotes the composition of hydrous wadsleyite and hydrous ringwoodite. See more information in Supplementary Note 1 about the effect of Mg/Si ratio on the stability of hydrous minerals.



Supplementary Figure 9. Stability of hydrous minerals in $\text{MgO-SiO}_2\text{-H}_2\text{O}$ systems.

(a) Comparison of the stability of hydrous minerals in $\text{Mg}_2\text{SiO}_4 + 2 \text{ wt}\% \text{H}_2\text{O}$ by this study and previous studies with high bulk water content. Stability of PhA, PhE, sPhB, PhD and phase assemblages (in black labels) are from the $\text{Mg}_2\text{SiO}_4 + 20 \text{ wt}\% \text{H}_2\text{O}$ system¹³. Stabilities of monomineralic sPhB (O01, O03)^{14,15} and PhD (O01, GS14)^{12,14} are also shown. (b) Comparison of the stability of hydrous minerals in $\text{Mg}_{1.3}\text{SiO}_{3.3} + 2 \text{ wt}\% \text{H}_2\text{O}$ by this study and previous studies with high bulk water content. The black labels denote the phase assemblages reported in $\text{Mg}_{1.5}\text{SiO}_{3.5} + 13 \text{ wt}\% \text{H}_2\text{O}$ ¹⁶. Stabilities of PhA, sPhB are from $\text{Mg}_{1.4}\text{SiO}_{3.4} + 3.66 \text{ wt}\% \text{H}_2\text{O}$ (PhA, sPhB) and PhE, PhD are from $\text{Mg}_{1.39}\text{SiO}_{3.39} + 12.2 \text{ wt}\% \text{H}_2\text{O}$ systems (K05, KM06)^{17,18}.



Supplementary Figure 10. A schematic drawing of a cell assembly using a Cr₂O₃-doped MgO octahedron with 8-mm edge length. For experiments using 10-mm edge length MgO octahedra, the cylindrical LaCrO₃ heater has an outer diameter of 3.0 mm and an inner diameter of 2.2 mm.

Supplementary Note 1 : Effect of Mg/Si ratio on the stability of hydrous minerals

Different Mg/Si molar ratios yield distinct phase relations and the stability of hydrous minerals. The Mg-rich composition prefers the formation of high Mg/Si phases (e.g., PhA, sPhB) while the Si-rich composition expands the stability of low Mg/Si phases (e.g., PhE). In this study, at 12 GPa and 1000–1100 °C, a three-phase assemblage of hydrous wadsleyite, clinoenstatite, and PhE appeared in the system of Mg/Si=1.3. By contrast, a two-phase assemblage of hydrous wadsleyite and PhA formed in the system of Mg/Si=2, owing to the bulk composition being situated on the PhA-hydrous wadsleyite tie-line (Supplementary Fig. 7a). Consequently, the thermal stability of PhA decreased while that of PhE expanded in the system with a low Mg/Si ratio, although the general dehydration temperature at 8–12 GPa remained constant in the two systems (Fig. 1).

At 14–18 GPa and 1000–1200 °C, hydrous wadsleyite and sPhB formed in Mg/Si=2 system while hydrous wadsleyite and clinoenstatite or stishovite were observed in Mg/Si=1.3 system, which could be explained by hydrous wadsleyite-sPhB tie-line and hydrous wadsleyite-clinoenstatite/stishovite tie-lines (Supplementary Fig. 7b). As a result, the dehydration temperatures of sPhB are ~100 °C higher in Mg₂SiO₄ + 2 wt% H₂O system than those in Mg_{1.3}SiO_{3.3} + 2wt% H₂O system at 14–18 GPa. Thermal stability of sPhB under water-saturated conditions also tends to be larger in high Mg/Si ratio system^{13,17,18}(Supplementary Fig. 8).

Although the P-T stability of PhD is nearly identical in two systems studied, it is expanded in the Si-rich composition under water-saturated conditions. At 20–24 GPa and 1000–1200 °C, sPhB, stishovite, and fluid were observed in Ohtani et al. (2000)² (Mg/Si=2, H₂O=11 wt%). However, PhD appears alongside sPhB and fluid in Komabayashi and Omori (2006)¹⁸ (Mg/Si=1.4, H₂O=12.2 wt%) at 21.5 GPa and 1000–1200 °C and Irifune et al. (1998)¹⁶ (Mg/Si=1.5, H₂O=13 wt%) under similar P-T conditions. With more silica incorporated, PhD coexists with stishovite and sPhB at 22 GPa and 1200–1400 °C¹² (Mg/Si=0.5, H₂O=10.09 wt%).

Supplementary Note 2 : Water-content gradient in the subducting slab

Water initially present in the oceanic plate originates from the hydration of slab prior to subduction. The seismological evidence indicates that subducting oceanic lithosphere undergoes extensive hydration even down to the cold core or lower portion of slab via bending-fault¹⁹⁻²³ or interactions with hydrated mantle plume^{24,25}. Upon subduction, hydrous minerals (e.g., serpentine) formed at shallow depths progressively transform into DHMSs (e.g., PhA) as the slab descends to the 410-km discontinuity^{13,17,18} when the hydrated lithosphere remains below “choke point”²⁶. The water stored in these DHMSs, in combination with that in NAMs (e.g., olivine), constitutes the primary water budget for subducting lithosphere below 410 km depth.

In the cold interior of a slab, NAMs remain dry, and water is mainly accommodated in hydrous minerals. Within the stability of hydrous minerals, water

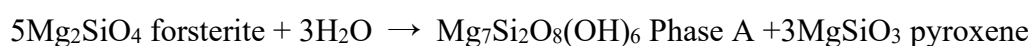
remained partitioned between hydrous minerals and NAMs. The interactions between hydrous minerals and NAMs with increasing temperature result in progressive hydration of coexisting NAMs and diminishing amounts of DMHSs. Hence, the water-content gradient within the hydrated interior results from solid-state reactions between DHMSs and NAMs via short-range redistribution of hydrogen (Fig. 4).

As temperatures approach the complete dehydration temperature near the slab surface (at the top of the harzburgitic layer and the bottom of the peridotitic layer), the water contents in NAMs peak. This maximum water content in NAMs can occur near the surface or at the region between the slab surface and the cold core. The precise location depends on the thermal structure. An older, colder slab would tend to place the gradient closer to the slab surface, whereas a younger, warmer slab would shift the gradient toward the interior between the surface and the cold²⁷. If the slab temperature near the surface is above the complete dehydration temperature, water is partitioned between hydrous fluid/melt and NAMs. This water redistribution is enhanced by the transport of hydrous fluid or melt. Thus, the water-content gradient in the vicinity of slab surface is mainly mediated via hydrous fluid or melt (Fig. 4).

We note that the hydrogen diffusion in major NAMs, as determined by previous studies^{28,29}, is difficult to permit efficient kilometer-scale redistribution from the cold slab interior to its warmer outer regions over geological timescales. On the other hand, hydrogen can be fully homogenized at grain-size scale (on the order of millimeters to centimeters). Here we emphasize that our model does not rely on long-range hydrogen

diffusion responsible for generating the suggested gradient. Instead, the hydration-dehydration model proposed in this study arises from: 1) the water budget inherited from the initial hydration nature of descending plate shallower than 410 km depths; 2) short-range redistribution relevant to mineral transformation and solid-state reactions within the hydrated interior; and 3) efficient fluid/melt-mediated transport near the slab surface, alongside the slab thermal structure (Fig. 4).

We also emphasize that limited hydrogen mobility between NAMs and DHMSs does not imply local hydration of NAMs, nor does it promote metastable olivine transformation through locally hydrated reaction zones. This is because the thermodynamic driving force overwhelmingly favours hydrogen incorporation into DHMSs rather than into NAMs. Indeed, Ishii and Ohtani (2021)³⁰ demonstrated that even when hydrous forsterite was used as the starting material, forsterite coexisting with Phase A became anhydrous (<1 ppm H₂O). This is consistent with the reaction:



which shows that DHMSs act as a strong sink for hydrogen, efficiently extracting hydrogen from forsterite. Therefore, even transient hydrogen exchange at grain boundaries does not lead to meaningful hydration of NAMs through DHMS, and olivine remains effectively dry at cold core temperatures. Several previous studies have specifically investigated the kinetics of phase transformation using pre-hydrated olivine with homogeneous hydrogen distribution³¹⁻³², which differs fundamentally from the transformation kinetics in hydrous-phase-bearing coexistence systems investigated

here and water-partitioning experiments²⁸. Future kinetic experiments under controlled water-undersaturated and hydrous-phase-bearing coexistence conditions would be valuable for further quantifying this behaviour.

Supplementary Note 3: Pressure calibration of multi-anvil press

Pressures of the high-pressure experiments were calibrated based on several phase transitions: SiO₂ coesite-stishovite (9.2 GPa, 1200 °C)³³, Mg₂SiO₄ forsterite-wadsleyite (13.6 GPa, 1200 °C)³⁴, Mg₂SiO₄ wadsleyite-ringwoodite (20.8 GPa, 1600 °C)³⁵, Mg₂SiO₄ akimotoite-bridgmanite (21.0 GPa, 1600 °C)³⁶, Mg₂SiO₄ ringwoodite-bridgmanite + periclase (23.6 GPa, 1600 °C)³⁶, and Mg₃Al₂Si₃O₁₂ pyrope-bridgmanite + corundum (25.6 GPa, 1600 °C)³⁷.

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