The Effect of Water on the 410-km Discontinuity: An Experimental Study

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Abstract. In order to evaluate the effect of H on the pressure and transition interval of the olivine-wadsleyite transformation, synthesis experiments were carried out in hydrous and anhydrous peridotite compositions at 1400°C. Under hydrous, near-saturated conditions, the pressure of the transition is 1.0 ± 0.5 GPa lower than under anhydrous conditions. In addition, the two-phase interval appears to broaden from about 0.4 GPa (12 km) in the anhydrous system to as much as 1.3 GPa (40 km) in the hydrous system. Under hydrous conditions the spatial distribution of olivine and wadsleyite appears to be controlled by H diffusion. Extrapolating previously measured diffusion coefficients for H in olivine and in liquids, it appears that H diffusion driven by gravitational equilibration may serve to narrow the transition interval between olivine and wadsleyite.

Introduction

The seismic discontinuities at depths of 410 and 660 kilometers, which bound the Transition Zone, are generally thought to be due to phase changes in an olivine-dominated mantle composition. The discontinuity at 410 kilometers corresponds to the olivine-wadsleyite transition with an increase in both density and S-wave velocity of about 5% (Dziewonski and Anderson, 1981). In the dry peridotite system the olivine-wadsleyite transition is observed experimentally at 13.5 to 14 GPa at about 1600 °C with a transition interval of at least 0.5 GPa (~15 km) (Zhang and Herzberg, 1994). Based on thermochemical measurements, Akaogi et al (1989) suggest an interval of 18 km. Seismically, the discontinuity is seen as a very abrupt rise in velocity. Several authors have reported discontinuity thicknesses of four km or less (Benz and Vidale, 1993, Yamazaki and Hirahara, 1994, Neele, 1996).

Based on electrostatic bond strength calculations, Smyth (1987) suggested that wadsleyite has the potential to hydrate by protonation of its non-silicate oxygen (O1). If this site were fully protonated, the structure might contain as much as 3.3 weight percent H₂O (Smyth 1994). Gasparik (1993), Inoue (1994), and Kohlstedt et al (1996) observed 3.1 weight percent or more H₂O in wadsleyite under near water-saturated conditions. This is a geologically significant amount of hydrogen. If saturated, a transition zone containing 70 modal percent wadsleyite could contain four times the amount of water in the hydrosphere and be the largest reservoir of H in the planet (Inoue et al., 1995).

Kudoh et al. (1996) and Smyth et al. (1997) reported the crystal structure of hydrous wadsleyites and observed cation vacancies in both the Mg and Si sites. Smyth et al. (1997)

and Kudoh and Inoue (1999) observed that hydrous wadsleyite may be metrically monoclinic (*I2/m*) instead of the nominally orthorhombic (*Imma*) structure, but that there is complete crystalline solution between the anhydrous and hydrous varieties. Recently, we have observed that the deviation from orthorhombic symmetry appears to vary with trivalent cation content. Also, the incorporation of H into this structure has a relatively small effect on the unit cell volume so that the hydrous wadsleyite is still about five percent denser than olivine, which can incorporate only up to about 0.2 weight percent H₂O.

Smyth and Kawamoto (1997) reported an additional variation of the wadsleyite structure similar to spineloid IV (Horioka et al., 1982), which may be optically indistinguishable from wadsleyite. This structure is intermediate in density and pressure stability between wadsleyite and ringwoodite, but also may require significant amounts of H and trivalent cations for stabilization. Most model mantle compositions have sufficient Al for this phase to be a possible constituent of the Transition Zone, and its presence is another possible explanation for the splitting of the discontinuity at 520-km recently reported by Deuss and Woodhouse (2001).

Wood (1995) explored some of the potential thermodynamic consequences of a hydrous Transition Zone. He reasoned that the presence of H in the Transition Zone, being more soluble in wadsleyite than olivine must expand the stability field of wadsleyite to lower pressures through configurational entropy of disorder. He then calculated, based on an assumed model for the incorporation of H into olivine and wadsleyite, that the presence of 500 ppm H would necessarily expand the transitional $\alpha + \beta$ loop interval from a minimum of about 7 km for a dry system to at least 22 km. He went on to assert that the observed sharpness of the 410-km discontinuity would be inconsistent with H₂O contents greater than about 200 ppm. Hellfrich and Wood (1996) estimated that the effect of a 10-km transition interval might appear seismically as a 5-km linear velocity gradient. They also concluded in their analysis that the reflectivity of the 410-km discontinuity is much more sensitive to varying H₂O concentration than to temperature. However, most estimates of the width of the transition interval for the dry system range from 15 to 18 km based on experimental studies. It therefore appears that the observed transition interval is too narrow for the dry system and would be much too narrow for a hydrous system, unless some other phenomenon is controlling the width.

In order to experimentally test the hypothesis of Wood (1995) that the presence of H₂O in the Transition Zone

would extend the stability of wadsleyite to lower pressures and broaden the two-phase region, we have conducted a series of synthesis experiments in the peridotite and olivine systems. We conducted a series of three preliminary synthesis experiments in a hydrous olivine system and a further series of three experiments each in hydrous and anhydrous peridotite systems. By measuring the compositions of the coexisting olivine and wadsleyite by the electron microprobe, we can obtain an estimate of the difference in the width of the transition between hydrous and anhydrous conditions. We also hoped to observe a significant pressure difference for the transition between the wet and dry conditions. We may also be able to observe additional phenomena that might affect the width of the transition interval.

Experimental

Three preliminary experiments were conducted in a hydrous olivine system with very low trivalent cation content. Starting material (Table 1) was formulated from powdered natural Fo₉₀ olivine, brucite, quartz, and wüstite to achieve a composition of Fo₉₂ plus three weight percent H₂O with a slight excess of silica. Three principal experiments were conducted in a hydrous peridotite system with a slight excess of silica and alumina. The starting material was formulated from powdered natural Fo₉₀ olivine, plus synthetic brucite, quartz, pyrope and wüstite to achieve a composition of Fo₈₈ plus three weight percent H₂O with a sufficient excess of silica and alumina to give approximately five normative percent each of pyroxene and pyrope-almandine garnet. Finally, three experiments were conducted in an anhydrous peridotite system. The starting material was formulated from powdered natural Fo₉₀ olivine, plus synthetic quartz, pyrope and wüstite to achieve a composition of Fo₉₀ with a sufficient excess of silica and alumina to give approximately five normative percent each of pyroxene and pyrope-almandine garnet.

The charges were sealed in welded Pt capsules 2 mm in diameter and 3.5 mm in length. A Cr₂O₃-doped MgO multianvil octahedron of 18 mm edge-length was employed with a stepped lanthanum chromate heater. The capsule was placed into the assembly inside an MgO sleeve. A 5000-ton press at the Bayerisches Geoinstitut was used to compress the octahedra using 52-mm edge-length WC anvils with 11-mm corner truncations. The heating time for each experiment was nominally 5 hours. The temperature was monitored and controlled to within 1°C during the experiments using a W3%Re/W25%Re thermocouple that was inserted axially into the assembly. The 18/11 multi-anvil assembly was calibrated using the coesite to stishovite transition at 1000°C (Zhang et al., 1996), the Mg₂SiO₄ forsterite to wadsleyite transition at 1200°C (Morishima et al., 1994) and the Mg₂SiO₄ wadsleyite to ringwoodite transition at 1200°C (Suzuki et al., 2000). The Fe-Mg partitioning between phases in the anhydrous peridotite experiments was compared with the results of Katsura and Ito (1989) to provide an additional constraint on the reproducibility of the pressure measurements. The accuracy in the absolute pressure measurement is around 1 GPa when the uncertainties in the calibrant pressure estimates are considered but the reproducibility of the experimental pressure is within 0.5 GPa.

The possible presence of wadsleyite II (Smyth and Kawamoto, 1997) or of monoclinic wadsleyite (Smyth et al., 1997), necessitated characterization of run products by single crystal X-ray diffraction if possible. Capsules were opened and polished for chemical analysis by electron microprobe. In addition, crystals were selected for single-crystal X-ray diffraction study. Experimental compositions, run-conditions, and products are listed in Table 1. Determination of H contents run products is beyond the scope of the current report. The most reliable methods for H content determination for olivine is by ¹⁵N nuclear reaction analysis or by polarized IR spectroscopy on oriented, faceted single crystals (Bell et al, 2002). Reliable calibration of the IR spectra of wadsleyite for H-content is not yet available, but is the subject of ongoing study.

Discussion

The compositions of co-existing olivine and wadsleyites are shown in Figure 1. In the anhydrous system, the width of the two-phase region at 1400°C appears to be about 0.4 GPa or about 12 km depth interval. This is consistent with previous experimental and thermochemical studies, although at the high end of the 4 to 12 km advocated by Hellfrich and Wood (1996). In the hydrous system, the two phase regions widens to about 1.3 GPa or about 40 km, and the pressure of the transition is 1.0 ± 0.5 GPa lower than in the anhydrous system. The estimate of the width of the two-phase region is based on the chemical analyses, and not on the pressure calibration. The results of our synthesis experiments are thus consistent with the prediction of Wood (1995) that the presence of H₂O extends the stability of wadsleyite to 0.6 to 1.0 GPa lower pressure and would necessarily broaden the two-phase loop to as much as 40 km. So, both the observed 40 km interval in the hydrous system and 12 km interval in the anhydrous system are still too broad to be consistent with the seismic observations.

It should be noted that in the hydrous system, the largest difference between the wadsleyite and olivine compositions is not the Fe content, but the H content, with the H content of wadsleyite being about ten times that of olivine. Moreover, in all of the experimental hydrous runs containing both olivine and wadsleyite, there appears a sharp boundary between the regions of olivine and wadsleyite. The texture of the hydrous runs may be responding to small thermal gradients and does not appear to be a simple chemical equilibrium, but rather a diffusion-controlled boundary. This is in sharp contrast to the anhydrous runs in which the wadsleyite grains appear evenly distributed throughout the capsule. Hydrogen is known to diffuse very rapidly in these materials. This raises the possibility that diffusion of H might control the texture and may affect the sharpness of the boundary in the natural system.

Solid-state diffusion of H is rapid in single-crystal olivine (Mackwell and Kohlstedt, 1990). Charge compensation is provided by a divalent cation vacancy, and vacancy diffusion is also very rapid (Mackwell et al., 1988). Hydrous wadsleyite is still about 5% more dense than anhydrous olivine (e.g. Smyth et al., 2000). This is comparable to the relative density difference between water and ice. In a hypothetical two-phase region consisting of olivine and wadsleyite plus lesser amounts of garnet and clinopyroxene extending over a depth of 20 km in a hydrous

system, gravitational equilibrium can be approached by diffusion of H without the much slower movement of iron. This gravity-driven diffusion may enrich the lower parts of the two-phase region in wadsleyite and the upper part in olivine, thus sharpening the boundary. This plausible mechanism may sharpen the apparent boundary to four kilometers or less as indicated in some seismic studies (Benz and Vidale 1993, Vidale et al., 1995). Because diffusion of H is considerably more rapid than convection over short distances, it might cause the transition zone to serve as a trap for H in the Earth.

It is possible to estimate some of the parameters that might constrain such an effect. First, the velocity of diffusion will constrain H distribution equilibrium. Mackwell and Kohlstedt (1990) measured diffusion coefficients for H in solid-state, single-crystal olivine at between 10^{-9} and 10^{-10} m^2s^{-1} at 800 and 1000 °C. These would extrapolate to about 10^{-8} to 10^{-9} at 1400°C. These values are large enough to achieve H distribution equilibrium in grains that are millimeters in diameter in a matter of minutes or, over a 5 cm annual convection movement in less than a day. Because the equilibration time scales with the square of distance, H diffusion could not stay ahead of convection in an actively convecting region of the mantle. If extrapolated, these values indicate equilibration over 10 km in about 300 million years. These diffusion coefficients values were obtained at pressures of 300 MPa where olivine has as relatively low H content (~200 ppm H_2O by weight). At pressures above 10 GPa, Kohlstedt et al. (1996) observed H contents an order of magnitude higher (~2000 ppm H_2O by weight). If diffusion coefficients increase with increasing concentration of H defects, diffusion would be even more rapid due to the higher H contents. Also, there are no solid-state diffusion data for H in wadsleyite, but the maximum H_2O contents are an order of magnitude higher than the high-pressure olivine (~33,000ppm by weight), and Chakraborty et al (1999) report cation diffusions rates one to two orders of magnitude faster in wadsleyite than in olivine.

Grain boundaries in mantle rocks that have undergone phase changes tend to be complex polyphase assemblages that can constitute up to several percent of the rock (McCormick et al., 1994). In such rocks, movement of H may be controlled by diffusion in grain boundaries, so the single-crystal values cited above can be regarded as lower bounds. Grain boundary diffusion of H is likely to approach that of mafic magmas. Chekhmir et al (1985) suggest a diffusion coefficient of 10^{-6} m^2s^{-1} for H in molten basalt, which is two to three orders of magnitude larger than in the crystals, and comparable to the diffusion of heat (Watson, 1991). It is difficult to estimate the contribution of grain boundaries, but it will vary with grain size and may constitute one percent of the cross-sectional area in some mantle rocks. Whole-rock diffusion coefficients for H in peridotite at 1400°C may then be one to two orders of magnitude larger than for single-crystals, or sufficient to allow equilibration over a 10 km interval in a few tens of millions of years. Some additional experimental data on whole-rock and single-crystal diffusion of H under pressure and temperature conditions relevant to the 410-km discontinuity may help to further constrain this phenomenon.

Figure 1. Plot of experimental results at 1400°C. Solid symbols represent olivine and open symbols wadsleyite. Gray symbols and boundaries represent the hydrous experiments and black the anhydrous.

Second, it is possible to estimate the driving force for establishment of gravitational equilibrium. If we take a hypothetical one-by-one meter volume across a 20-km two-phase region, and re-establish gravitational equilibrium by moving all of the wadsleyite to the bottom and olivine to the top we release an energy, E , given by

$$E = V\Delta\rho gh,$$

where V is the volume which would be 2500 m^3 for wadsleyite in the upper half of the volume. $\Delta\rho$ is the density difference between wadsleyite and olivine taken as 5% or 150 kg/m^3 ; g is the acceleration of gravity, 9.8 ms^{-2} ; and h is distance the volume moves to re-establish equilibrium. This is approximately 6 km for the 20-km column (center of mass for the upper volume to the center of mass of the lower volume). This gives about 2.2×10^{10} j for the volume undergoing reaction, or about 2.5 kJ/kg, enough to raise the temperature of the volume by approximately one degree Celcius. This neglects smaller PdV term and assumes balance in the heats of reaction, but it may provide a sufficient amount of energy to act as a driving force for the establishment of gravitational equilibrium in regions where there is little vertical movement of material across the boundary. Comprehensive computer modeling will be required to further constrain the possibility of this phenomenon, but is beyond the scope of the current study.

In summary, we have carried out a series of experimental synthesis runs in order to evaluate the effect of H_2O on the position and sharpness of the 410-km discontinuity. Run products have been carefully characterized by electron microprobe and single-crystal X-ray diffraction. Under near-saturated conditions, the pressure of the transition is 1.0 ± 0.5 GPa lower than under anhydrous conditions. In addition, the two-phase interval appears to broaden from about 0.4 GPa (12 km) in the anhydrous system to as much as 1.3 GPa (40 km) in the water-saturated system. Chemical analysis indicates that the largest chemical difference between olivine and wadsleyite is the hydrogen content. Textures in the hydrous experiments suggest that H diffusion controls the spatial distribution of the olivine and wadsleyite phases. Evaluating this possibility, diffusion coefficients for H in solid-state single-crystal olivine appear to be large enough to allow H distribution equilibration over a 20-km interval in a few hundred million years, or less if grain boundary effects

are considered. In any case, H diffusion cannot outpace convection in regions of significant convective flow across the boundary. However such regions are confined to subduction zones and spreading centers, and seismic observations of the narrow boundary regions were not made in these regions.

If this diffusion mechanism does indeed function to control the distribution of olivine and wadsleyite near the 410 km discontinuity, then the presence of H may cause the transition at 410 km to be sharper and shallower and velocities in the region slower than under anhydrous conditions. Under cool, anhydrous conditions the transition would be shallower and velocities faster than under hot anhydrous conditions. Further work will be required to quantify the effect of H on elastic properties, but it appears that the effect of H on the depth of the transition and on velocities will be larger than possible variation in temperature.

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Table 1. Results of Synthesis Experiments

Run	SS0001	SS0003	SS0004	SZ0101	SZ0102	SZ0103	SZ0109	SZ0110	SZ0111
Composition*	1	1	1	2	2	2	3	3	3
P(GPa)	12.0	12.0	12.0	13.2	11.8	12.5	13.2	13.9	12.5
T(°C)	1200	1400	1600	1400	1400	1400	1400	1400	1400
Olivine									
Fo#	97.8	92.7	92.		91.0	92.9	91.2		92.4
Modal%	15	100	100	0	90	50	50	0	98
Wadsleyite									
Fo#	90.8			87.7	84.0	84.2	88.2		88.2
Modal%	85	0	0	100	10	50	50	95	2
Garnet									
Modal%	0	0	0	3	5	5	5	5	5
Clinopyroxene									
Modal%	0	0	0	5	5	5	0	0	0

*Composition 1. Hydrous olivine: approximately Fo₉₂ olivine plus 3% H₂O.

*Composition 2. Hydrous peridotite: approximately Fo₈₉ olivine plus 3% H₂O with minor garnet and excess silica.

*Composition 3. Anhydrous peridotite: approximately Fo₉₂ olivine with minor garnet and excess silica.