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Elasticity of hydrous wadsleyite to 12 GPa: Implications for Earth's transition zone

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[1] Knowledge of the pressure effect on elasticity of hydrous olivine polymorphs is necessary to model seismic wave speeds for potential hydrous regions of the mantle. Here we report single-crystal elastic properties of wadsleyite, β-Mg_2SiO_4, with 0.84 wt.% H_2O measured to 12 GPa by Brillouin scattering. Pressure derivatives of the aggregate bulk modulus, K'_{S0}, and shear modulus, G'_0, of hydrous wadsleyite are 4.1(1) and 1.4(1) respectively. These values are indistinguishable within uncertainty from those of anhydrous wadsleyite. We estimate that $\sim$1 wt.% H_2O in wadsleyite at 410-km depth can reconcile seismic bulk sound velocities with a pyrolite-composition mantle by using our measured high-pressure elastic constants. If the H_2O content of the mantle is much less than 1 wt.%, then other factors need to be considered to explain the velocity contrast of the 410-km discontinuity. Variations in water content with depth may also contribute to the anomalously steep seismic velocity gradient in the mantle transition zone.

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1. Introduction

[2] Wadsleyite, β-Mg_2SiO_4, is potentially a major hydrogen host in the transition zone because of its high water storage capacity (up to 0.9 wt.% H_2O at 15 GPa, 1400°C [*Demouchy et al.*, 2005]). Determination of the effect of water on the elasticity of wadsleyite can provide constraints on the water content in the Earth's transition zone through comparison with seismic data. Our previous study [*Mao et al.*, 2008] showed that the elastic moduli of wadsleyite decrease strongly with increasing water content at ambient conditions: 1 wt.% H_2O in wadsleyite leads to a 7–8% decrease in the bulk and shear moduli. This corresponds to $\sim$3% decrease in the compressional and shear wave velocities at ambient conditions. Pressure derivatives of the bulk and shear moduli are needed to extrapolate elastic moduli to high pressures. The pressure derivative of the isothermal bulk modulus, $(\partial K_T/\partial P)_{T0} = K'_{T0}$, for hydrous wadsleyite was previously investigated by static compression studies and no obvious systematic trend between the pressure derivatives and water content has emerged [*Yusa and Inoue*, 1997; *Holl et al.*, 2008].

[3] Here, we report the single-crystal elasticity of wadsleyite with 0.84 wt.% H_2O to 12 GPa by Brillouin scattering. Using our measured elastic constants, we examine the effect of water on the bulk sound velocities of wadsleyite at Earth's transition zone conditions. The effect of water on the velocity contrast between olivine and wadsleyite at 410-km depth is also evaluated. We also examine how variation of water content in wadsleyite with depth may affect the seismic velocity gradient in the transition zone.

2. Experimental Details

[4] Single crystals of wadsleyite, β-Mg_2SiO_4 with 0.84 wt.% H_2O were synthesized from mixed oxide powders in a multi-anvil press at 14–16 GPa and 1200–1400°C. Recovered samples were examined by single-crystal x-ray diffraction and infrared and Raman spectroscopy [*Mao et al.*, 2008]. The concentration of H_2O was estimated from the empirical relationship between the *b*/*a* ratio and water content as determined by *Jacobsen et al.* [2005]. The uncertainty in H_2O content is estimated to be ±10%. Further details about sample synthesis and characterization are reported elsewhere [*Mao et al.*, 2008].

[5] Three individual platelets with different crystal orientations were used in the Brillouin measurements. Each was plane-parallel polished to $\sim$30 μm thickness and loaded into diamond anvil cells together with a methanol-ethanol-water pressure medium. Several ruby chips were also loaded in each cell for pressure determination. For each platelet, Brillouin measurements were performed at 7 pressures up to 12 GPa (Figure S1 of the auxiliary material).[1] All the spectra were measured in a symmetric forward scattering geometry using a six-pass tandem Fabry-Parot interferometer. We estimate there is ±0.5–1% uncertainty in the measured velocities [*Zha et al.*, 1996]. Further details of the Brillouin system are provided elsewhere [*Speziale and Duffy*, 2002].

3. Results

[6] Figure 1 compares the C_{ij} values determined in this study with those of anhydrous wadsleyite at high pressures [*Zha et al.*, 1997] (Table S1). The pressure derivative of each C_{ij} is obtained by fitting the measured moduli and density to third-order or fourth-order Eulerian finite strain equations (Table S2). At ambient pressure, most C_{ij}s of

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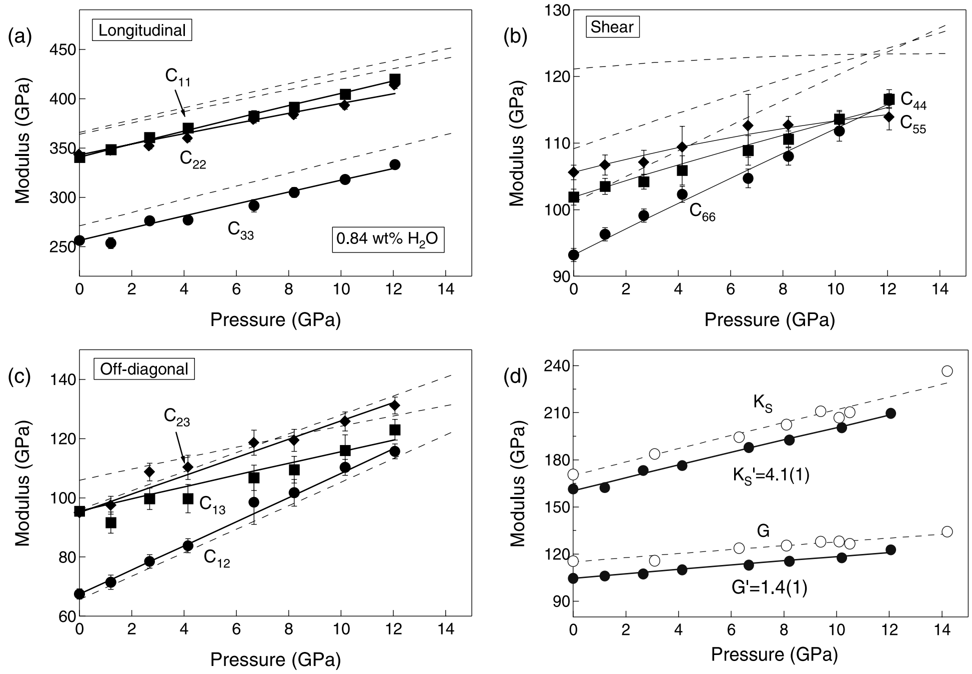

Figure 1. Elastic moduli of hydrous wadsleyite as a function of pressure compared with those of anhydrous wadsleyite [*Zha et al.*, 1997]. Solid symbols and lines: wadsleyite with 0.84 wt.% H_2O, (this study); dashed lines: anhydrous wadsleyite, [*Zha et al.*, 1997]. (a–c) Individual C_{ij} of hydrous wadsleyite; (d) adiabatic bulk, K_S, and shear moduli, G.

hydrous wadsleyite are offset to lower values compared with the anhydrous phase, with shear moduli showing the largest reduction. Upon compression, the moduli of both the hydrous and anhydrous phases increase with generally similar slopes with largest differences observed for C_{55}, C_{13}, C_{23} (Figure 1 and Table S2). Some of the differences in trends may reflect uncertainty in the C_{ij}s of anhydrous wadsleyite [*Zha et al.*, 1997] for which Brillouin data were recorded in only two crystal planes, compared with three in this study.

[7] From the single-crystal elastic moduli, we calculated the VRH (Voigt-Reuss-Hill) average to determine the adiabatic bulk and shear moduli for polycrystalline aggregates (Table S1). Figure 1d shows the aggregate bulk and shear moduli as a function of pressure. Third-order finite Eulerian strain equations are used to obtain the pressure derivatives of the adiabatic bulk and shear moduli, yielding: K'_{S0} = 4.1(1), G'_0 = 1.4(1), where numbers in parentheses are 1 standard deviation uncertainties in the last digit(s). For comparison, measured pressure derivatives of anhydrous wadsleyite are K'_{S0} = 4.3(2) and G'_0 = 1.4(2) [*Zha et al.*, 1997; *Li et al.*, 1998]. Thus, wadsleyite with 0.84 wt.% H_2O has pressure derivatives of the bulk and shear moduli that are indistinguishable from the anhydrous phase. While further studies of hydrous wadsleyites with a range of iron and water contents are needed, the pressure derivatives of aggregate elastic moduli do not appear to be sensitive to H_2O at least for water contents of ~1 wt.%. At a pressure of 12 GPa and 300 K, the compressional, bulk, and shear velocities in wadsleyite with 0.84 wt.% H_2O are 10.03(3), 7.53(3), and 5.75(3) km/s, which are 2.1–2.4% below anhydrous values at this pressure. For comparison, to produce a similar velocity reduction in anhydrous wadsleyite requires 500–600 K increase in temperature [*Mayama et al.*, 2004; *Isaak et al.*, 2007] or ~10 mol.% increase in iron content [*Mao et al.*, 2008].

4. Discussion

[8] To understand the implications of these results for the Earth's mantle, we have computed the bulk sound velocities as a function of depth for olivine and wadsleyite along a 1400°C adiabat using the approach outlined by *Duffy and Anderson* [1989] and the parameters listed in Table S3. We focus exclusively on bulk sound velocities here because these are least affected by anelasticity [*Karato*, 1993], which can markedly affect the temperature dependence of the shear velocity and may be enhanced by incorporation of hydrogen [*Karato*, 1995].

[9] To determine bulk sound velocities as a function of depth for olivine and wadsleyite, we need to specify for each phase its composition (Fe, H_2O content), density, thermal expansivity, adiabatic bulk modulus, and pressure and temperature derivatives of the bulk modulus. The effect of hydrogen on the elasticity of olivine is small at ambient conditions: 0.8 wt.% H_2O leads to 2–2.5% reduction for the bulk and shear moduli, with 0.2–0.5% reduction in the P and S wave velocities for fosterite [*Jacobsen et al.*, 2008]. Measurements of the effect of hydrogen on the pressure derivatives of hydrous olivine are in progress with publication forthcoming. Therefore, we restrict comparison of hydrous wadsleyite to the properties of anhydrous olivine in this study. The Fe contents of olivine and wadsleyite were taken to be 10 mol.% on average, and include the expected increase in Fe content of wadsleyite relative to olivine across the 410-km discontinuity [*Irifune and Isshiki*, 1998;

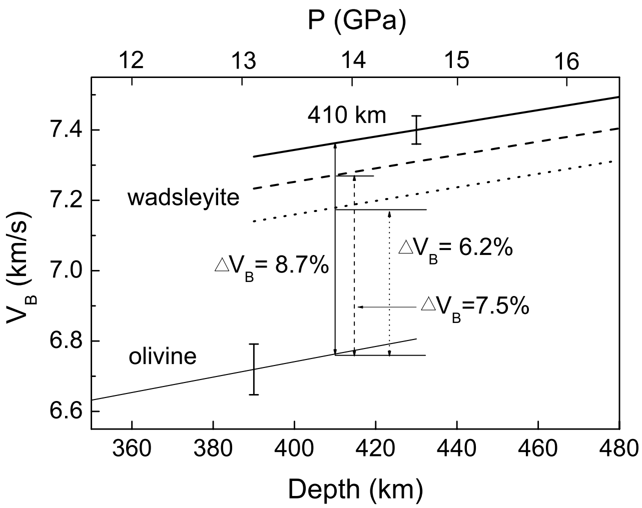

Figure 2. Bulk sound velocity as a function of depth along a mantle adiabat for anhydrous olivine and wadsleyite with variable hydrogen content. Pressure converted to depth using Earth model PREM [*Dziewonski and Anderson*, 1981]. Solid line: dry conditions; dashed line: 0.4 wt% H_2O in wadsleyite; dotted line: 0.8 wt% H_2O in wadsleyite. Representative uncertainties of the calculation are also shown.

Frost, 2003]. Thermal expansivity data for wadsleyite with 2.4 wt.% H_2O [*Inoue et al.*, 2004] were used with the assumption that thermal expansivity decreases linearly with water content. However, effects of hydrogen content on thermal expansion are small and results are not sensitive to this parameter.

[10] The seismic velocity increase at 410 km is often used to estimate the olivine volume content in the Earth's upper mantle [e.g., *Duffy and Anderson*, 1989]. Figure 2 shows bulk sound velocities in olivine and wadsleyite along a 1400°C adiabat. Under anhydrous conditions, the velocity contrast at 410-km depth between wadsleyite and olivine is 8.7(12)%. Because water affects the elasticity of wadsleyite strongly, the velocity contrast between hydrous wadsleyite and anhydrous olivine decreases to 6.2(13)% for wadsleyite with 0.8 wt.% H_2O.

[11] In the pyrolite model for the mantle, the olivine fraction is close to 60 vol.%. However, previous comparisons of mineral physics and seismic data have found that the olivine volume fraction at 410-km depth is only 35–53% [*Duffy and Anderson*, 1989; *Duffy et al.*, 1995; *Li et al.*, 1998; *Cammarano and Romanowicz*, 2007]. By studying PP and SS precursors from the 410-km discontinuity, *Chambers et al.* [2005] found values of the impedance contrast for both P and S waves significantly below those expected for a pyrolite mantle. Although *Shearer and Flanagan* [1999] have found impedance contrast consistent with pyrolite, other seismic studies [e.g., *Gaherty et al.*, 1999; *Rost and Weber*, 2002] also found a lower impedance contrast than for a pyrolite mantle. Hydration of the mantle has the potential to explain why the seismic velocity and impedance contrast at 410 km are lower than expected for pyrolite [*Li et al.*, 2001; *Chambers et al.*, 2005]. Bulk sound velocity profiles for the mantle can be obtained from global or regional seismic models that report P and S wave velocities [*Kennett et al.*, 1995; *Gaherty et al.*, 1996]. The bulk sound velocity contrast at 410 km in mantle seismic models is typically 3–5% [e.g., *Kennett et al.*, 1995; *Gaherty et al.*, 1996]. The olivine fraction at this depth can be estimated from the ratio of the seismic bulk velocity contrast to that calculated between olivine and wadsleyite from mineral physics data. Table 1 shows that the inferred olivine fraction of the mantle increases with increasing water content in wadsleyite for different seismic models. This is due to the reduction in bulk sound velocity of wadsleyite with hydration. A pyrolite composition is consistent with global model AK135 [*Kennett et al.*, 1995] and STW105 [*Kustowski et al.*, 2008] if wadsleyite contains more than 1 wt.% H_2O at 410-km depth. Seismic model PA5 for the Pacific Corridor [*Gaherty et al.*, 1996] requires ~0.6 wt.% H_2O in wadsleyite to match the pyrolite composition. For these cases, the amount of water requires a very hydrous transition zone that is close to or above the maximum H_2O storage capacity of wadsleyite at these conditions [*Demouchy et al.*, 2005]. Geochemical and geophysical evidence suggests the H_2O content of the transition zone may be much lower than this ($\leq$0.2 wt.%) [e.g., *Dixon et al.*, 2002; *Huang et al.*, 2005; *Hirschmann*, 2006]. In this case, the inferred olivine content is about 40(6)% for AK135, and 51(8)% for PA5 (Table 1), similar to the anhydrous case. The water content in wadsleyite at 410-km depth needed to match the pyrolite composition may also exhibit regional variations.

[12] A long-standing problem in mineralogical modeling of the mantle is that the seismically observed gradient in the transition zone is much greater than inferred from mineral physics modeling assuming homogeneous mantle compositions [e.g., *Duffy and Anderson*, 1989; *Li et al.*, 2001]. The bulk sound velocity-depth gradient, dV_B/dz, for anhydrous wadsleyite is 0.0019(6) s^{-1} compared with values of 0.0024–0.0033 s^{-1} in seismic models [e.g., *Kennett et al.*, 1995; *Gaherty et al.*, 1996]. The difference of the velocity gradient between some seismic models and mineral physics modeling is within the uncertainties of mineralogical calculations, e.g., AK135 [*Kennett et al.*, 1995]. But some seismic models reported very large velocity gradients [e.g., *Gaherty et al.*, 1996] which need to be explained by other factors. One recent study [*Cammarano and Romanowicz*, 2007] proposed that an increase in basaltic fraction with

Table 1. Olivine Fraction of Mantle From Bulk Sound Velocity Contrast at 410-km Depth[a]

H_2O Content in Wadsleyite (wt %)	Seismic Model: PA5	Seismic Model: AK135
0	47(7)%	37(5)%
0.2	51(8)%	40(6)%
0.4	56(9)%	43(7)%
0.6	61(11)%	47(9)%
0.8	67(13)%	52(10)%
1.0	74(17)%	58(14)%

[a]Olivine fraction ≈ $\Delta V_{Bol\text{-}wa}/\Delta V_{Bseis} \times 100\%$. $\Delta V_{Bol\text{-}wa}$ is the velocity contrast between hydrous wadsleyite and anhydrous olivine. ΔV_{Bseis} is the velocity contrast at 410-km from seismic model.

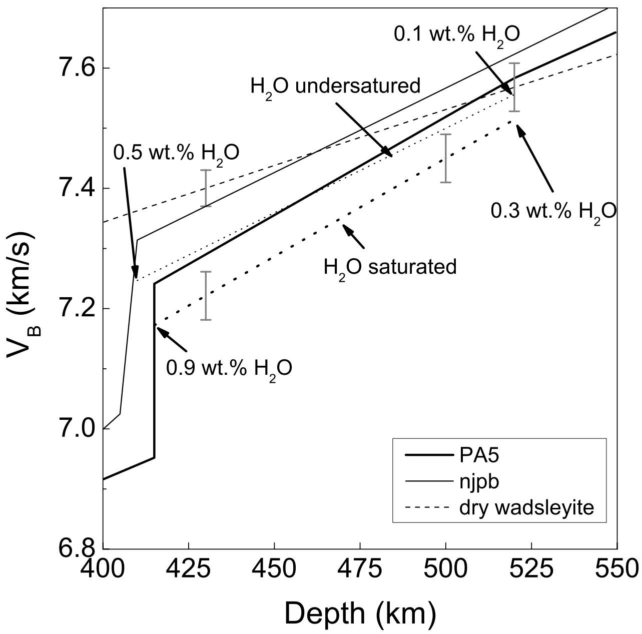

Figure 3. Effect of changes in H_2O content on seismic gradient in the upper part of the transition zone. Seismic model PA5 and njpb are shown as solid lines. Dashed line: anhydrous wadsleyite; bold dotted line: H_2O gradient under water saturated conditions; light dotted line: H_2O decreases from 0.5 wt.% to 0.1 wt.%. Uncertainties of the calculation are also shown.

depth could explain the seismic gradient at 250–350 km depth. Here we explore the role of depth variations of H_2O content on the seismic gradient between 410- and 520-km depth.

[13] Figure 3 shows bulk sound velocities with depth in wadsleyite (dashed line) compared with selected regional seismic models (njpb [*Kennett et al.*, 1994] and PA5 [*Gaherty et al.*, 1996]). The H_2O content in wadsleyite under water-saturated conditions is expected to decrease along the P-T conditions of a mantle adiabat from ~0.9 wt.% at 410-km depth to ~0.3 wt.% H_2O at 520-km depth because of the strong temperature and pressure dependence of hydrogen solubility [*Litasov and Ohtani*, 2003; *Demouchy et al.*, 2005]. This variation in water content for wadsleyite produces a velocity gradient comparable to seismic model PA5 [*Gaherty et al.*, 1996] (Figure 3). Here we have assumed a linear variation in water content over these depths consistent with available experimental data [*Litasov and Ohtani*, 2003; *Demouchy et al.*, 2005]. Even if mantle H_2O contents are below saturation, gradients in H_2O content may be produced if, for example, water is injected into the transition zone from slab dehydration reactions at different depths. Figure 3 also shows the seismic gradient results from an assumed linear variation in H_2O content from 0.5 wt.% at the top of the transition zone to 0.1 wt.% at 520 km. The resulting gradient is shallower in this case, but comparable to that of seismic model njpb. The calculation was also carried out for some other global and regional seismic models (Table S4). Because other models have a steeper velocity gradient than global model AK135, a greater variation in water content from 410 to 520 km is needed to match the seismic gradients. Also, the contribution of other mantle phases to the velocity gradient also needs to be accounted for.

5. Conclusion

[14] The elastic properties of hydrous wadsleyite are needed for inferring the potential effects of mantle hydrogen on seismic properties. Our experimental results and modeling indicate that hydrogen-rich (~1 wt.% H_2O) mantle wadsleyite could reconcile a pyrolite (60 vol.% olivine) mineralogy to match seismic bulk sound velocity models at 410-km depth. The gradient in H_2O content expected for such a water-saturated transition zone can also provide an explanation for the steep transition zone seismic gradient. On the other hand, if the mantle H_2O content is much lower ($\leq$0.1–0.3 wt.%), the presence of H_2O will have only a small effect on the seismic velocity discontinuity at 410-km depth, and the mantle olivine content consistent with bulk sound velocity data is approximately 45(8)%. In this case, large regional variations in bulk sound velocity contrast at 410 km cannot be explained by variations in H_2O content. However, gradients in H_2O content with depth could also partially explain the transition zone seismic gradient in this case. The presence of H_2O may also promote partial melting in or above the transition zone. The effects of partial melting and the amount of H_2O needed to satisfy compressional and shear wave profiles for the mantle will be addressed in future studies.

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