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High-pressure single-crystal elasticity of wadsleyite and the seismic signature of water in the shallow transition zone

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ABSTRACT

Earth's transition zone at depths between 410 km and 660 km plays a key role in Earth's deep water cycle since large amounts of hydrogen can be stored in the nominally anhydrous minerals wadsleyite and ringwoodite, $(Mg,Fe)_2SiO_4$. Previous mineral physics experiments on iron-free wadsleyite proposed low seismic velocities as an indicative feature for hydration in the transition zone. Here we report simultaneous sound wave velocity and density measurements on iron-bearing wadsleyite single crystals with 0.24 wt-% H_2O. By comparison with earlier studies, we show that pressure suppresses the velocity reduction caused by higher degrees of hydration in iron-bearing wadsleyite, ultimately leading to a velocity cross-over for both P-waves and S-waves. Modeling based on our experimental results shows that wave speed variations within the transition zone as well as velocity jumps at the 410-km seismic discontinuity, both of which have been used in previous work to detect mantle hydration, are poor water sensors. Instead, the impedance contrast across the 410-km seismic discontinuity that is reduced in the presence of water can serve as a more robust indicator for hydrated parts of the transition zone.

1. Introduction

Water strongly affects physical and chemical properties of rocks in Earth's mantle including viscosity (Hirth and Kohlstedt, 1996; Fei et al., 2017) and melting temperatures (Hirschmann, 2006). Tracing the deep hydrogen or H_2O ("water") cycle through Earth's interior thus remains a key challenge in understanding global geodynamic processes and their surface expressions. Several studies have argued that Earth's transition zone, at depths between 410 km and 660 km, might act as a deep water reservoir that remains stable for geological timescales (Bercovici and Karato, 2003; Kuritani et al., 2011) and plays a key role in the dynamic and geochemical evolution of the entire mantle (Bercovici and Karato, 2003). Aqueous fluids released by dehydration reactions may cause melting atop (Song et al., 2004) and below the transition zone (Schmandt et al., 2014) as well as trigger deep earthquakes (Richard et al., 2007). Furthermore, it has been proposed that a hydrous transition zone might be the source of continental flood basalts (Wang et al., 2015).

The hypothesis of the transition zone being a deep water reservoir is motivated by high-pressure high-temperature experiments showing that the nominally anhydrous minerals wadsleyite and ringwoodite, $(Mg,Fe)_2SiO_4$, that constitute up to 60 vol-% of transition zone rocks (Frost, 2008), can incorporate significant amounts of H_2O equivalents as point defects into their crystal structures (Inoue et al., 1995; Kohlstedt et al., 1996). The recent discovery of a hydrous ringwoodite inclusion in a natural diamond (Pearson et al., 2014) confirms the experimental predictions and strengthens the hypothesis of a (at least partly) hydrated transition zone. Further direct evidence for water in Earth's transition zone comes from ice-VII inclusions in diamonds that were interpreted to represent former aqueous fluids entrapped at pressures of the transition zone (Tschauner et al., 2018). However, the actual amount of water stored in the transition zone on a global scale cannot be constrained by these isolated observations.

Instead, global-scale three-dimensional mapping of the water content in the transition zone requires geophysical remote sens-

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ing. Electrical conductivity and seismic wave propagation are both sensitive to water incorporation and have thus been proposed as promising geophysical observables to quantify hydration (Huang et al., 2005; Smyth and Jacobsen, 2006; Yoshino et al., 2008; Mao et al., 2008a; Kelbert et al., 2009; Meier et al., 2009; Houser, 2016). Unfortunately, attempts to map hydration within the transition zone using electrical conductivity variations are hampered by contradicting experimental determinations on the quantitative effects of hydration on mineral electrical conductivity (Huang et al., 2005; Yoshino et al., 2008) as well as the limited spatial resolution in electrical induction measurements (Kelbert et al., 2009).

In contrast, seismological methods provide higher three-dimensional resolution and offer a variety of observables that have been proposed to be sensitive to hydration, including lateral wave speed variations as depicted by seismic tomography and the characteristics of seismic discontinuities (Chambers et al., 2005; Meier et al., 2009; Thio et al., 2016; Houser, 2016). Previous attempts to map the water distribution using seismic data found the transition zone to be strongly hydrated (Mao et al., 2008a), essentially dry (Houser, 2016), or partly hydrated away from subducting slabs (Meier et al., 2009), a conclusion that opposes the common assumption of water being carried into the mantle through subduction processes (Thompson, 1992; Ohtani et al., 2004). In addition to discrepancies arising from the use of different seismic data and their uncertainties, it remains difficult to unravel the competing effects of hydration, temperature, and possible changes in iron content on the seismic signature. Here we show that the disagreement among previous conclusions is in part caused by incomplete information on the expected effects of hydration on the seismic signal at conditions of the Earth's transition zone.

Earlier studies have shown that, at pressures close to the top of the transition zone (~14 GPa), hydration of Mg_2SiO_4 wadsleyite with 0.84 wt-% H_2O lowers P-wave and S-wave velocities by 2.7% and 3.6%, respectively (Zha et al., 1997; Mao et al., 2008a). Changing the Fe/(Mg + Fe) ratio from 0 to 0.08 has a similar effect (Zha et al., 1997; Wang et al., 2014). The combined effect of iron and hydrogen on seismic velocities, however, remains unclear at high pressure due to the lack of elasticity data for iron-bearing compositions with different hydrogen contents but equal Fe/(Mg + Fe) ratios (Mao et al., 2011). In particular, to separate the effects of iron and hydrogen, elasticity data on iron-bearing wadsleyite with an as small as possible but well characterized hydrogen content (<0.5 wt-% H_2O) is required. Since water fugacity strongly affects wadsleyite grain growth (Nishihara et al., 2006), a small hydrogen content in wadsleyite is unavoidable in order to synthesize single crystals of suitable size and quality (Kawazoe et al., 2015).

We performed simultaneous high-pressure Brillouin spectroscopy and X-ray diffraction experiments on wadsleyite single crystals with Fe/(Mg + Fe) = 0.112(2), an iron-content expected for mantle wadsleyite (Irifune and Isshiki, 1998; Frost, 2003a), and 0.24(2) wt-% H_2O up to a pressure of 19.7 GPa. From our data, the effect of water on the high-pressure sound wave velocities can be directly inferred by comparison to previous experimental results on wadsleyite with the same iron content, but 1.93 wt-% H_2O (Mao et al., 2011).

In contrast to previous studies on the high-pressure elasticity of supposedly anhydrous iron-bearing wadsleyite (Li and Liebermann, 2000; Liu et al., 2009; Wang et al., 2014), we determined the hydrogen concentrations of the wadsleyite crystals used in our high-pressure experiments. Any quantitative assessment of the effect of hydration on the elastic properties of wadsleyite requires a reliable determination of hydrogen concentrations. Since water enhances the crystal growth of wadsleyite (Nishihara et al., 2006), the determination of hydrogen concentration is especially important in single-crystal studies. The here-studied wadsleyite crystals complement existing elasticity data by providing the first iron-bearing composition with low but known hydrogen concentration. A comprehensive review of available equation of state parameters for different wadsleyite compositions has been reported in Buchen et al. (2017).

2. Experimental

2.1. Sample synthesis and characterization

Large single crystals of iron-bearing wadsleyite were synthesized from San Carlos olivine powder in a multi-anvil press as previously reported (Kawazoe et al., 2015). Electron microprobe analyses on four grains showed homogeneous compositions with Fe/(Mg + Fe) = 0.112(2). Hydrogen concentrations were determined by Fourier transform infrared (FTIR) spectroscopy on the same oriented single-crystal sections used for subsequent high-pressure experiments. Polarized infrared absorption spectra were recorded at 10 different spots distributed over 4 crystals. Absorption bands attributed to structurally bonded hydroxyl groups in wadsleyite (Jacobsen et al., 2005) were decomposed into individual peaks and the total integrated absorbances (Libowitzky and Rossman, 1996) converted to H_2O concentrations (Libowitzky and Rossman, 1997). The average of all four crystal sections yields 0.24(2) wt-% H_2O. Mössbauer spectroscopy on a powder sample prepared from the same experimental run (H4015) as the crystals gave $Fe^{3+}/\Sigma Fe = 0.15(3)$. The fraction of hydroxyl groups that combine with Fe^{3+} to substitute for tetrahedral Si^{4+} can be estimated from the deconvolution of FTIR spectra (Kawazoe et al., 2016) and is minor in our samples. More details on sample characterization can be found in Buchen et al. (2017). All chemical information was combined to calculate the molar mass and densities of the wadsleyite crystals.

2.2. High-pressure experiments

Oriented, double-sided polished single-crystal thin sections were prepared parallel to either the (120) or the (243) crystallographic planes with final thicknesses of 19 μm for experiments below 17 GPa and 10 μm for higher pressures. Semicircular disks were cut out of these thin sections using a focused ion beam (Marquardt and Marquardt, 2012). Two disks of complementary orientations were loaded together into a BX90 (Kantor et al., 2012) diamond anvil cell (DAC) along with a ruby sphere for provisional pressure determination (Mao et al., 1986; Dewaele et al., 2008). Precompressed neon was loaded as pressure-transmitting medium (Kurnosov et al., 2008). The two-crystal approach (Fig. 1a) facilitates sound wave velocity measurements in sufficient crystallographic directions to obtain all nine orthorhombic single-crystal elastic constants at consistent stress conditions while, at the same time, reducing the correlations between individual constants (Mao et al., 2015; Schulze et al., 2017; Kurnosov et al., 2017).

2.3. Brillouin spectroscopy and X-ray diffraction

At 8 pressures up to 19.7 GPa, sound wave velocities were determined by Brillouin spectroscopy (Speziale et al., 2014) for at least 13 different wave vectors within the plane of each platelet. Individual wave vectors were typically spaced by a 15° rotation around the DAC compression axis. Brillouin spectra were recorded at DESY Hamburg in forward symmetric scattering geometry with a scattering angle close to 50°. The actual scattering angle was calibrated with an oriented MgO single crystal using known elastic constants (Spetzler, 1970). The scattered light was analyzed for frequency shifts with a six-pass tandem Fabry-Perot interferometer. A Brillouin spectrum recorded at 19.7 GPa is shown in Fig. 1b. At each pressure, single-crystal X-ray diffraction experiments were