



Hydration-reduced lattice thermal conductivity of olivine in Earth's upper mantle

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Earth's water cycle enables the incorporation of water (hydration) in mantle minerals that can influence the physical properties of the mantle. Lattice thermal conductivity of mantle minerals is critical for controlling the temperature profile and dynamics of the mantle and subducting slabs. However, the effect of hydration on lattice thermal conductivity remains poorly understood and has often been assumed to be negligible. Here we have precisely measured the lattice thermal conductivity of hydrous San Carlos olivine ($Mg_{0.9}Fe_{0.1}SiO_4$ (Fo90) up to 15 gigapascals using an ultrafast optical pump–probe technique. The thermal conductivity of hydrous Fo90 with ~7,000 wt ppm water is significantly suppressed at pressures above ~5 gigapascals, and is approximately 2 times smaller than the nominally anhydrous Fo90 at mantle transition zone pressures, demonstrating the critical influence of hydration on the lattice thermal conductivity of olivine in this region. Modeling the thermal structure of a subducting slab with our results shows that the hydration-reduced thermal conductivity in hydrated oceanic crust further decreases the temperature at the cold, dry center of the subducting slab. Therefore, the olivine–wadsleyite transformation rate in the slab with hydrated oceanic crust is much slower than that with dry oceanic crust after the slab sinks into the transition zone, extending the metastable olivine to a greater depth. The hydration-reduced thermal conductivity could enable hydrous minerals to survive in deeper mantle and enhance water transportation to the transition zone.

hydration | thermal conductivity | geodynamics | metastable olivine | subducting slab

H₂O plays a critical role in driving and affecting many geophysical phenomena and dynamic processes in Earth's interior (1–3). It has been suggested that olivine, a primary mineral in the upper mantle, and its high-pressure polymorphs (wadsleyite and ringwoodite) could store a large amount of water (hydrogen ions) in their crystalline defects and act as a major water reservoir within Earth (2–4). Incorporation of water in these mantle minerals has been shown to influence their physical properties (5–12) and the dynamics of the mantle and subducting slabs (13). In particular, water enrichment could influence the minerals' thermal transport properties, which, in turn, alters the temperature gradient in the mantle and subducting slabs. At the center of subducting slabs, the cold temperature inhibits the olivine–wadsleyite/ringwoodite phase transformation (14). A thin wedge of olivine could therefore persist in a metastable state far below the 410-km depth. Previous experiments on transformation kinetics have shown that the presence of water inside the slabs greatly enhances the olivine–wadsleyite/ringwoodite transformation rate under the same pressure and temperature conditions (15–18). However, the hydration effect on the lattice thermal conductivity of mantle minerals and its consequential influence on the temperature inside the slabs that also controls the transformation rate remain largely unknown. Precise determination of the thermal conductivity of hydrous upper mantle minerals at relevant conditions would provide critical insight into the temperature evolution and dynamics of subducting slabs, and therefore the fate of metastable olivine in the mantle

transition zone, with potential implications for the origin of deep earthquakes (19).

In the past few decades, the lattice and radiative thermal conductivities of upper mantle minerals have been extensively studied, as these two mechanisms transfer heat through Earth's upper mantle in different ways. Previous optical absorption measurements have indicated that, under relevant pressure–temperature (*P-T*) conditions in the upper mantle, small amounts of incorporated water may strongly influence the radiative thermal conductivities of olivine (20), wadsleyite (21), and ringwoodite (21). By contrast, precise measurements of the lattice thermal conductivity of upper mantle minerals under extreme *P-T* conditions are very challenging. Therefore, little experimental data are available and the measurement accuracy is insufficient due to the limitations of previous experimental methods. Although the lattice thermal conductivity and diffusivity of nominally anhydrous olivine have been investigated for decades (22–25), the reported results at ambient and high pressures remain inconclusive among different studies. Moreover, in contrast to radiative thermal conductivity, which is sensitive to the hydration content, the role that hydration plays in the lattice thermal conductivity of olivine has often been assumed to be negligible (20, 25) due to its limited water storage capacity, typically of the order of ~0.1 wt% to 1 wt%. The validity of this hypothesis, however, has not been experimentally tested, and whether the influence of hydration changes as pressure increases remains unknown. Our study of the hydration effect on the olivine lattice thermal conductivity under relevant mantle conditions, in combination with our joint thermomechanical modeling of a subducting slab, provides insight into the extent of metastable olivine in the mantle transition zone.

Significance

Thermal conductivity of mantle minerals is critical for controlling the temperature profile and dynamics of the mantle and subducting slabs. However, the effect of hydration on lattice thermal conductivity remains poorly understood. We studied lattice thermal conductivity of olivine ($Mg_{0.9}Fe_{0.1}SiO_4$ (Fo90) to 15 GPa using ultrafast optics. The thermal conductivity of hydrous Fo90 with ~7,000 wt ppm water is 2 times smaller than its anhydrous counterpart at transition zone pressures. Modeling thermal structure of a subducting slab shows that the hydration-reduced thermal conductivity in the oceanic crust further decreases the temperature within the subducting slab, which substantially lowers the olivine–wadsleyite transformation rate and extends the metastable olivine to greater depths. Such an effect could enhance water transportation to the transition zone.

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Results and Discussion

We combined ultrafast time-domain thermoreflectance (TDTR) with high-pressure diamond anvil cell (DAC) techniques to precisely measure the lattice thermal conductivity (Λ) of anhydrous (polycrystalline) and hydrous (single crystal) San Carlos olivine ($\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_4$ (Fo90) at room temperature and high pressures up to ~ 15 GPa. The results are shown in Fig. 1. Given the anisotropic thermal conductivity along different crystal orientations (23), the pressure-dependent thermal conductivity of our anhydrous polycrystalline Fo90 (Λ_{anhy} ; black symbols in Fig. 1) shows a slight variation comparable to the measurement uncertainty of about 10 to 15% (see [Supporting Information](#) for the details). At ambient pressure, $\Lambda_{\text{anhy}} \approx 3.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, in reasonable agreement with previous measurements of polycrystalline Fo90 (22) and slightly lower than single-crystal Fo92 (25) and Fo93 (23), suggesting that, due to the iron mass disorder effect (cf. ref. 26), the more iron ions in the olivine, the stronger the phonon-defect scattering, leading to a lower lattice thermal conductivity. Upon compression, Λ_{anhy} increases rapidly with increasing pressure to $\sim 11.6 (\pm 2.3) \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 13.1 GPa, a pressure near the 410-km discontinuity.

Compared with the anhydrous Fo90, the lattice thermal conductivity of the hydrous single crystal Fo90 (Λ_{hy}) shows a weaker pressure dependence (see the red symbols in Fig. 1). Note that a different set of red symbols represents the measurement along a different orientation of the single-crystal sample. As expected, the incorporation of $\sim 7,000$ wt ppm (0.7 wt%) water in Fo90 does not significantly change the lattice thermal conductivity at ambient pressure or even below $P \approx 5$ GPa (the characterization of water content is described in [Calibration of Water Content](#) and [Figs. S2](#) and [S3](#)). However, the Λ_{hy} remains low upon further compression and reaches only $\sim 5.7 (\pm 0.6) \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 13.6 GPa, a factor of

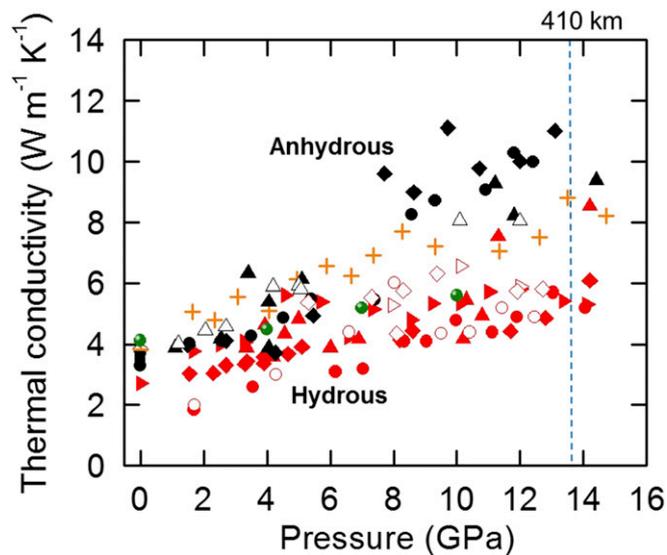


Fig. 1. Lattice thermal conductivity of anhydrous (black symbols) and hydrous (red symbols) San Carlos olivine Fo90 as a function of pressure at room temperature. Each set of symbols represents an individual measurement run, with solid symbols for compression and open symbols for decompression cycles. The effects of hydration and pressure on the thermal conductivity of Fo90 incorporated with 7,000 wt ppm water are insignificant until ~ 5 GPa, after which its increase with pressure is suppressed and becomes much smaller than its anhydrous counterpart by a factor of 2 at the pressures near the mantle transition zone, which is marked by the blue dashed line. The orange cross symbols represent the conductivity of Fo90 with a water content lower than 200 wt ppm, which is similar to the anhydrous Fo90 until ~ 10 GPa. The measurement uncertainties are $\sim 10\%$ in the low-pressure regime and $\sim 15\%$ at 15 GPa. Literature results for polycrystalline anhydrous Fo90 (green symbols) at 298 K from ref. 22 are plotted for comparison.

2 smaller than the Λ_{anhy} at a similar pressure. The concentrated distribution of thermal conductivity as a function of pressure based on four sets of measurements along different orientations suggests that the anisotropy of the hydrous Fo90 thermal conductivity may have been suppressed by the hydration effect under high pressure. The orange cross symbols in Fig. 1 represent the Λ_{hy} of Fo90 incorporated with a water content lower than 200 wt ppm (see [Calibration of Water Content](#) and [Fig. S2](#)), which is essentially the same as the Λ_{anhy} in the low-pressure regime but becomes slightly smaller by ~ 20 to 25% at $P > 10$ GPa. Note that, because the variation in the thermal conductivity during 8 GPa to 11 GPa is within our measurement uncertainty of about 15% , there isn't a thermal conductivity anomaly within such a pressure range. Compared with the effect of proton weakening on the sound velocities and thermodynamic properties (e.g., heat capacity) of olivine and its high-pressure polymorphs [typically of the order of only a few percent (5, 11, 12)], the proton-induced lattice thermal conductivity reduction at high pressures, by a factor as large as 2, is dramatic and much more significant than previously expected. Such a large pressure-induced suppression of hydrous Fo90 lattice thermal conductivity is likely due to higher-density ionic defects when compressed to higher pressures. Such weaker pressure dependence of thermal conductivity was also reported in MgO doped with ionic defects of iron at high pressures (26). At low-pressure regime, the replacement of cations (Si^{4+} , Mg^{2+} , and Fe^{2+}) in Fo90 with a small amount of H^+ does not substantially affect the heat capacity, phonon group velocities, and phonon scattering rates. As pressure increases, these ionic defects may aggregate and form a higher-density network that serves as an additional phonon-defect scattering mechanism to substantially reduce the phonon mean free path for thermal energy transport.

In Fig. 2, we plot the ratio of our lattice thermal conductivity of hydrous Fo90 against its anhydrous counterpart ($\Lambda_{\text{hy}}/\Lambda_{\text{anhy}}$) as a function of pressure to characterize the influences of hydration and pressure on thermal conductivity. The decrease of this ratio with pressure indicates an enhancement of the hydration effect with increasing pressure, which could be general for other hydrous mantle minerals.

In addition to the lattice thermal conductivity, which increases with pressure but decreases with temperature, the radiative thermal conductivity (Λ_{rad}) of olivine is important as well for heat transfer in Earth's upper mantle, particularly at high-temperature conditions. Hofmeister (20) showed that the Λ_{rad} of Fo90 is sensitive to the hydration content and grain size of a crystal. At 1,500 K, the Λ_{rad} of subcentimeter-sized Fo90 incorporated with 10 wt ppm to 100 wt ppm water is smaller than its anhydrous counterpart and the lattice thermal conductivity by a factor of ~ 4 and ~ 8 , respectively, if the hydration effect on the lattice thermal conductivity was assumed to be negligible (cf. figures 5 and 6 in ref. 20). If we assume that the temperature dependence of hydrous Fo90 lattice thermal conductivity (Λ_{hy}), which is incorporated with proton defects follows a typically assumed $T^{-1/2}$ dependence (see our high-temperature measurements at ambient pressure in [High-Temperature Thermal Conductivity Measurements](#), [Fig. S6](#), and [Table S1](#) as well as refs. 22 and 26), the Λ_{hy} of Fo90 with $\sim 7,000$ wt ppm water would then be reduced to $\sim 2.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 13.6 GPa and 1,500 K. This estimation suggests that the lattice thermal conductivity of hydrous olivine at relevant P - T conditions near the mantle transition zone remains significantly larger than the radiative component, indicating that thermal conduction is a major contributor to heat transfer in the hydrous mantle, compared with the thermal radiation mechanism. Therefore, it is reasonable to use the lattice thermal conductivity to evaluate the effect of hydration on the thermal profile of subducting metastable olivine slabs in Earth's transition zone.

We further investigated the resultant temperature anomaly of subducting slabs using 2D thermomechanical models (27) (see [Supporting Information](#) for the modeling details). The degree of hydration below the oceanic Moho before subduction remains

necessarily linked to a completely dry condition in all parts of the slab. In addition, the hydrated slab will slow down the heat dissipation during frictional sliding of earthquakes, and therefore promotes transformational deep earthquakes.

Although the water content in the entire slab had been suggested to vary widely, depending on the slab's age and spreading rate (cf. ref. 31), our data for the thermal conductivity of hydrous Fo90 with <200 wt ppm H₂O (orange cross symbols in Fig. 1) already showed an about 25% lower value than the anhydrous Fo90 at about 14 GPa. We therefore can expect that, even if the hydrous olivine in the oceanic crust contains water content of only a few hundred to 1,000 wt ppm, it will still play an effective role in the thermal insulating effect. In other words, the possible low water content in olivine in the oceanic crust still lowers the temperature at the center of the subducting slab and slows down the olivine–wadsleyite/ringwoodite transformation rate around the 410-km depth.

Conclusions

The hydration-induced significant reduction of lattice thermal conductivity at higher pressures could be a general feature in other mantle minerals, such as garnet, wadsleyite, and ringwoodite, which can contain comparable or even larger amounts of hydration than

olivine (5). Therefore, the effective lattice thermal conductivities of the hydrous subducted slab, upper mantle, and transition zone aggregates are likely much smaller than previously expected. The stability of hydrous minerals is strongly affected by temperature, and lower thermal conductivity can help hydrous minerals in subducted slabs survive to greater depths, enhancing water transportation to the transition zone, which will lower the thermal conductivity further. There is potentially positive feedback between the water concentration and reduced thermal conductivity in the transition zone. Future experimental studies of the hydration effect on the lattice thermal conductivity of other mantle minerals at relevant *P-T* conditions is essential to further constrain the heat transfer and to model the dynamics in Earth's upper mantle and transition zone.

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