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Electrical conductivity of hydrous silicate melts: Implications for the bottom-up hydration of Earth's upper mantle

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Abstract:

The upwelling of the hydrous mantle transition zone triggers dehydration-induced partial melting atop the 410-km discontinuity. Here we investigate the electrical conductivity of hydrous silicate melts in the 200-400 km depth range and explore whether melting at the 410-km depths is responsible for the hydration of the upper mantle. Our experimental electrical conductivity data demonstrate that the mantle at 180-350 km depths is mostly melt free, confirming the H₂O under-saturated conditions. However, the residual mantle from partial melting atop the 410km discontinuity may contain various possible amounts of water according to the initial mantle transition zone and melt concentrations. This residual H₂O could contribute to the hydration of the upper mantle either through diffusion or material replacement by upwelling. Our calculations suggest that the diffusion may not be responsible for the hydration of the upper mantle to present H₂O concentration of 50–200 ppm wt. Melting of the upwelling mantle transition zone with less than 1500 ppm wt. H₂O produces residual peridotites with ~ 200 ppm H₂O at the 410-km discontinuity. Continuous upwelling of such hydrous residues would gradually replace the dry upper mantle with depleted residual hydrous peridotites in less than 260 Ma. In this study, we propose a bottom-up hydration mechanism for the Earth's upper mantle driven by dehydration-melting at the 410-km

discontinuity. The hydrous partial melting at the top of the asthenosphere appears to be a consequence of H₂O saturation in the upwelling residual peridotites.

Keywords: electrical conductivity, water storage capacity, hydrous melt, upper mantle, mantle upwelling, mantle transition zone

1. Introduction:

The average H₂O content of the Earth's upper mantle is determined to be about 50-200 ppm wt. (Hirschmann, 2006; Michael, 1988; Saal et al., 2002; Simons et al., 2002). These estimates are significantly lower than the H₂O storage capacity of peridotite mineral assemblages (Ardia et al., 2012; Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012), indicating water undersaturated conditions for the Earth's upper mantle. The origin of H₂O in the upper mantle is still a subject of debate. However, two possible scenarios have been proposed. In the first hypothesis, the H₂O in the upper mantle has been attributed to hydrous residues left behind following partial melting in the upper mantle. In the second scenario, H₂O in the upper mantle can be derived from the mixing of dry upper mantle with a hydrous primitive source (Hirschmann, 2006).

At present, three distinct partial melting regimes, that could explain the origin of H₂O in the upper mantle have been proposed; dehydration melting at the 410-km discontinuity (Bercovici and Karato, 2003), melting in mantle wedges (Hirth and Kohlstedt, 1995) and partial melting of mantle plumes (Morgan and Morgan, 1999), all arguing that the residues left behind by the melting events could be responsible for H₂O contents in the upper mantle. While the depleted signature of mid oceanic ridge basalt (MORB)-source upper mantle strongly favors the partial melting hypotheses, the exact nature of the melting regime that led

to the hydration of the upper mantle, consistent with geochemical, geophysical and geodynamic observations, remains poorly constrained (Hirschmann, 2006).

A recent study based on *in situ* sound velocity measurements confirmed the dehydration induced melting in the upwelling mantle across the 410-km discontinuity (Freitas et al., 2017). Geophysical observations of high electrical conductivity (up to 1 S/m) (Toffelmier and Tyburczy, 2007) and reduced seismic wave velocities (δV_s 4-9%) (Revenaugh and Sipkin, 1994; Tauzin et al., 2010) suggest a 50-60 km thick melt layer above the Mantle Transition Zone (MTZ) (Revenaugh and Sipkin, 1994; Song et al., 2004; Tauzin et al., 2013, 2010; Vinnik and Farra, 2007). The upwelling of hydrous MTZ with ~ 0.1 - 0.2 wt.% of H_2O (Freitas et al., 2017; Huang et al., 2005; Karato, 2011) and the significant differences in H_2O storage capacities between the MTZ (Bolfan-Casanova et al., 2000; Hirschmann et al., 2005; Kohlstedt et al., 1996) and the overlying mantle (Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012) is the governing factor that triggers melting in the upwelling mantle across the MTZ (Bercovici and Karato, 2003; Freitas et al., 2017). The reported 3.6-9.4 % shear wave velocity drop is compatible with a 0.5-2.5 % melt fraction (Freitas et al., 2017) in the partial melt layer.

The melt produced at the 410-km discontinuity is found to have high H_2O contents (Freitas et al., 2017; Matsukage et al., 2005). The mineral-melt equilibrium following melting at the top of the MTZ causes H_2O to diffuse from the hydrous melt into solid mineral phases both above and below the melt layer. The H_2O diffusion may occur between melt and wadsleyite, transferring H_2O back into the underlying MTZ and between melt-olivine/pyroxene/garnet, hydrating the overlying mantle. In addition to the diffusion process, the mantle upwells passively away from subduction zones in response to intrusion of slab material into Earth's interior (Bercovici and Karato, 2003; Karato et al., 2006). The mantle upwelling is significantly faster than H_2O diffusion into mantle minerals (Bercovici and

Karato, 2003), replacing the dry upper mantle with residual peridotites. The dehydration melting in the upwelling mantle and subsequent H₂O transport mechanisms therefore play a crucial role in water transfer between the upper mantle and the mantle transition zone (Bercovici and Karato, 2003).

Laboratory measurements of electrical conductivity have been extremely useful in characterizing the material circulation in the Earth's mantle (Dai and Karato, 2009, Yoshino, 2010). The laboratory measurements suggest electrical conductivity is a powerful tool for detecting melt or fluid and their occurrence within the solid mineral matrix at deep mantle conditions (Freitas et al., 2019; Grotenhuis et al., 2005; Manthilake et al., 2016, 2015; Maumus et al., 2005). It is also sensitive to the chemical composition of melt, particularly to volatiles (Gaillard et al., 2008; Karato, 1990; Manthilake et al., 2009; Yoshino, 2010) and alkalis (Ni et al., 2011). In this study, we measure the electrical conductivity of hydrous peridotite at various pressures up to 12 GPa above their melting temperatures. The results were compared with magnetotelluric profiles to gain insight into the extent of melting and melt percolation if hydrous conditions are assumed for the Earth's upper mantle. Based on our results, we discuss different scenarios that could be responsible for the H₂O circulation in the upper mantle.

2. Materials and Methods

2.1 Starting materials

Starting materials with a composition similar to KLB-1 peridotite (Wang and Takahashi, 2000) were prepared using reagent grade oxides, initially mixed in the absence of the required amount of Al(OH)₃ to yield the correct mineral composition (ranging from 1.63 to 4.86 mg to obtain 500 mg of peridotite powder with 400 to 1500 ppm H₂O, respectively). These powders were dried at 300 °C overnight to remove any adsorbed moisture. Na and K

were added as NaCO_3 and K_2CO_3 and then de-carbonated. The decarbonation of the oxide mixtures was carried out by slowly increasing the temperature to 1000°C ($1.6^\circ\text{C}/\text{min}$) and keeping it at 1000°C for about 10 hours to ensure complete decarbonation. The decarbonated powder mixture was then cooled to 200°C and stored in a high vacuum furnace at 120°C prior to the hot pressing runs. The required amount of $\text{Al}(\text{OH})_3$ was then mixed with the decarbonated powder mixture to obtain the desired amount of H_2O in peridotite samples.

Samples with $\sim 0, 500, 700$ and 1200 ppm wt. H_2O as initial content in powders were prepared in order to have hydrous under saturated conditions at the different targeted pressures. As water solubility increases with pressure in nominally hydrous phases (Ardia et al., 2012; Férot and Bolfan-Casanova, 2012), we always used initial water content under solubility limit at each pressure. However, due to the absorbed moisture in the high pressure synthesis assembly, the water content may be subjected to some possible water (H) diffusion into the samples even with sealed capsules. The X-Rays powder diffraction analyses using a Philips PW 1830 (Cobalt wave-length) and micro-Raman analyses were carried out prior to the electrical conductivity measurements to ensure the absence of additional hydrous phases such as super hydrous Phase-B and partial melts in pre-sintered samples.

2.2 Experimental

The resulting powder mixtures were hot pressed to obtain solid sintered samples for electrical conductivity measurements. Hydrous and dry samples were synthesized at different pressures and temperature conditions in a first high pressure run with multi-anvil apparatus, these samples were recovered for *in situ* measurements that were performed in a second multi-anvil run. The details on samples conditions (pressure, temperature and time at temperature) are given in the table S1. Dry samples were synthesized at target or lower pressures to ensure low water solubility of the material (Ferot and Bolfan 2012, Kohlstedt et

al. 1996). Hydrous samples were synthesized at target pressure (for 6 and 9 GPa) and 14 GPa. The synthesis of hydrous samples at 14 GPa performing *in situ* experiments at 12 GPa allows us to simulate the dehydration induced melting at the 410-km discontinuity (Freitas et al. 2017).

Dry samples and hydrous samples were synthesized for ~2 hours in Gold-Palladium (Au-Pd) capsules and temperatures between 1373 and 1573 K (table S1). These undersaturated conditions are required during the synthesis process, in order to avoid hydrous silicate melts along grain boundaries, which could otherwise interfere with the electrical conductivity. Cylindrical core samples ~1.0 mm in length and ~1.2 mm in diameter were prepared from these pre-synthesized sample specimens.

The high-pressure and high-temperature experiments were performed using a 1500-ton Kawai-type multi-anvil apparatus at the Laboratoire Magmas et Volcans, Clermont-Ferrand, France. Both sample synthesis and the *in situ* measurements were performed in multi-anvil apparatus using an octahedral pressure medium composed of MgO and Cr₂O₃ in a 14/8 multi-anvil configuration (octahedron edge length / anvil truncation edge length) for experiments at 12 GPa and 18/11 for experiments at 6 and 9 GPa (Supplementary Figure S1). The pre-synthesized cylindrical sample was inserted into a MgO sleeve. The sleeve also helps insulate the sample electrically from the furnace. This furnace, composed of a 25 μ m thick cylindrical Re foil, has apertures for the electrode and the thermocouple wires. A zirconia sleeve around the furnace was used as a thermal insulator. Thermal gradient was also taken into account for the design of these experiments (Hernlund et al., 2006), and were estimated to be negligible <5° with our sample dimensions (Supplementary Figure S2). We placed two electrodes, made of Mo discs, at the top and bottom of the cylindrical sample. A tungsten-rhenium (W₉₅Re₅-W₇₄Re₂₆) thermocouple junction was placed at one end of the sample to monitor the temperature. The opposite end was connected to a single W₉₅Re₅ wire. We

collected impedance spectra between the two W₉₅Re₅ wires. All ceramic parts of the cell assembly, including the pressure medium, were fired at 1373 K prior to their assemblage in order to remove any adsorbed moisture, and kept in vacuum furnaces (10⁻² Torr and 150°C) before assembling the experiment. Oxygen fugacity of the sample was not controlled during *in situ* measurements, but was expected to remain below the Mo-Mo₂ buffer.

EC measurements were performed using the ModuLab MTS Impedance/Gain-phase analyzer in the frequency range of 10¹-10⁶ Hz. Polyphasic samples are characterized by a combination of resistor-capacitor/constant phase element (R-C/CPE) circuits and the resistance can be obtained by fitting the impedance spectra to appropriate equivalent circuits (Supplementary Figure S3). Once the sample resistance has been determined, conductivity can be calculated using the sample diameter and length. The experimental procedure for electrical conductivity measurements is discussed elsewhere (Manthilake et al., 2016, 2015). Importance of a moisture removal procedure is crucial in these experiments and is detailed in Supplementary Figure S3.

2.3 Chemical and micro-textural analyses

The chemical composition and micro-textures of pre-sintered samples and experimental run products after electrical conductivity measurements were investigated using a Cameca SX100 electron probe micro analyzer and a Scanning Electron Microscope (SEM) JEOL Jeol JSM-5910 LV, respectively, at the Laboratoire Magmas et Volcans of Clermont-Ferrand. Energy-dispersive X-ray spectroscopy (EDS) chemical mapping was used to determine the mineral proportions and chemical compositions by selecting a large area of each phase. The fine melt micro-textures were observed using a ZEISS supra 55VP field emission gun (FEG) SEM at 2MATech, Aubière, France. Major element compositions of solid phases are given in Supplementary Tables S2 and S3.

The presence of water (OH) in the crystalline phases of our peridotite samples was qualitatively analyzed both before and after electrical conductivity measurements. These analyzes were performed with micro-Raman spectroscopy (Freitas et al., 2017) (Supplementary Figures S4-S6). Raman spectra were collected using an InVia confocal Raman micro spectrometer, equipped with a 532 nm diode laser (output power of ~140 mW), a Peltier-cooled CCD detector, a motorized XY stage and a Leica DM 2500 M optical microscope, at the Laboratoire Magmas et Volcans, Clermont-Ferrand. The Raman spectroscopy technique was preferred over FTIR analysis mainly due to its capacity to analyze both OH and Si bonds in a non-destructive manner and its high spatial resolution allowing the analysis of small objects (~1 μm). However, due to the unavailability of H₂O calibration curves for pyroxene and garnet, we were not able to perform a complete estimate of the bulk H₂O contents of our samples (qualitative analysis only).

3. Results

The melting of hydrous peridotite can be detected by a sudden or discontinuous increase in electrical conductivity at high temperature (Fig. 1). The electrical conductivity of partially molten peridotites varies from 0.1-1 S/m. The activation enthalpies of hydrous peridotites before melting indicate the conduction mechanism is characteristic of electron hopping (small polaron conduction) (Katsura et al., 2009).

The high resolution image analyses of samples after electrical conductivity measurements indicate the presence of melt in hydrous samples (Fig. 2). The experimental run products indicate well developed polygonal textures with average grain size of <30 μm . The melt distribution appears to be homogeneous throughout the samples and occurs as thin films along grains boundaries and as small pockets of a few hundred nanometers in size at

triple junctions (Fig. 2 b, c, d). The melt forms an interconnected network showing a very low dihedral angle with complete wetting of the grain boundaries (Freitas et al., 2017) (Fig 3).

Due to the extremely small size of the melt pockets ($<1\ \mu\text{m}$), we were not able to measure the chemical composition of the resulting hydrous silicate melt at 6 and 9 GPa. The melt fraction appears to be low (up to a few %). The chemical analyses of melt at 12 GPa indicate significant enrichment in incompatibles elements (Ca, Al, Na, K, Fe, Ti) (Freitas et al., 2017).

Samples synthesized prior to electrical conductivity measurements were melt-free (confirmed by X-ray powder diffraction/micro-Raman) and their mineralogy was mainly composed of olivine, clinopyroxene and garnet (Fig. 2a). No evidence of hydrous phases such as super-hydrous B were found in the starting materials. The proportions of the different crystalline phases vary between the samples depending on the degree of melting. The starting samples recovered after synthesis at 12 GPa have mineral proportions of about $60\pm2\%$ olivine, $20\pm2\%$ garnet and $20\pm2\%$ clinopyroxene. Upon partial melting at 12 GPa, the proportion of olivine remained almost constant, while the proportion of Cpx decreased with increasing melt fractions. The proportion of garnet slightly increased with melting, as would be expected with mantle mineralogy (Kaminsky, 2012). For all experiments, the Mg number of olivine is between 90 and 93. Nominally anhydrous sample have lower Mg numbers of 88-89, which is consistent with melt-absent conditions. Chemistry of solid phases for synthesis and experimental runs are given in Supplementary Tables S2 and S3.

4. Discussion

4.1 Melting in the upper mantle

The magnetotelluric and geomagnetic depth sounding profiles of the upper mantle indicate significant regional variation in electrical conductivity (Grayver et al., 2017;

Kuvshinov et al., 2005; Toffelmier and Tyburczy, 2007). The majority of geophysical profiles are compatible with electrical conductivity resulting from hydrous upper mantle minerals (Novella et al., 2017b). The comparison of these electrical conductivities of hydrous silicate melts with global electromagnetic and magnetotelluric profiles of the upper mantle implies that the upper mantle in the depth range 180-350 km is mainly melt free (Fig. 4). However, the estimated high electrical conductivity anomaly above the 410-km discontinuity (Toffelmier and Tyburczy, 2007) appears to be compatible with the presence of melt in this zone (Fig. 4). Thanks to our measurements, the melt fraction required to explain this observed anomaly is estimated to be <10 vol. % (Fig. 4). This estimation is significantly higher than the melt fraction of 0.5-2.5 % obtained by sound velocity measurements (Freitas et al., 2017) to explain the shear velocity reduction (δV_s) of 3.6-9.4 %. The possible over-estimation of melt fraction by laboratory electrical conductivity methods has been discussed in a recent study (Freitas et al., 2019) in addition to assumptions made on forward and inversion models of electromagnetic data (Toffelmier and Tyburczy, 2007). In our discussion, we use our electrical conductivity data to qualitatively determine the vertical extents of melting in the upper mantle.

The absence of melt in the depth range 180-350 km confirms that the water content of the upper mantle is below the saturation limits of peridotite (Ardia et al., 2012; Férot and Bolfan-Casanova, 2012). This water undersaturated mid-upper mantle is sandwiched between two partial melt layers. Melting at the top of the asthenosphere (above 180 km) has been discussed in terms of the reduced water solubility of aluminous orthopyroxene (Mierdel et al., 2007), and melting below 350 km has been linked to the reduced water solubility in peridotites, in both cases melting is caused by the presence of excess H₂O. Here we investigate whether the dehydration-induced water saturation and subsequent melting at the

top of the mantle transition zone has an influence on water circulation in the Earth's upper mantle.

4.2 H₂O circulation in the upper mantle

The strong evidences supporting the presence of a melt layer atop the mantle transition zone and the strict requirement of H₂O for deep mantle melting underline the importance of understanding the mechanism that is responsible for H₂O circulation in the upper mantle. Assuming that the mantle was initially dry and well mixed after the establishment of the vigorously convecting global magma ocean, initiation of subduction in a plate tectonic context can be considered as having been the major process responsible for mantle hydration. With average downward slab velocities of between 5 and 10 cm per year (Gordon, 1995a) and assuming plates thicknesses of 7 km and a total ridge length of 70000 km, the annual amount of subducted material is estimated to be 24-49 km (Smyth and Jacobsen, 2006). With a mean age of 80 Ma, the average subducted material is estimated to have between 0.1 and 1.5% of H₂O (Dixon et al., 2002) leading to an H₂O flux of 0.1 to 1 km³ into the mantle per year. However, studies of ophiolites indicate that most hydrous minerals break down at shallow mantle conditions and subducted rocks in eclogite facies were shown to be significantly impoverished in volatiles, with values of up to 0.25% H₂O (Katayama and Nakashima, 2003). Even in the case of extreme slab dehydration of up to 97% (Dixon et al., 2002), H₂O contents of up to 1000 ppm wt. can be preserved in subducting slabs and transported down to the transition zone (Dixon et al., 2002).

Using slab downward speed of 5 cm per year and the MTZ thickness of 250 km, the time required for a portion of slab to cross the MTZ is estimated to be 5 Ma. While this time is relatively short on a planetary timescale, the slab deflection and stagnation in the MTZ observed in seismic tomography studies (Zhao, 2004) suggests that residence time of

subducted material would be much greater than the values assumed in our calculations. With a diffusion coefficient of 10^{-8} - 10^{-10} m²/s (Hae et al., 2006; Richard et al., 2006), a significant transfer of H₂O from the slab to the MTZ could be expected during that time.

The appearance of the melt layer above the MTZ can be inferred given the estimation of H₂O slab fluxes in the mantle transition zone. If we assume 24.5 km³ slab material reaches the MTZ per year (volume produced at the ridges for 5 cm/year spreading velocity along 70000 km of ridges with an average thickness of 7 km (Gordon, 1995b)), and that these rocks contain 1000 ppm wt. H₂O, a mass of 9.4×10^{10} kg of H₂O is transferred to the MTZ every year. Assuming the volume of the MTZ to be 6.5×10^7 km³, with a mass of 2.6×10^{20} kg (mean density of 4000 kg/m³), and assuming complete slab dehydration into the MTZ, only 2.2-3.3 Ma after the beginning of modern subduction is required to reach 800-1200 ppm wt. in the MTZ, which is roughly equal to the maximum H₂O solubility in nominally anhydrous minerals (NAM's) forming the upper mantle (Ardia et al., 2012; Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012). Using the upper limit of the inferred H₂O content of the mantle transition zone from geophysical methods of about 0.1-0.2 wt.% H₂O (Freitas et al., 2017; Huang et al., 2005), the time required to transport H₂O from surface to the MTZ is estimated to be 6 ± 0.6 Ma. This very simple estimation highlights the efficiency of the hydration of the MTZ. It indicates clearly that melting at the 410-km discontinuity would have been initiated early in the Earth's history. However, H₂O diffusion into wadsleyite is a slow process (Hae et al., 2006). For example, the time required to diffuse 2000 ppm wt. H₂O to reach 500 ppm wt. at 500 km laterally along the MTZ is estimated to be 175 Ga. Given that the surface area of the MTZ is significantly large and the number of active subduction zones is extremely limited, it is possible that the H₂O distribution in the MTZ remains heterogeneous, with regions of the MTZ maintaining dry conditions (Yoshino et al., 2008). The absence of seismic anomalies in some regions (Tauzin et al., 2010)

indicates that melting is not pervasive above the MTZ. The upwelling of H₂O deprived regions (< 800-1200 ppm wt.) of the MTZ may not undergo partial melting upon entering the upper mantle.

The H₂O under-saturated MTZ, as demonstrated by recent experimental studies (Freitas et al., 2017; Huang et al., 2005; Karato, 2011), can be a result of continuous removal of H₂O from the MTZ (upward and/or downward) or partial dehydration of slabs at the MTZ. The first scenario implies the presence of a global H₂O circulation cycle extracting H₂O from the MTZ from an early stage. However, the quantity of H₂O stored in the melt layer above the MTZ together with the moderately hydrous olivine present in equilibrium with melt over the 60 km thick layer, only represents a small portion of the total amount of H₂O in the MTZ (<1%). This amount is a few orders of magnitude lower than the total amount transported by subducted slabs per year. Similarly, the known deep volcanic sources of OIB show that source material may be only moderately hydrous with H₂O content estimated between 300 and 1000 ppm wt. (Rüpke et al., 2006; Simons et al., 2002). These calculations demonstrate the possibility of large scale melting events occurring periodically in the Earth's history. Large scale deep mantle melting has occurred in the past, as demonstrated by the presence of komatiites, kimberlites and numerous large igneous provinces (Vinnik and Farra, 2007). However, at present, such large scale volcanism does not exist anymore and hot spot volcanism and mantle plumes are alternative candidates for removing H₂O from the deep Earth.

On the other hand, it also appears possible that slabs that are not fully dehydrated at the MTZ, and instead carry a substantial amount of H₂O on down to the lower mantle (Dixon et al., 2002). The high H₂O contents of up to 2 wt. % observed in some enriched mantle sources of oceanic island basalts (OIB) indicate that some H₂O may be incorporated into dense hydrous silicate phases (Nishi et al., 2014; Pamato et al., 2014) that penetrates down

into the lower mantle via subduction processes (Rüpke et al., 2006). These would imply a slower hydration rate of the MTZ and more tardive apparition of the LVL.

4.3 Bottom-up hydration of the upper mantle

In this section, different possible mechanisms of bottom-up hydration of Earth's upper mantle and their consequences are explored.

Due to the H₂O under-saturated conditions of the upper mantle, the water contained in hydrous melts produced at the 410-km discontinuity may diffuse in to the overlying mantle. Based on the one dimensional diffusion equation (Crank, 1975), we calculate the diffusion time required to hydrate the upper mantle assuming that the upper mantle is initially dry (Supplementary Figure S7),

$$C(x, t) = C_0 + (C_1 - C_0) * \operatorname{erfc}\left(\frac{x}{2\sqrt{D * t}}\right)$$

where C_0 is the initial concentration, C_1 is constant local concentration, x is the distance to the border of concentration C_1 , t is the diffusion time and D the diffusion coefficient of hydrogen in olivine. The value for D was estimated to be $7.82 \times 10^{-9} \pm 6.1 \times 10^{-8}$ m²/s at 1723 K for a randomly oriented polycrystalline material (average of the three crystallographic direction) (Novella et al., 2017b). Using average H₂O solubility for mantle (Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012), increasing from 300 ppm wt. at 200 km up to 1000 ppm wt. at 410 km, we calculate the time required for peridotite in equilibrium with the hydrous melt layer to reach H₂O saturation. Our results indicate that the time required to reach the H₂O saturation in the uppermost mantle is unrealistically high based on diffusion alone, and cannot be achieved during the planetary life time (Supplementary Figure S7, Fig. 5a).

On the other hand, the mantle away from subducting slabs is rising passively in response to the material injection by subduction. Estimated values using slab flux yield to

upwelling velocities of 1 mm per year (Bercovici and Karato, 2003). The estimation based on convection models indicates convection rate as high as 1.5 cm/year (Becker et al., 1999). The upwelling rates from subduction fluxes indicates that only 260 Ma is sufficient to bring rocks from the top of the transition zone to the uppermost mantle (150 km depth) and even faster considering the highest convection rates. This estimation indicates that residual hydrous peridotite should be rising from the LVL to the uppermost mantle and saturates the upper mantle at various depths depending on the initial H₂O contents in peridotite (Fig. 5b).

Here we investigate the possible H₂O contents of peridotites in contact with the melt layer. The shear wave velocity reduction (δV_s) at the melt layer is estimated to vary between 3.6-9.4 % (Revenaugh and Sipkin, 1994; Tauzin et al., 2010). Based on our previous study (Freitas et al., 2017), the resulting melt fraction corresponding to the δV_s at the low velocity layer is estimated to be 0.5-2.5 vol.%

The average water content of the MTZ has been constrained by different methods with variable results. Despite potential high water solubilities of MTZ minerals (Bolfan-Casaonova et al., 2018b), the actual content of the MTZ is believed to be lower than saturation limit. Water contents inferred from inclusions in diamonds (Pearson et al., 2014) with up to 2 % would suggest potentially high water content which also supported by viscosity measurements (Fei et al. 2017). Particularly, the near-water saturated conditions implied by hydrous ringwoodite inclusions found in natural diamond (Pearson et al., 2014) may represent an episode of subduction-zone-related melting. However, most of geophysical techniques such as elasticity, sound velocities (Inoue et al., 1998, Chang et al., 2015, Freitas et al., 2017) and electrical conductivities (Huang et al., 2005, Yoshino et al., 2008, Yoshino and Katsura 2012) indicating major part of MTZ contains < 0.2 wt. % H₂O.

Assuming a batch melting scenario and H₂O partitioning between peridotite and melt (Novella et al., 2014), readjusted to the mineral proportions expected at 12 GPa

(Supplementary Text S1), we could estimate the H₂O contents of peridotite in equilibrium with hydrous melt for variable water contents in the mantle source for different melt fractions (Fig. 6). Different scenarios could occur given the potential range of water content of the residual peridotites (Fig. 6). Thus, if residual peridotite in the upwelling mantle contains about 200 ppm wt. of H₂O, mantle saturation can be expected at range of depths in the upper mantle (Fig. 6). If we assume the H₂O saturation curves of Férot and Bolfan-Casanova (Férot and Bolfan-Casanova, 2012), Hirschmann et al. (Hirschmann et al., 2009), Tenner et al. (Tenner et al., 2012, 2009) and Ardia et al. (Ardia et al., 2012) (for garnet/olivine partitioning of H₂O of 9.0), peridotite in the upper mantle reaches H₂O saturation limits above 200 km. In contrast, by using the extreme H₂O saturation scenario proposed by Ardia et al. (Ardia et al., 2012) with water partitioning between olivine and garnet of 0.9, the mantle saturation above 200 km depths can be expected for the water contents of 450 ppm. However, this value is inconstant with the upper mantle water contents of 50-200 ppm.

The ascent of residual hydrous peridotites would initiate melting upon reaching their storage capacities and could be responsible for the hydrous melting at the top of the asthenosphere. Melting above a depth of 180 km can only be explained by moderate water content in the MTZ (< 1500 ppm wt) for geologically relevant melt fractions at the 410-km discontinuity (Fig. 6). The upwelling of the MTZ with water contents of less than 800 ppm wt. may not undergo melting upon entering the upper mantle and may continue to hydrate the upper mantle. However, such fertile peridotites are not able to explain the depleted signature of the upper mantle. Melting at the 410- km discontinuity is therefore a primary requirement for a compatible hydration of the upper mantle with geochemical constrains.

The lithosphere-asthenosphere boundary (LAB) is defined as intersection of the conductive geotherm of mechanically strong crust with the fluid-saturated peridotite solidus in mechanically weak upper most part of the mantle (McKenzie and Bickle, 1988). The

location of the LAB is therefore corresponding to the depth at which the fluid saturation occurs in upwelling peridotites. The geochemical constraints suggest that the LAB is not a stable feature, which appear to move up or down over geological time scale (O'Reilly and Griffin, 2010). If other parameters remain constant, the lateral variation of the depth of the LAB may indicate the variable water contents in residual peridotites, which corresponds to the lateral variations of water contents in the MTZ. Further, the movement of the depth of the LAB at the same location may suggest episodic variation of water contents in the MTZ over geological time scale. The upwelling of MTZ with a water content greater than 2000 ppm wt. ppm could produce residual peridotites reaching the maximum water storage capacity (Fig. 6). The upwelling of such peridotites could continue to melt and form a gravitationally unstable melt column in the upper mantle. The upwelling of the MTZ with high water contents would trigger large-scale melting (Vinnik and Farra, 2007) and could be responsible for the formation of upper mantle plumes.

5. Conclusions

Based on our data, we propose a hydration mechanism for the Earth's upper mantle that is consistent with geophysical observations (magnetotelluric and seismic profiles) and geochemical constraints. In this bottom-up water transportation mechanism, the residual hydrous peridotites, formed as a result of melting at the 410-km discontinuity, upwell across the upper mantle, replacing initially dry mantle peridotites. The resulting water content in the residues is directly related to the water content of the mantle transition zone and the degree of melting. The upper limit of 200 ppm observed for the upper mantle can be maintained in residual peridotites if the water content of the MTZ is less than 1500 ppm. The slow H₂O diffusion rates in wadsleyite may enforce large gradient in H₂O distribution within the MTZ, and as a result, regional variations of H₂O contents in the upper mantle would be expected.

The ascent of residual hydrous peridotites could be responsible for the hydrous melting at the top of the asthenosphere.

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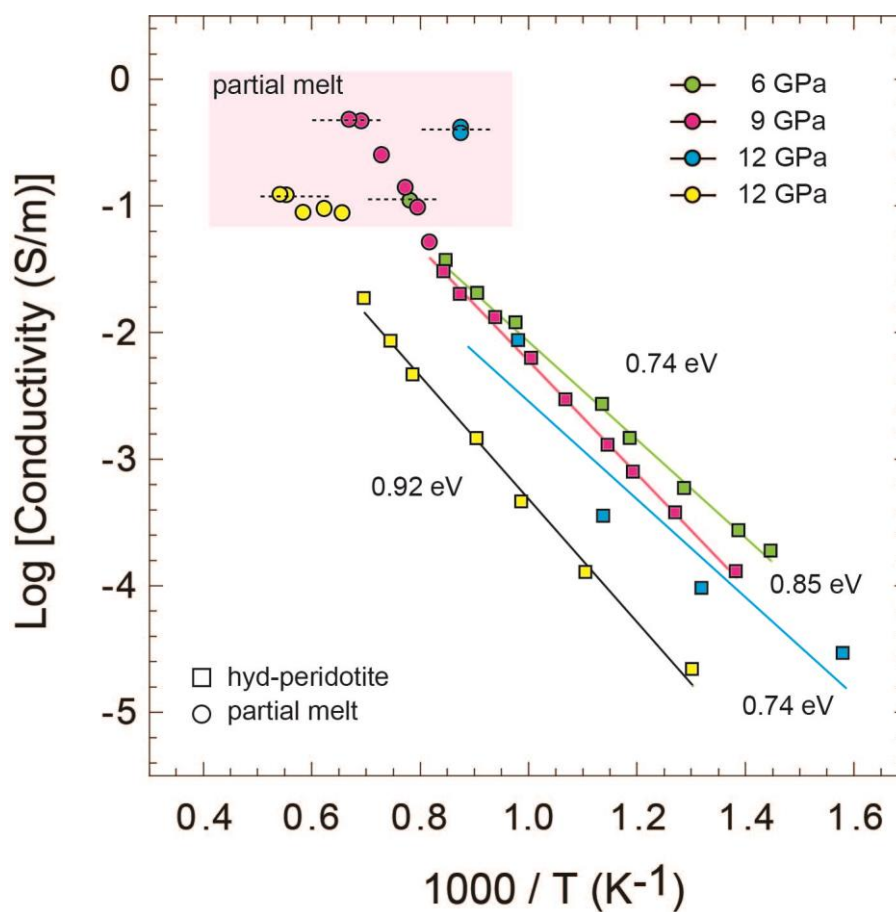
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655

656 **Figure 1. Electrical conductivity of hydrous peridotites at 6, 9 and 12 GPa.** The
 657 peridotites before melting are indicated in squares and after partial melting are in circles. The
 658 melt conductivities used for the discussion are shown with horizontal lines. Activation
 659 enthalpies are given in eV, next to the individual fits. The error bars associated with the
 660 electrical conductivity data measurements are less than the symbol size at high temperatures.
 661 The uncertainties in the estimation of the electrical conductivity result from the estimations of
 662 temperature, pressure, and sample dimensions as well as data fitting errors and are less than
 663 5%.

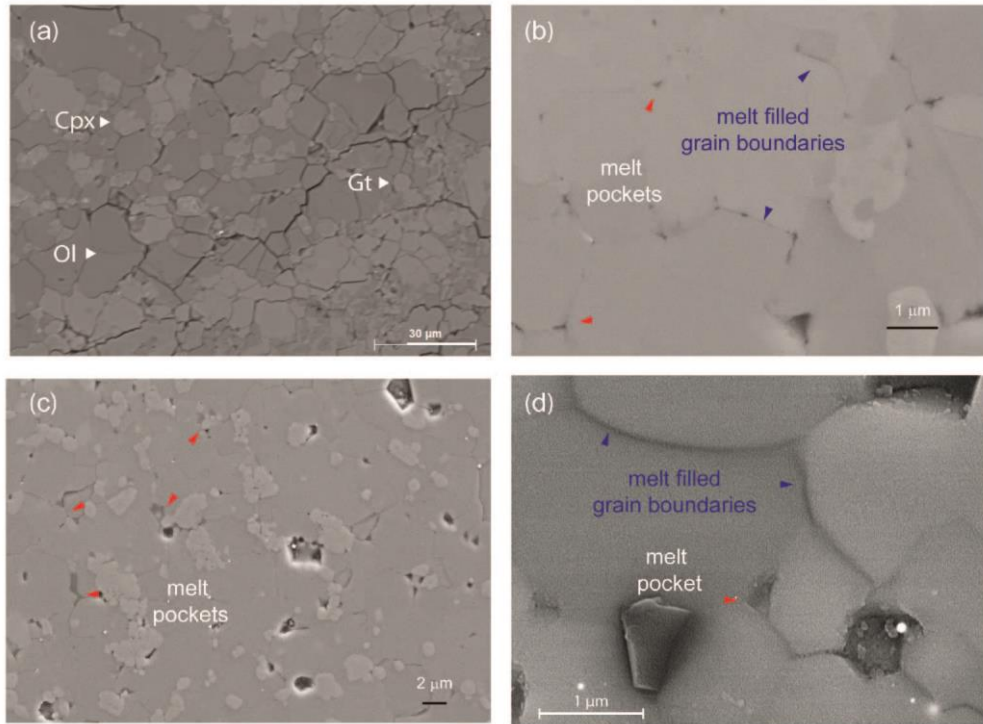


Figure 2. High resolution microphotographs of the recovered samples before and after the electrical conductivity measurements. (a) Hydrous peridotite sample showing mineral distribution before melting at 12 GPa (M421). (b) The mineral and melt distribution of partially molten sample after the electrical conductivity measurements at 6 GPa (M667), (c) at 9 GPa (M668), (d) and at 12 GPa (M425).

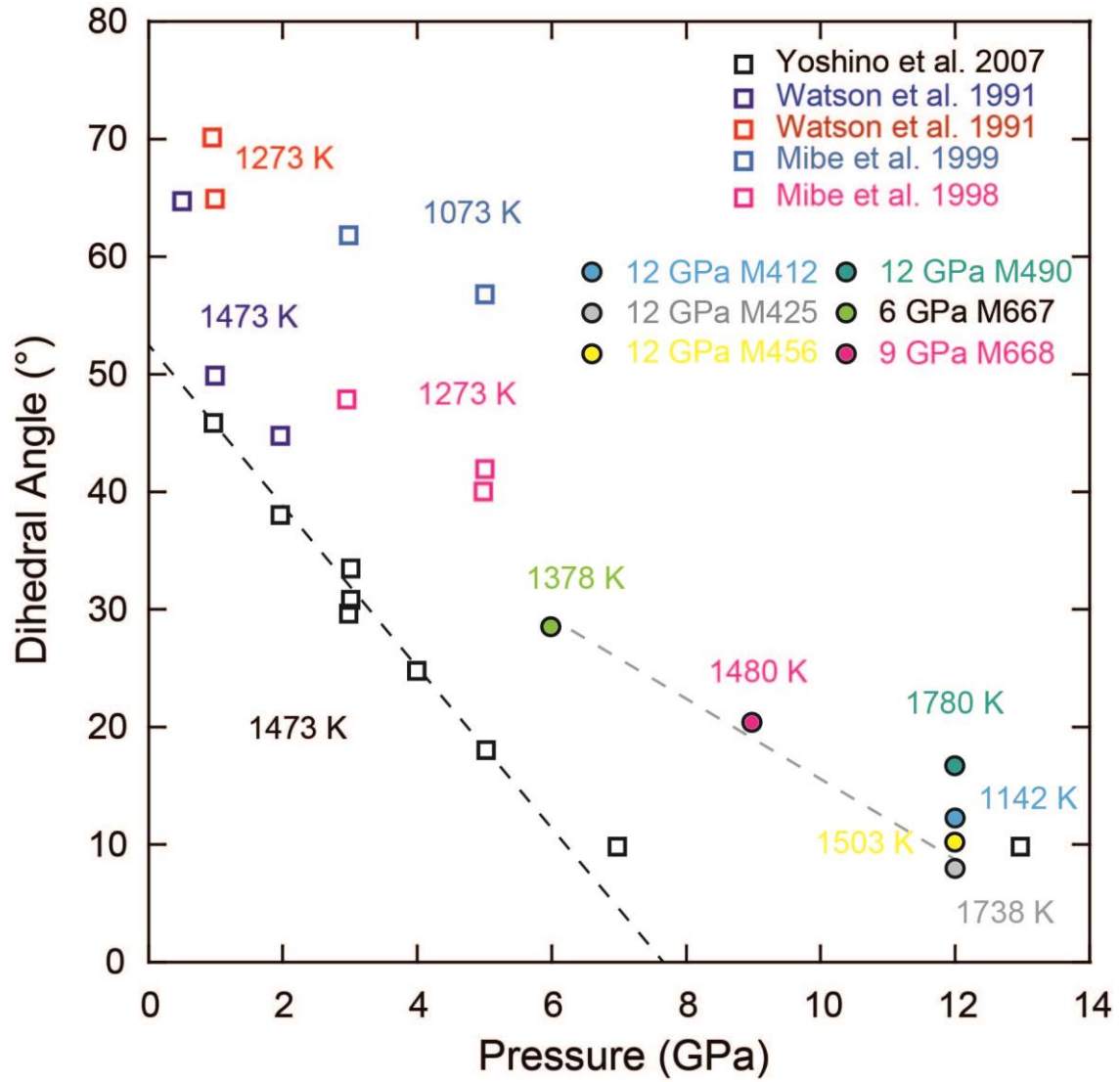


Figure 3. Dihedral angles estimated in our partially molten samples as a function of pressure. The maximum temperature of each experiment is indicated next to its symbol. Dihedral angles were estimated using FEG SEM and SEM image analysis with Matlab code to measure the angles by the tangent method. Analytical error is estimated to be 5° (1σ). Relevant literature values for similar or analogue systems at lower pressures have been represented for comparison (Mibe et al., 1999, 1998; Watson et al., 1991; Yoshino et al., 2007).

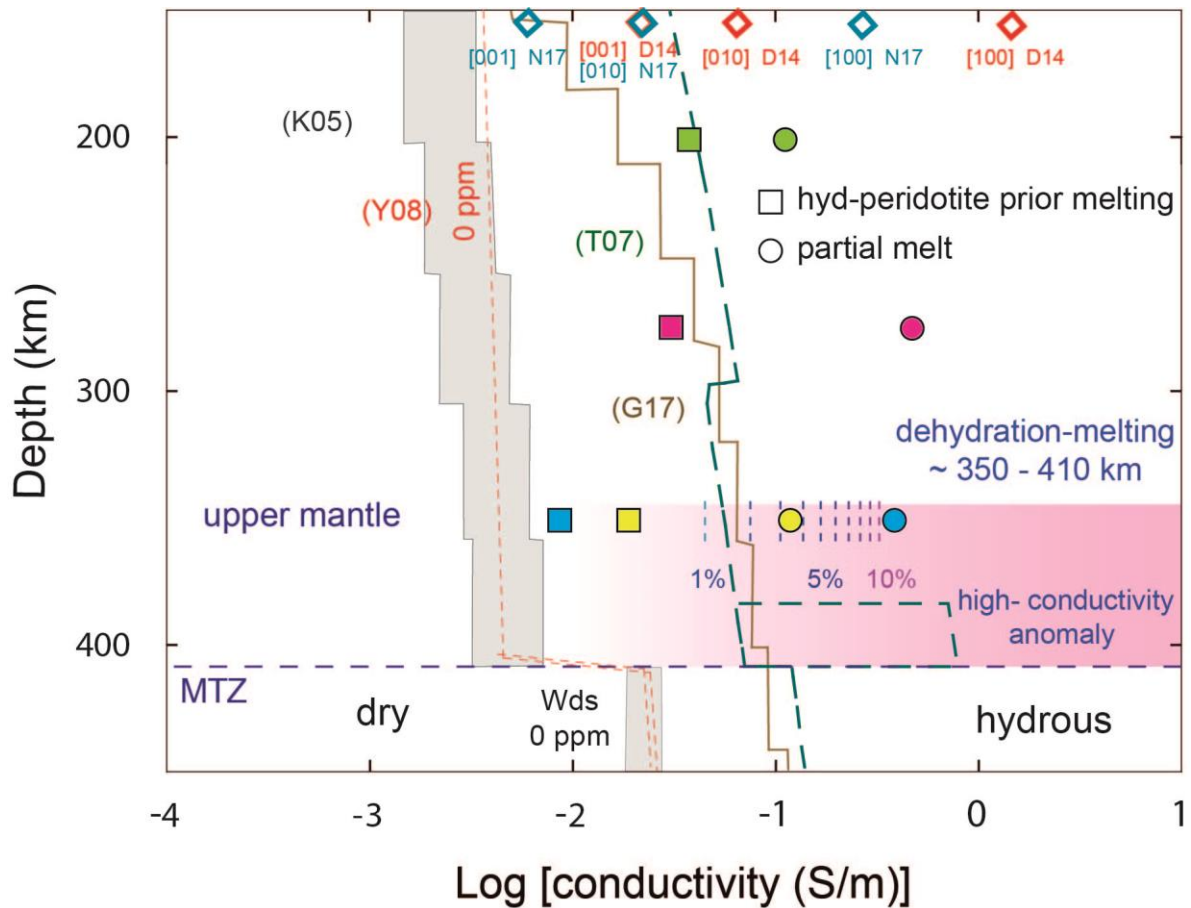


Figure 4. Electrical conductivity of hydrous silicate melts compared with geophysical profiles. Electrical conductivities of peridotite prior to melting (squares) and partially molten peridotites (circles) at 6, 9 and 12 GPa are displayed as a function of depth. Hashin Shtrikman upper bound (HS+) conductivities for different melt fraction are displayed for 12 GPa samples with dashed lines. The conductivity profiles are for oceanic mantle (pacific, K05) (Kuvshinov et al., 2005), continental (Tuscan SW USA, T07) (Toffelmier and Tyburczy, 2007) and global electrical conductivity model of Earth's mantle based on inverting satellite magnetic field measurements (G17) (Grayver et al., 2017). Pink shaded area indicates the possible distribution of hydrous melt at the top of the mantle transition zone. In this area the high conductivity anomaly modeled from EM data has been represented (Toffelmier and Tyburczy, 2007). Dry upper mantle electrical conductivity vs depth profile based on high pressure electrical conductivity measurements are also displayed (Yoshino et al., 2008). Electrical conductivities of oriented single crystal olivine from measurements (Dai and

Karato 2014) (4 GPa and extrapolated to mantle temperatures) and diffusion experiments (Novella et al., 2017b) (2 GPa, extrapolated to mantle temperatures and with 80 ppm of water) have been reported. Electrical conductivity anomaly of 350% cannot be reproduced by moderately anisotropic solid rock but rather require presence of melts. Our solid hydrous peridotites conductivities agree well with randomly oriented olivine values.

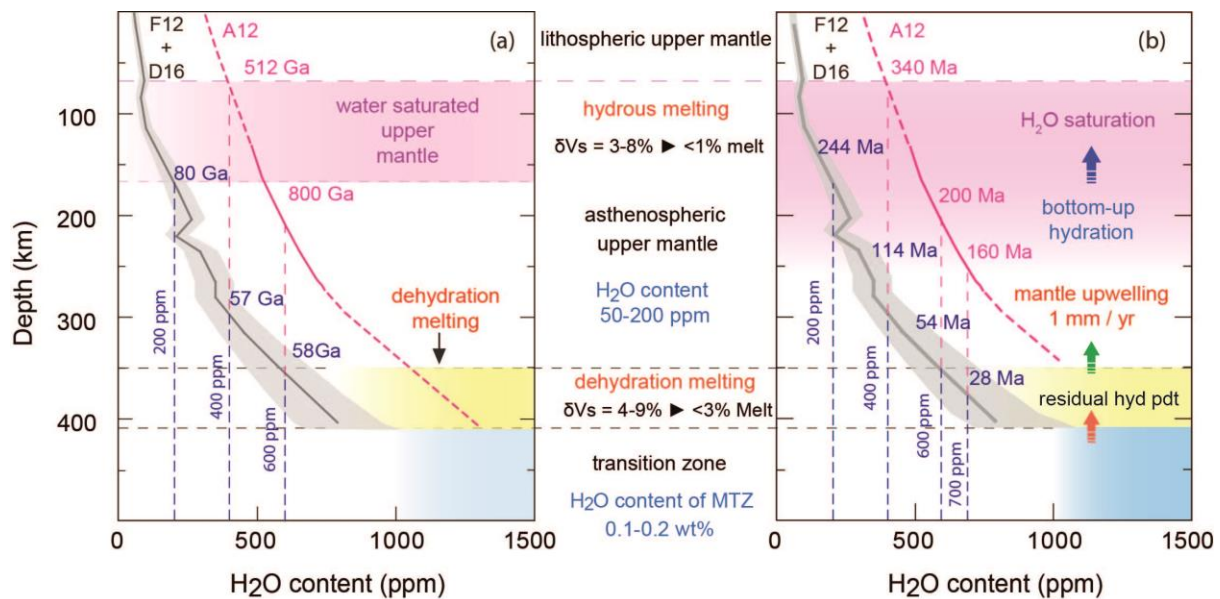


Figure 5. Comparison of water transport mechanisms of the Earth's upper mantle. (a) Hydration of the Earth's upper mantle by diffusion. The estimations are based on the diffusion coefficient of $7.89 \times 10^{-9} \text{ m}^2/\text{s}$ (Novella et al., 2017a) at 1723K with initial H₂O concentration of 800 ppm wt. and variable diffusion lengths (from 410-km to the saturation depth at a given water content) (see also Supplementary Fig. S7). The water saturation curves are for peridotite without considering garnet (F12) (Férot and Bolfan-Casanova, 2012) and considering water partition between olivine and garnet (0.9) (A12) (Ardia et al., 2012). The broken lines indicate the time required to saturate the upper mantle by diffusion at given depths for a given bulk water content. The shaded in yellow indicates regions where dehydration-melting is expected. The shaded in pink indicate depth range for mantle H₂O saturation which can't be explained by diffusion process. (b) Hydration by upwelling. The dehydration melting at the 410-km discontinuity (Freitas et al., 2017) produces residual peridotites with different H₂O concentration according melt fractions and compositions. The continuous upwelling of hydrous peridotite replaces the initially dry peridotite in the upper mantle, forming hydrous upper mantle. The ascending peridotite reaches water saturation levels at different depths (Ardia et al., 2012; Férot and Bolfan-Casanova, 2012), and should initiate hydrous melting. The broken lines indicate the time required to saturate the upper

mantle at given depths for a given bulk water content. The water saturation curves are for peridotite without considering garnet (F12) (Férot and Bolfan-Casanova, 2012) and with considering water partition between olivine and garnet (A12) (Ardia et al., 2012). The shaded in yellow indicates regions where dehydration-melting is expected. The shaded in pink indicate depth range for mantle H₂O saturation.

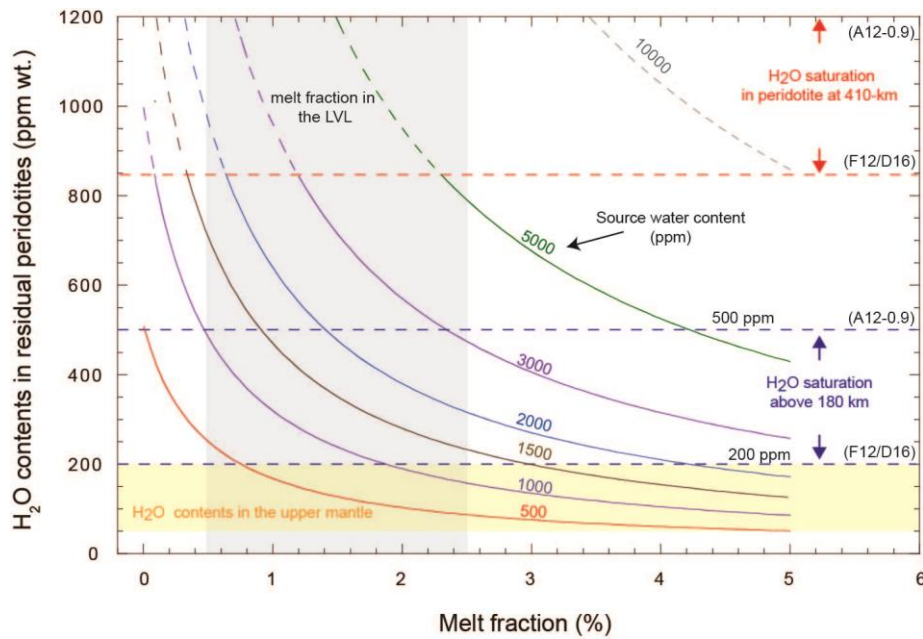


Figure 6. Bottom-up hydration of the upper mantle. The H₂O contents are calculated assuming batch melting scenario at the 410-km discontinuity. The H₂O contents in the source (MTZ) are marked next to individual lines. The Gray shaded area indicates the melt fraction expected at the partially molten layer above the MTZ. The area shaded in Yellow indicates H₂O content of the upper mantle. Vertical lines (F12/D16) (Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012) and (A12) (Ardia et al., 2012) indicate water saturation limits at different depths based on water solubilities of peridotites.

Supplementary materials for:

Electrical conductivity of hydrous silicate melts: Implications for the bottom-up hydration of Earth's upper mantle

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Supplementary materials

Here we give supplementary information concerning:

- **The estimation of the H₂O partition coefficient for peridotite/melt at 12 GPa (Text S1)**
- **Experimental conditions and parameters of syntheses and *in situ* measurements (table S1)**
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- **Water diffusion calculations for the hydration of Earth's upper mantle. (Figure S7)**

Supplementary Text S1: The estimation of the H₂O partition coefficient for peridotite/melt at 12 GPa.

The H₂O partition coefficient between peridotite and melt is available at 6 GPa (Novella et al., 2014). Due to the significant variation of mineral proportions at 12 GPa, we have estimate the H₂O partition coefficient between peridotite and melt based on mantle mineralogy at 12 GPa. The bulk partition coefficient can be calculated by:

$$D_{H_2O}^{pdt/melt} = \sum X_i Kd_i$$

Where X is the fraction of each mineral (i) and Kd its partition coefficient for water with melt. The Kd values are taken from Novella et al. 2014 (Novella et al., 2014) and mineral proportions where estimated using the mineral proportions estimated at 12 GPa in our study. Mineral proportion at 12 GPa are observed to be 60 % of olivine, 28 % of garnet (Gt) and 12 % of clinopyroxene (Cpx).

	Kd (Novella et al., 2014) STD	
Melt/olivine	0.004	0.0006
Melt/Opx	0.0064	0.0004
Melt/Cpx	0.0115	0.0016
Melt/garnet	0.0032	0.0008
D _{Melt/pdt} 6 GPa	0.006288	0.000343
D _{Melt/pdt} 12 GPa	0.004676	0.000424

Table S1. Conditions of experimental runs (syntheses and *in situ* measurements). Each run conditions are given in the first section: pressure, temperature and duration at maximum temperature. *In situ* experiments were only kept for short times above melting temperature (<1h) to avoid any melt escape or reaction. Water content are then given within the 3 next columns: initial starting water content in powders, water content in solid samples after syntheses and water content after *in situ* electrical conductivity measurements. Water content on solid samples/samples after *in situ* measurements was estimated qualitatively (or with large errors) by Raman spectroscopy (see methods section and Freitas et al. 2017). Partially molten samples characteristics are given, with melt fractions and estimated melting temperatures based on electrical conductivity. Dihedral angles measurements are given with average and median angles for each partially molten sample. In the last column (remarks), the type of each experiment is given; synthesis run number used for electrical conductivity measurements is given in parenthesis. Errors given in parenthesis are 1 standard deviation.

¹ *Water content of the bulk sample estimated with quenched glass quantification and partition coefficient (Novella et al. 2014) with mineral proportions inferred from BSE image analyses.*

² *Melt fractions estimated by mass balance, partition coefficients and image surface analyses.*

³ *Melt fractions estimated by image surface analysis only.*

	Experimental conditions			Water contents (ppm)			Partial melting		Dihedral angle Θ			
Run #	Pressure (GPa)	Tmax (K)	Time	initial powder	Solid sample	After <i>in situ</i> EC ¹	%melt ²	Tmelting (K) ⁴	Number of measurements	Mean	Median	Remarks
M405	5	1523 (20)	3h45	0	-	-	-	-	-	-	-	Synthesis
M407	5	1473 (20)	1h	0	-	-	-	-	-	-	-	Synthesis
M408	14	1373 (20)	7h	≈500	>3000	-	-	-	-	-	-	Synthesis
M410	14	1373 (20)	2h10	≈500	3267 (650)	-	-	-	-	-	-	Synthesis
M412	12	1738 (5)	<1h	0	-	3098 (959)	7.00 (0.27)	1143 (5)	77.00	12.20 (11.68)	10.51	EC (M405)
M415	5	1450 (20)	2h30	0	-	-	-	-	-	-	-	Synthesis
M421	12	1385 (5)	<1h	0	-	993 (260)	-	-	-	-	-	EC (M415)
M425	12	1738. (5)	<1h	≈500	≈3000	3030 (627)	2.00 (0.45)	1493 (5)	139.00	7.88 (8.79)	6.23	EC (M408)
M455	14	1373 (20)	2h	≈700	≈3600	-	-	-	-	-	-	Synthesis
M456	12	1503 (5)	<1h	≈700	≈3600	-	3.50 (2.44)	1303 (5)	59.00	10.12 (10.26)	8.50	EC (M455)
M481	2	1473 (20)	2h	0	-	-	-	-	-	-	-	Synthesis
M482	14	1323 (20)	2h10	-	500-700	-	-	-	-	-	-	Synthesis (M481)
M490	12	1780 (5)	<1h	0	500-700	800-1650	25.65 (2.50)	1561 (5)	36.00	16.60 (9.52)	17.09	Freitas et al. 2017 (M481)
M492	12	1363 (5)	<1h	0	500-700	490 (125)	-	-	-	-	-	Freitas et al. 2017 (M482)
M665	6	1273 (20)	4h15	≈700	1579 (413)	-	-	-	-	-	-	Synthesis
M666	9	1323 (20)	3h30	≈700	1740 (523)	-	-	-	-	-	-	Synthesis
M667	6	1378 (5)	<1h	≈700	1579 (413)	-	3.68 (1.86)³	1163 (5)	104.00	28.46 (13.19)	27.26	EC (M665)
M668	9	1480.20 (5)	<1h	≈700	1105 (422)	-	6.33 (1.01)³	1181 (5)	109.00	20.32 (9.53)	19.49	EC (M666)

Table S2. Average chemical composition of solid mineral phases at 6 and 9 GPa both before and after EC measurements. Each value is an average of 3 to 10 analyses; the standard deviation is indicated in parenthesis. Chemical analyses of quenched melt were not possible due to the extremely small size of the melt pockets.

	Starting material at 6 GPa			After EC exp. at 6 GPa			Starting material at 6 GPa			After EC exp. at 6 GPa		
	Ol	Cpx	Gt	Ol	Cpx	Gt	Ol	Cpx	Gt	Ol	Cpx	Gt
SiO ₂	40.00 (0.90)	54.82 (0.22)	46.13 (0.92)	40.65 (0.87)	54.42 (0.73)	42.13 (2.84)	39.30 (1.00)	53.58 (0.63)	39.81 (2.45)	39.28 (1.39)	51.14 (0.96)	40.68 (0.22)
TiO ₂	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
Al ₂ O ₃	0.29 (0.37)	2.34 (0.64)	14.66 (0.49)	0.44 (0.43)	2.59 (0.36)	15.41 (6.69)	1.03 (0.03)	1.48 (0.15)	18.50 (4.68)	0.20 (0.20)	1.79 (0.22)	20.53 (0.54)
Cr ₂ O ₃	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
FeO	10.71 (1.86)	3.93 (1.35)	8.80 (0.77)	10.79 (2.54)	2.77 (0.09)	9.01 (1.00)	8.81 (0.22)	4.22 (3.06)	10.84 (1.85)	9.18 (1.34)	3.06 (0.30)	9.17 (0.12)
MnO	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
MgO	40.00 (0.90)	19.12 (0.46)	22.75 (0.74)	47.17 (2.57)	19.70 (0.68)	42.13 (2.84)	50.00 (1.25)	19.30 (0.94)	23.29 (5.35)	48.25 (2.45)	20.91 (2.57)	20.47 (2.93)
CaO	0.09 (0.05)	18.38 (2.84)	6.79 (1.53)	0.16 (0.11)	18.97 (0.60)	5.04 (2.87)	0.17 (0.01)	18.88 (4.42)	4.81 (1.22)	0.14 (0.10)	18.65 (1.82)	5.87 (0.71)
Na ₂ O	0.22 (0.28)	1.91 (0.68)	0.72 (0.11)	0.31 (0.37)	1.79 (0.27)	0.60 (0.19)	0.58 (0.02)	1.78 (0.90)	0.49 (0.28)	0.27 (0.24)	1.42 (0.19)	0.17 (0.21)
NiO	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)
K ₂ O	-	-	-	-	0.01 (0.02)	0.01 (0.02)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	-	0.01 (0.01)	0.05 (0.02)
Total	99.91 (1.15)	100.52 (0.57)	99.84 (0.86)	99.55 (0.94)	100.19 (0.33)	99.89 (1.36)	99.9 (2.50)	99.24 (0.47)	97.74 (1.11)	97.36 (0.28)	96.97 (0.58)	96.95 (1.53)
Ol: olivine, Cpx: clino-pyroxenes, Gt: garnet												

Table S3. Average chemical composition of solid mineral phases at 12 GPa. Each value is an average of 5 to 20 analyses. The standard deviation is given on the average value in parenthesis.

	M412-7 (0.27)% Melt			M421-0% Melt			M425-2 (0.45)% Melt			M456-3.5 (2.44)% Melt		
	Ol	Cpx	Gt	Ol	Cpx	Gt	Ol	Cpx	Gt	Ol	Cpx	Gt
SiO₂	41.60 (0.38)	56.48 (1.01)	47.23 (0.89)	40.90 (0.87)	54.62 (0.65)	40.69 (1.38)	41.36 (0.26)	54.78 (0.67)	43.95 (1.46)	41.99 (0.48)	55.73 (0.36)	46.51 (4.01)
TiO₂	0.00 (0.00)	0.03 (0.01)	0.78 (0.38)	0.04 (0.03)	0.10 (0.04)	0.57 (0.39)	0.04 (0.03)	0.06 (0.03)	0.43 (0.16)	0.05 (0.04)	0.10 (0.05)	0.32 (0.26)
Al₂O₃	0.12 (0.02)	2.66 (0.29)	8.87 (2.21)	0.04 (0.08)	1.66 (0.46)	22.19 (3.04)	0.44 (0.60)	1.74 (0.57)	17.13 (5.31)	0.15 (0.04)	1.83 (0.40)	13.17 (10.42)
Cr₂O₃	0.12 (0.02)	0.01 (0.02)	0.01 (0.01)	0.01 (0.02)	0.01 (0.02)	0.03 (0.02)	0.01 (0.01)	0.02 (0.01)	0.03 (0.02)	0.00 (0.00)	0.00 (0.02)	0.00 (0.00)
FeO	7.15 (0.24)	4.83 (0.48)	9.77 (1.77)	8.99 (1.12)	2.97 (0.71)	7.73 (0.58)	7.01 (0.78)	2.90 (0.35)	6.34 (0.61)	5.55 (1.96)	2.96 (0.15)	5.17 (0.99)
MnO	0.02 (0.02)	0.02 (0.02)	0.05 (0.02)	0.02 (0.02)	0.01 (0.01)	0.06 (0.02)	0.02 (0.02)	0.02 (0.02)	0.06 (0.03)	0.01 (0.01)	0.02 (0.01)	0.05 (4.59)
MgO	50.78 (0.58)	33.61 (0.73)	13.64 (8.72)	48.80 (0.68)	18.74 (5.83)	18.51 (4.17)	50.10 (1.20)	19.09 (1.18)	24.97 (7.53)	52.67 (2.02)	17.83 (0.62)	24.57 (4.59)
CaO	0.18 (0.03)	1.70 (0.23)	15.99 (4.72)	0.11 (0.12)	20.04 (4.33)	8.77 (7.09)	0.36 (0.30)	19.79 (1.07)	6.94 (1.97)	0.23 (0.14)	20.96 (0.50)	10.58 (2.07)
Na₂O	0.02 (0.02)	0.21 (0.13)	1.46 (0.42)	0.02 (0.04)	1.31 (0.29)	0.06 (0.02)	0.03 (0.02)	0.97 (0.18)	0.11 (0.07)	0.03 (0.02)	1.23 (0.12)	0.31 (0.25)
NiO	0.01 (0.01)	0.07 (0.02)	0.00 (0.00)	-	-	-	-	-	-	0.01 (0.01)	0.00 (0.00)	0.02 (0.01)
K₂O	0.01 (0.01)	0.01 (0.02)	0.06 (0.06)	0.01 (0.01)	0.03 (0.02)	0.01 (0.02)	0.00 (0.00)	0.01 (0.01)	0.01 (0.01)	0.01 (0.01)	0.02 (0.01)	0.02 (0.01)
Total	99.88 (0.93)	100.16 (1.14)	97.86 (5.21)	98.94 (0.67)	99.50 (1.40)	98.62 (1.05)	99.38 (0.45)	99.39 (0.57)	99.95 (1.70)	100.70 (0.29)	100.68 (0.61)	100.71 (1.03)

Ol: olivine, Cpx: clino-pyroxenes, Gt: garnet

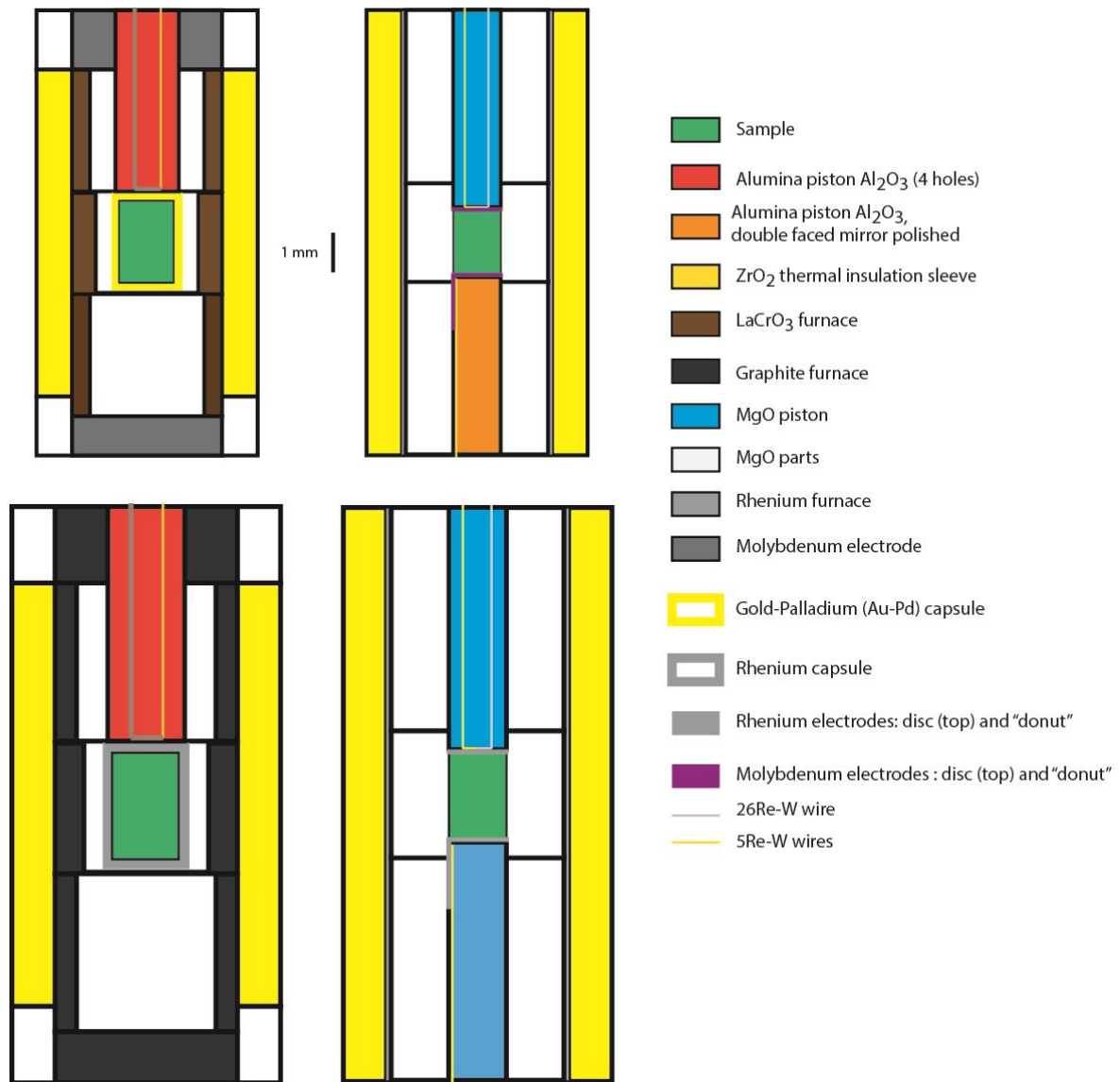


Figure S1. Schematic cross section of the high pressure assemblies. Assembly geometries on the left were used for the synthesis of the peridotite samples and on the right were used for electrical conductivity measurements at 6, 9 and 12 GPa. Top assemblies are 14/8 geometries whereas bottom ones are 18/11.

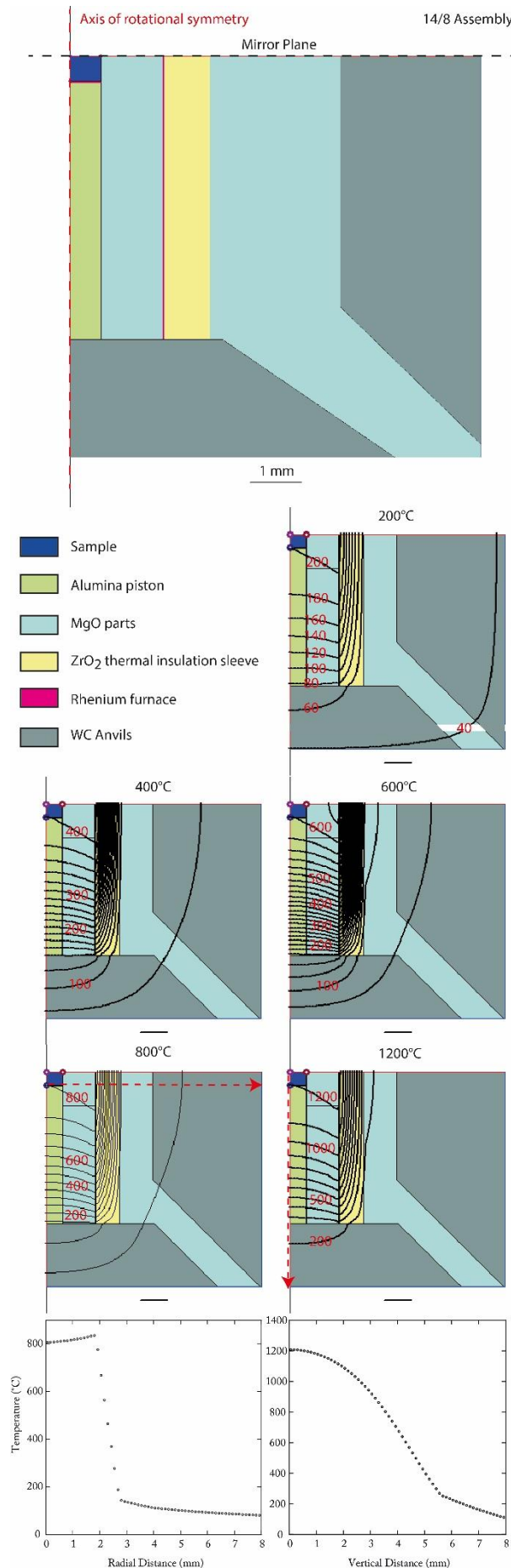


Figure S2. Modeling of thermal gradient in our assembly. Results of simulations using the software codes of Hernlund et al. 2006 (Hernlund et al., 2006) . The maximum temperature gradient of the assembly at 1500 K was estimated to be less than 20 K.

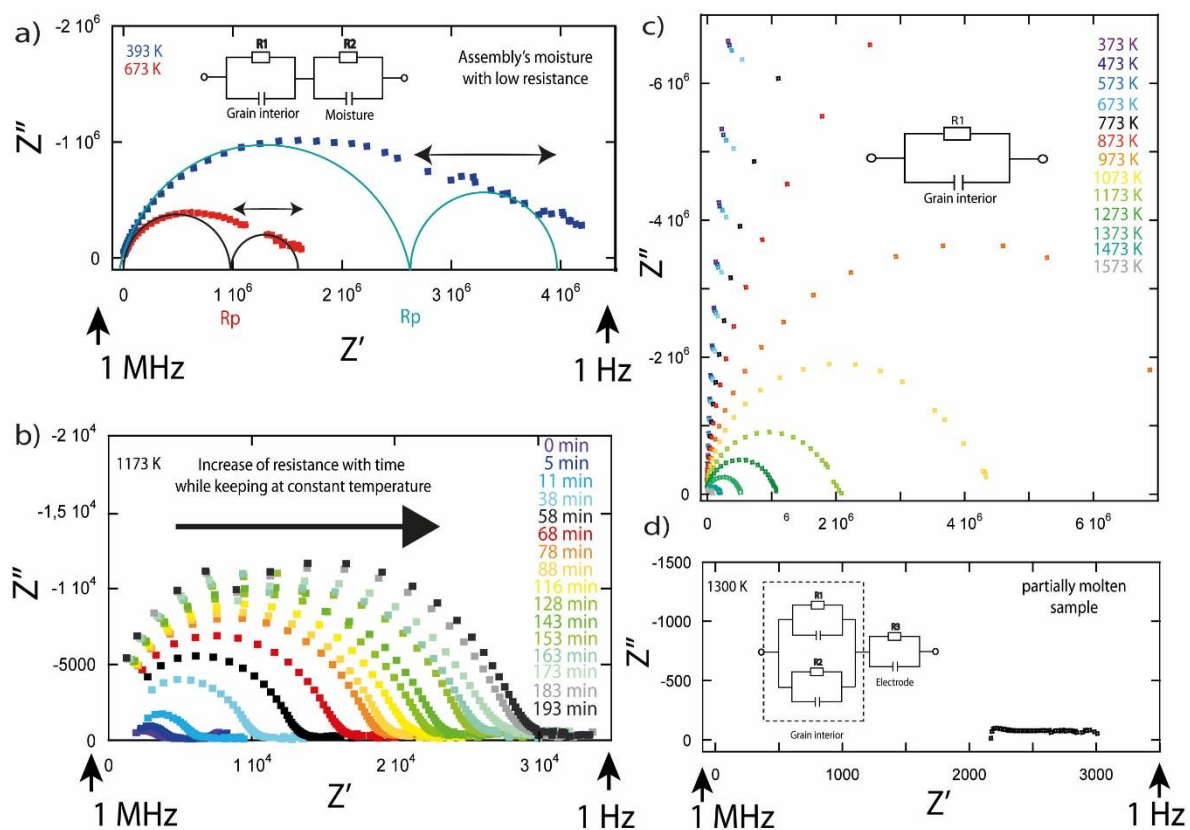


Figure S3. Impedance spectra of the sample at different stages of heating. (a) during assembly dehydration at 673 K (b) sample annealing at 1173 K. (c) Impedance spectra recorded after assembly dehydration and sample annealing steps. Moisture tails were not observed anymore during this cycle. (d) Impedance spectra recorded after the onset of melting, these spectra are characterized by the disappearance of the semicircular shape for induction patterns.

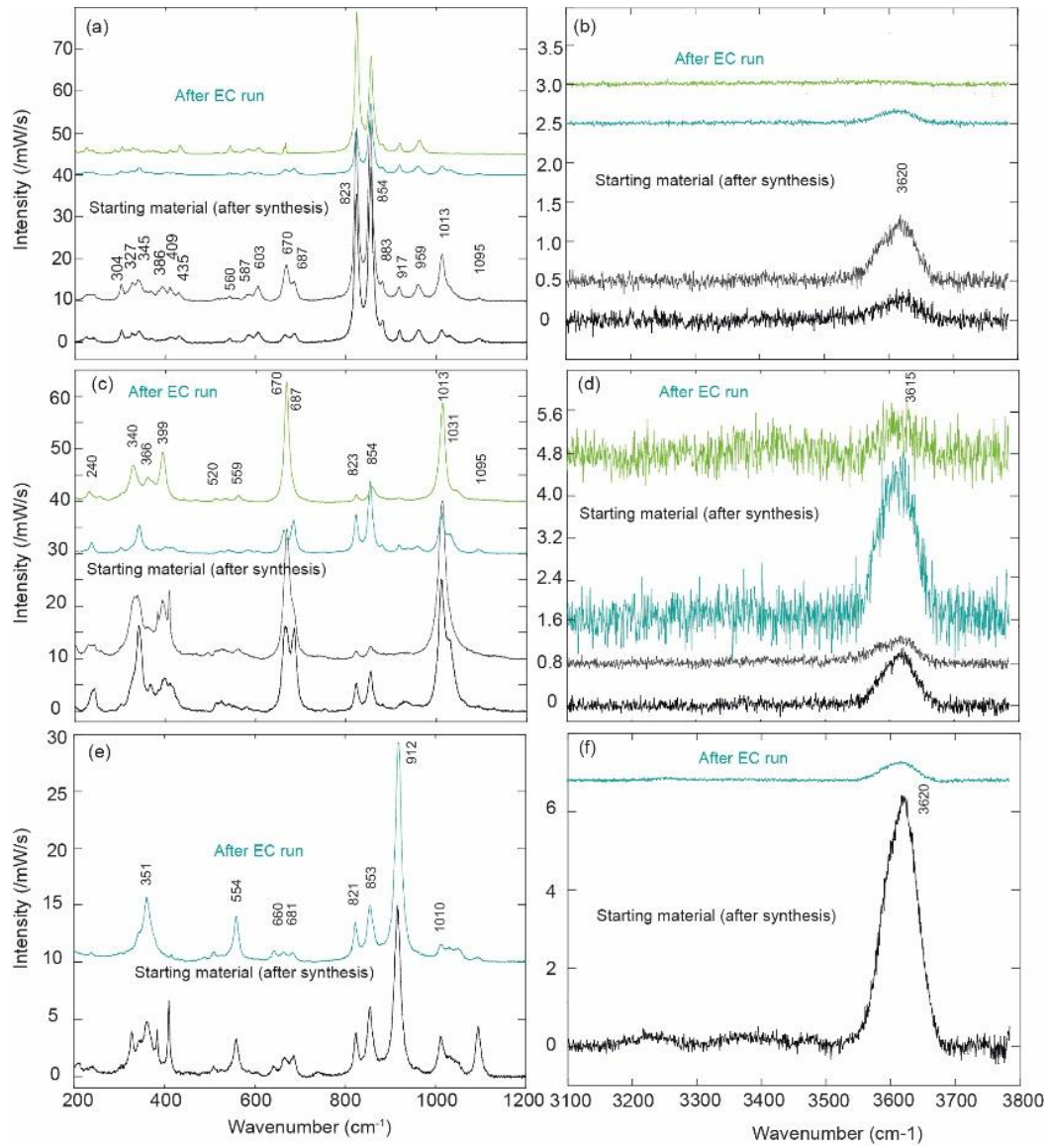


Figure S4. Representative Raman spectra from starting materials and recovered experimental runs at 6 GPa. Low-wavenumber (200-1200 cm⁻¹) bands corresponding to vibrations of the silicate network (a, c, e) and high-wavenumber bands (3100-3800 cm⁻¹) corresponding to O-H vibrations (b, d, f) of olivine, pyroxene and garnet, respectively. The presence of H₂O is visible in all samples. H₂O content is significantly reduced in the experiments that undergoes partial melting. Black and gray spectra were taken in starting material after the synthesis run, green spectra were acquired on samples recovered after the *in situ* measurements.

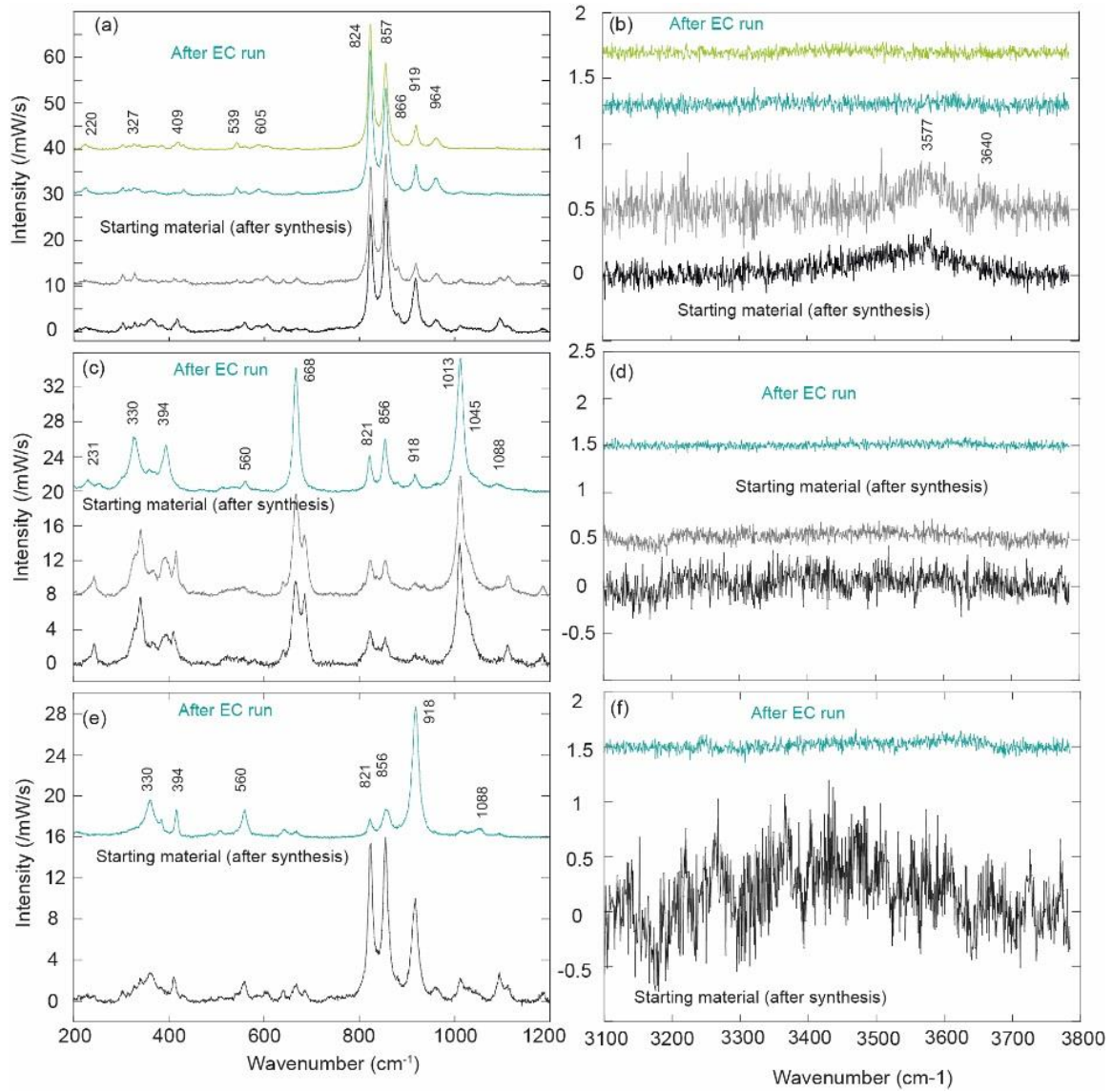


Figure S5. Representative Raman spectra from starting materials and recovered experimental runs at 9 GPa. Low-wavenumber (200-1200 cm^{-1}) bands corresponding to vibrations of the silicate network (a, c, e) and high-wavenumber bands (3100-3800 cm^{-1}) corresponding to O-H vibrations (b, d, f) of olivine, pyroxene and garnet, respectively. The presence of H_2O is visible in all samples. H_2O content is significantly reduced in the experiments that undergoes partial melting. Black and gray spectra were taken in starting material after the synthesis run, green spectra were acquired on samples recovered after the *in situ* measurements.

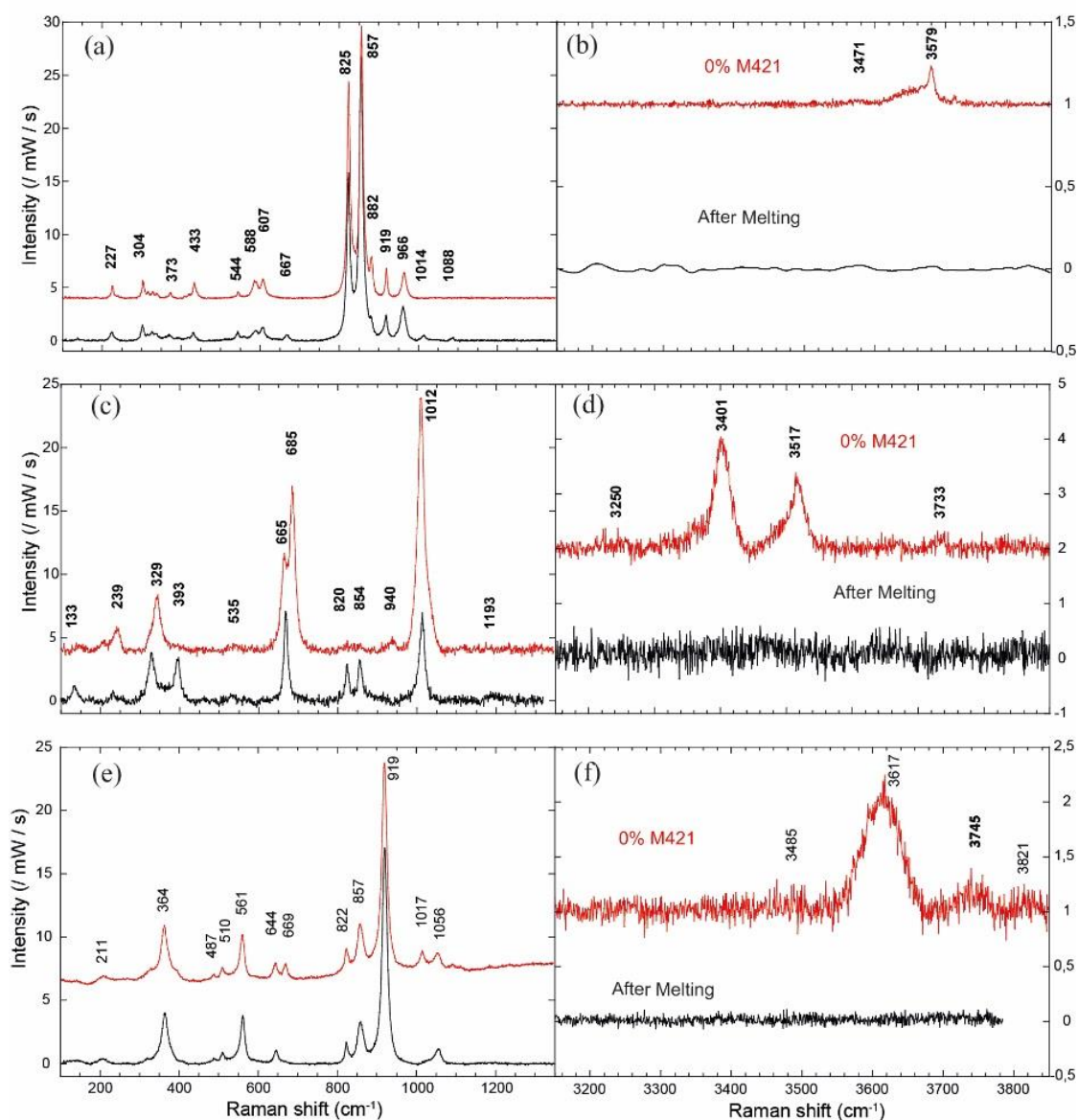


Figure S6. Representative Raman spectra from recovered experimental runs at 12 GPa.

Red spectra were measured on samples with 600 ppm wt. bulk H₂O contents. Black spectra were acquired on the samples that undergone partial melting. Spectra on the left column (a, c, e) were made on silicate frequency region (200-1200 cm⁻¹) whereas spectra displayed on the right column (b, d, f) were performed on O-H vibration frequency region (3100-3800 cm⁻¹) of olivine, pyroxene and garnet, respectively.

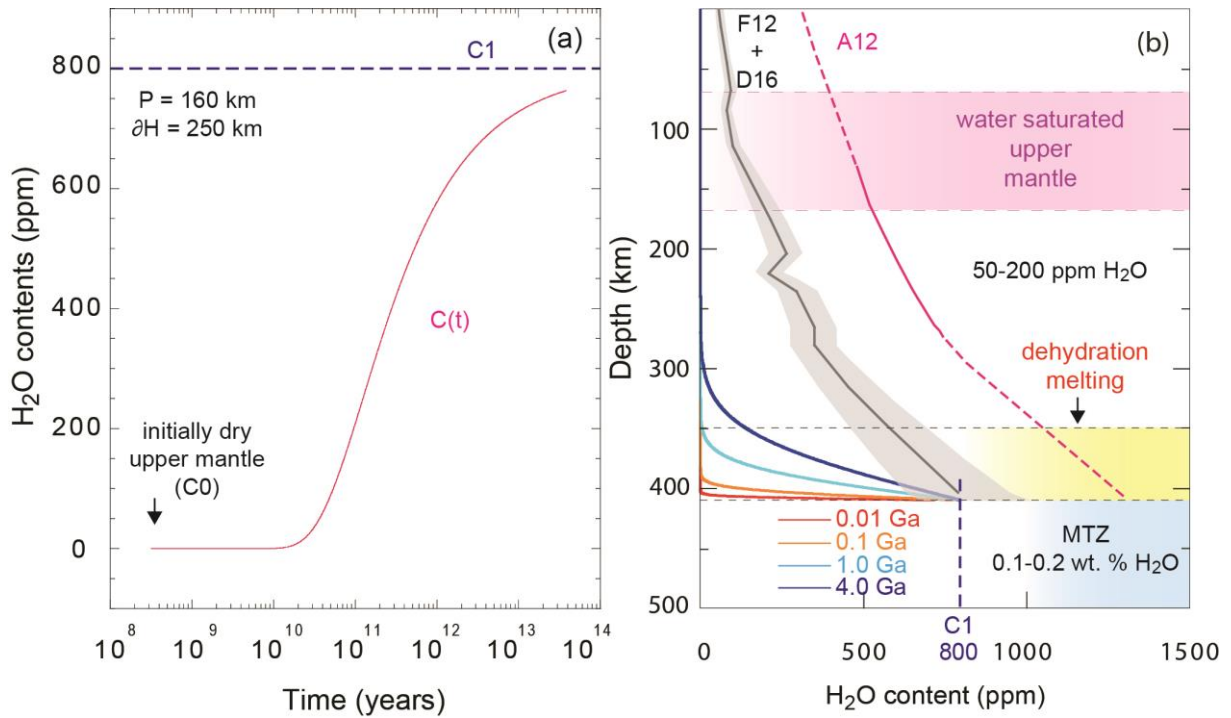


Figure S7. Water diffusion calculations for the hydration of Earth's upper mantle. (a). Water concentration profiles as a function of time at depth of 160 km (250 km of diffusion length). The diffusion profile is calculated for an initially dry upper mantle ($C_0 = 0$ ppm) and considering an olivine peridotite in contact with hydrous melt at 410-km discontinuity with a constant, sustained water concentration $C_1 = 800$ ppm wt. The calculations are based on the diffusion coefficient of $7.82 \times 10^{-9} \text{ m}^2/\text{s}$ (Novella et al., 2017) at 1723K (b) Water concentration profiles in the upper mantle for different times of diffusion (0.01 to 4 Ga) for an initially dry upper mantle and 800 ppm wt. olivine peridotite in equilibrium with hydrous melt at the top of the MTZ. The water saturation curves are for peridotite without considering garnet (F12+D16) (Demouchy and Bolfan-Casanova, 2016; Férot and Bolfan-Casanova, 2012) and with considering water partition between olivine and garnet (0.9) (A12) (Ardia et al., 2012). The diffusion profiles even for Earth's time scale can't reach the saturation curve. Diffusion is able to keep large gradients of water concentration in Earth's upper mantle over geological times.

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