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Dense hydrous silica carrying water to the deep Earth and promotion of oxygen fugacity heterogeneity

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ABSTRACT

Water has remarkable effects on the properties of mantle rocks, but, owing to the high temperatures in the mantle, uncertainties remain about how and how much water is transported into the deep Earth. Recent studies have shown that stishovite and post-stishovites as high-pressure phases of SiO_2 have the potential to carry weight percent levels of water into the Earth's interior along the geotherm of the subducting oceanic crust. As slabs are subducted to the deepest mantle, dehydration of these dense hydrous silica phases has the potential to change the physicochemical properties of the mantle by reducing melting points, forming new high-pressure phases, and enhancing the oxygen fugacity heterogeneity of the lower mantle.

Water plays a significant role in the chemical and physical properties of planetary interiors, such as mineral phase relations, melting temperatures, electrical conductivity, and seismic velocities. On the basis of the H_2O/Ce ratio of oceanic basalts and convecting mantle Ce concentration, Hirschmann[1] estimated that there was one ocean mass of water contained in the Earth's mantle. Subduction of the basaltic oceanic crust is thought to be an essential carrier transporting water from the surface to the interior, but, owing to the high temperatures in the mantle, it is still unclear how and how much water is transported to the deep mantle.

Previous studies have indicated that warmer subduction zones become dry when serpentine dehydrates at the first "choke point" at ~200 km depth and that water could be transported deeper by post-serpentine dense hydrous magnesium silicate (DHMS) phase A, phase E, superhydrous phase B, phase D, and phase E in cooler subducted slabs.[2] At the second "choke point" at ~700–800 km depth, beyond the transition zone (which is mainly composed of wadsleyite and ringwoodite), phase D, phase H, and ringwoodite break down to nominally anhydrous minerals dominated by bridgmanite and (ferro)periclase along slab geotherms, even in cooler subduction zones.[2] However, previous studies have shown that these identified DHMS phases can potentially stabilize water in subducted slabs or the lower mantle.[2,3]

Subducted lithosphere sinks gravitationally to the base of the mantle, with the oceanic crust, providing negative buoyancy when it becomes denser than lower mantle rocks at depths greater than ~700 km. In addition to DHMS from basaltic slabs, silica (SiO_2), as a free mineral phase, crystallizes as a major component of subducted oceanic crust (~20 wt. %) at depths greater than about 300 km, undergoing several polymorphic phase changes as the crust subducts to the core–mantle boundary (CMB).[4] Stishovite, a dense SiO_2 polymorph, is stable in oceanic crust along the mantle geotherm from ~10 to ~75 GPa, where it is converted to a distorted orthorhombic $CaCl_2$-type silica through a second-order displacive phase transition. Recent experimental work has shown that dense hydrous silica (hydrous stishovite, hydrous $CaCl_2$-type SiO_2 and hydrous seifertite), hereinafter referred to as DHS, can accommodate H_2O from ~3.5 wt. % (~0.7 wt. % in bulk crust) to ~1 wt. % (~0.2 wt. % in bulk crust) on subduction along the mantle geotherm (Fig. 1), which is equivalent to about one ocean mass of water transported by DHS into the lower mantle in the last 4 Ga, and *ab initio*

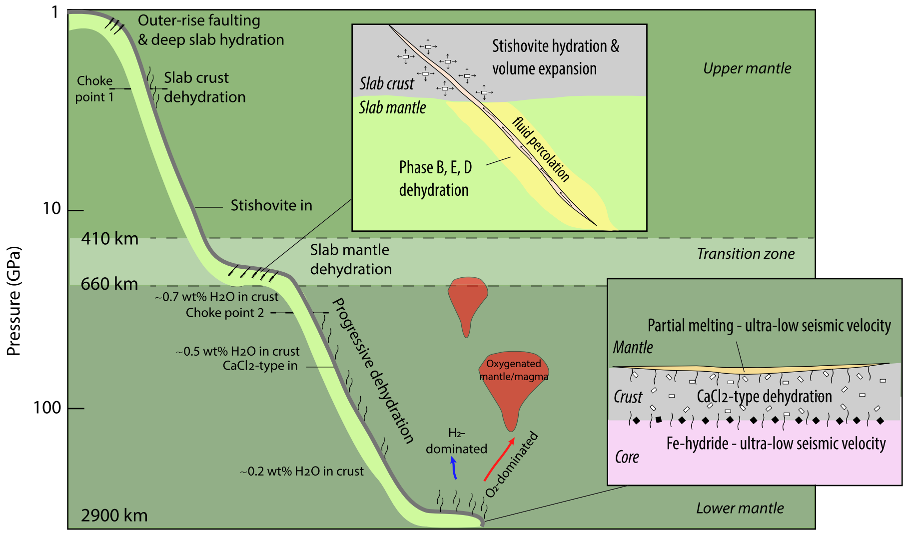

FIG. 1. Subduction of oceanic lithosphere drives material circulation in the Earth's mantle, providingan important conduit for the deep-water cycle and the coevolution of the hydrosphere and solid Earth. Dehydration of water-bearing stishovite, $CaCl_2$-type silica, and seifertite (DHS) during subduction results in the addition of water to mantle rocks, and this can significantly affect mineral phase relations, melting temperatures, and seismic velocities. Importantly, hydrous $CaCl_2$-type silica can continuously carry water into the base of the mantle at mantle temperatures over 3000 K.[6] The O_2-dominated volatiles decomposed from the released water or newly formed high-pressure peroxides can oxygenate the surrounding mantle and promote melting.[7–9]

calculations indicate that interstitial H_2O substitution is the main water incorporation mechanism.[5,6]

Water-bearing stishovite, as a stable and major phase of subducted basaltic oceanic crust at depths from ~300 to ~1700 km, has the potential to overcome the limit from the second "choke point" and continue to carry water to greater depths.[2,6] Then, hydrous $CaCl_2$-type SiO_2 at >75 GPa will take up the responsibility for transporting part of the water from hydrous stishovite down to the CMB, even though phase H as one of the DHMS phases will break down along the mantle geotherm.[6] As the mantle temperature increases during subduction, released water from DHS and DHMS exposed to the surrounding mantle rocks or the iron outer core may cause partial melting or the formation of new high-pressure phases (e.g., FeO_2H_x and FeH_x), which provides plausible explanations for the observed geophysical data in the deep Earth (Fig. 1).[7] The associated reactions are

$$4Fe + 2H_2O = FeO_2H_x(Py - phase) + 3FeH_x + 2(1 - x)H_2\uparrow, \quad (1)$$

$$FeO_2H_x(Py - phase) = Fe_2O_3 + xH_2O + \frac{1}{2}(1 - x)O_2\uparrow. \quad (2)$$

These processes show that hydrogen and oxygen can be derived in a step-by-step manner from water–iron reactions under CMB conditions. This promotes heterogeneity of the oxygen fugacity of the Earth's mantle.[7,9] Recent work has shown that the solidus (the temperature above which the first melt appears) of mantle rock can be decreased by increasing the oxygen fugacity.[10,11] That is, higher rock oxygen fugacity leads to easier rock melting. Thus, the dissociation of H_2O released from DHS in the lower mantle will enhance melting in the oxygenated mantle rocks. As a result, hydrous high-pressure silica provides a unique conduit for the deep Earth water cycle and polarization of oxygen fugacity of the lower mantle, promoting the geodynamics of the Earth's interior.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Yanhao Lin: Conceptualization (lead); Data curation (equal); Investigation (equal); Methodology (equal); Project administration (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **Ho-Kwang Mao**: Conceptualization (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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