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Molecular hydrogen bulk adsorption in crystalline silica: a mechanism for hydrogen transport and exchange in the Earth's interior?

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The presence of hydrogen exerts a fundamental control on geochemical reactions, redox equilibria, and the physical properties of Earth's interior. Deep hydrogen cycling has traditionally been attributed to H_2O and structurally bound hydroxyl (OH^-) in minerals, with subduction zones acting as major pathways for hydrogen transfer from shallow reservoirs to the mantle. However, recent observations of molecular hydrogen (H_2) preserved in eclogitic and mantle minerals suggest that additional mechanisms may contribute to hydrogen storage and mobility at depth.

Double-capsule piston-cylinder and multi-anvil experiments were conducted to investigate H_2 incorporation in quartz and coesite coexisting with an aqueous fluid externally buffered at ΔFMQ $\approx$ −6.0 to +0.4, at 1.5–9.6 GPa and 500–950 °C, representative conditions of a dehydrating subducting slab. Quadrupole mass spectrometry of experimental fluids revealed H_2 concentrations that, in most runs, exceeded thermodynamic predictions and blank measurements. We interpret this excess as evidence for bulk adsorption (physisorption) of H_2 within sub-nanometric structural cavities of silica polymorphs during experiments, followed by desorption during cooling and decompression to room conditions. Estimated adsorbed H_2 contents reach up to ~0.089 mol H_2 per mol SiO_2 (≈3000 μg g^{-1}), increasing with pressure and hydrogen fugacity but decreasing with temperature. The transition to stishovite, however, causes a marked decrease in adsorbed H_2.

These results were used to derive an empirical model describing H_2 bulk adsorption as a function of pressure and temperature under redox conditions relevant to dehydrating slabs. Because adsorption is governed by weak electrostatic interactions, evolving *P–T* conditions along slab trajectories and high-pressure structural densification (*i.e.*, transition toward stishovite) may promote H_2 release from crystalline silica. Such desorption could locally modify fluid compositions and redox conditions, generating transient H_2-rich fluids capable of stabilizing reduced species or triggering hydrous redox melting. These findings identify H_2 bulk adsorption in silicates as a potential mechanism for hydrogen storage and transfer in subduction environments.