

LETTERS

In-situ Raman spectra of dissolved silica species in aqueous fluids to 900 °C and 14 kbar

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ABSTRACT

The Raman spectra of fluids in equilibrium with solid quartz in the system $\text{H}_2\text{O}-\text{SiO}_2$ were measured to 900 °C and 14 kbar using an externally heated diamond cell. Dissolved silica species were identified by comparing the measured spectra with calculated normal mode frequencies and Raman intensities. At crustal pressures and temperatures, H_4SiO_4 is the dominant silica species in aqueous fluids, with a strong Raman band at 760–785 cm^{-1} . At the P - T conditions of the upper mantle, however, $\text{H}_6\text{Si}_2\text{O}_7$ dimers and possibly higher polymers coexist with H_4SiO_4 in the fluid. The most intense Raman bands of $\text{H}_6\text{Si}_2\text{O}_7$ occur at about 630, 920, and 230 cm^{-1} . The presence of high concentrations of ionized silica species in the solution can be ruled out. The observed speciation changes explain the drastic increase of silica solubility in water at lower crustal and upper mantle P - T conditions.