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Speciation and mixing behavior of silica-saturated aqueous fluid at high temperature and pressure

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Abstract

The effect of dissolved silica on the *PVT* properties of H_2O and structure of silica-saturated aqueous fluids in equilibrium with quartz in the SiO_2-H_2O system has been determined in situ with the materials at temperature (up to 800 °C) and pressure (up to 1350 MPa) in a constant-volume hydrothermal diamond anvil cell. Pressure was measured with the Raman shift of ^{13}C synthetic diamond and was also calculated from the *PVT* properties of pure H_2O. The two sets of pressures thus obtained differ by ≤50 MPa at $T < 500$ °C. At higher temperatures (and pressures), the pressure difference increases and reaches about 350 MPa at 800 °C.

The structure of the silica-saturated aqueous fluid was probed with microRaman and microFTIR methods. Coexisting molecular H_2O and OH-groups, bonded to Si^{4+}, exist above ~400 °C and ~0.4 GPa with their abundance-ratio, $OH/H_2O°$, positively correlated with temperature. Hydrogen bonding diminishes with temperature and cannot be detected in the silica-saturated aqueous fluid above ~400 °C and ~0.4 GPa. This behavior resembles that of pure H_2O under similar temperature and pressure conditions.

Speciation of dissolved silica in the fluid at 400 °C/760 MPa, and above, comprises Q^o and Q^1 species, whereas at lower temperature and pressure only Q^o species were detected. The Q^1/Q^o abundance ratio is positively correlated with temperature (and silica content). An excess volume of mixing, V_{H2O}^{excess}, derived from a comparison of EOS of pure H_2O and those of silicate-saturated H_2O, is related to the Q^1/Q^o abundance ratio in silicate-saturated fluid, X_{Q^1}/X_{Q^o}. The $V_{H2O}^{excess} = M_{H_2O}(1/d^{m}{}_{H_2O}^{-1}/0.88)$, $V_{H2O}^{excess} \sim 0$ when X_{Q^1}/X_{Q^o} approaches 0.

Keywords: Aqueous fluid, volume properties, spectroscopy, structure