Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.17424

Nanoscale heterogeneous structure and vibrational properties of hydrous silica at high pressure

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Volatile-bearing silicate melts are important agents in magmatic and metamorphic processes. The physical and thermodynamic properties of such melts depend on chemical composition, including volatile content, but also on pressure and temperature. As molecular speciation as well as volatile solubility in the melts change continuously with varying conditions, in situ measurements are needed to understand melt structure and properties at extreme conditions. In this study, we chose hydrous silica, i.e., amorphous SiO_2 containing approximately 10 % H_2O, as model system to study changes in the structure and vibrational properties with pressure. Both in situ Raman (at University of Cologne) and X-ray diffraction experiments (at beamline ID27, ESRF Grenoble) were performed in a pressure range up to 40 GPa using diamond anvil cells. The most prominent structural change in the silica network is the transition of Si coordination from 4-fold to 6-fold, which is essentially completed at the highest pressures and resembles the behavior observed in anhydrous silica. The Raman spectra indicate the presence of H_2O-rich domains on the sub-micrometer scale. With increasing pressure, the Raman bands in the region of O-H stretching vibrations change from water-like to ice-VI-like and eventually broaden significantly, which indicates a distribution of hydrous domains with different stress states in the compressed silica network. To support the interpretation of the measurements, especially the more subtle differences between the X-ray structure factors of the hydrous and the anhydrous samples, we performed ab initio molecular dynamics simulations (AIMD). The standard AIMD procedure to produce a glassy state by quenching a high temperature melt yielded structures with a dominant H_2O speciation as hydroxyl groups. Therefore, other methods, such as explicit H_2O substitution at lower temperatures, were employed to obtain structures containing sufficient molecular H_2O. Combining the different experimental and simulation results, we discuss the structural evolution of the nano-segregated hydrous silica under pressure and provide an outlook for studying the high temperature melt.

This work was supported by BMBF project 05K19PK2. Simulations were performed on the JUWELS supercomputer at Jülich Supercomputing Centre (JSC).