Influence of water on the high-pressure behavior of silica glass

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Seismic anomalies atop and below the transition zone (410-660 km) have been attributed to the occurrence of hydrous silicate melts [1,2]. Hence, a knowledge of the structural and physical properties of silicate melts and glasses (a potential structural analogue) is mandatory for the understanding of the melt processes and dynamics.

The SiO_2-H_2O mixture represents the simplest model system of a hydrous silicate melt. At ambient conditions, a phase separation into heterogeneously distributed SiO_2-rich domains that contain exclusively Si-OH groups and water-rich districts formed by molecular water takes place [3]. Already small amounts of water (1-2 wt.%) have a tremendous influence on the mechanical properties (e.g. the elasticity) of SiO_2 glass [4]. However, up to now, the influence of water on the structure of SiO_2 melt or glass is unknown, above all at high-pressure, and therefore, the origin of the seismic anomalies is still under debate.

In this study, we have investigated the influence of high-pressure on the structure of hydrous SiO_2 glass (10.4 wt.% water content) by a combination of lab-based high-pressure *in-situ* optical Raman spectroscopy up to 46 GPa and X-ray Raman scattering (XRS) measurements on the Si $L_{2,3}$ and O K edges up to 42 GPa perfomed at the beamline ID20 at the ESRF (Grenoble, France). Because the high-pressure behaviour of anhydrous SiO_2 glass was studied intensively in the past (among others by Raman spectroscopy [5] and XRS [6]), we compared our data with the previous results and used them as reference for our analysis.

The SiO_2 network of hydrous and anhydrous SiO_2 glass is affected by the pressure in a comparable way. At about 10 and 20 GPa (pressures as they occur in the transition zone), we observed significant changes in the vibrational properties of both samples in our optical Raman data. This is supported by our XRS data which, however, suggest that the Si-OH groups may hamper the transition and broaden the transition pressure by about 5 GPa compared to pure silica glass. We assume that at about 10 GPa the Si-O(H) bond lengths and angles begin to vary with increasing pressure. At around 20 GPa, our data indicate a structural phase transition from tetrahedrally coordinated Si into a mixture of 5/6-fold coordination. Moreover, at around 3 GPa, pronounced pressure-induced changes occur in the OH-modes of the molecular water. While at ambient conditions the shape of this feature is similar to that of liquid water, with increasing pressure it becomes more ice-like indicating a further structural phase transition in the water-rich districts. Based on a comparison of our data with ice spectra reported in the literature, we assume ice VII-like domains. Our results imply that the water in the structure of hydrous SiO_2 may play a key role for the melting processes in the transition zone.

[1] D. Freitas et al., Nature, Commun., 8, 2186 (2017)
[2] W. R. Panero et al., Geochem. Geophys. Geosyst., 21, e2019GC008712 (2020)
[3] E. Stopler, Contrib. Mineral. Petrol., 81, 1-17 (1982)
[4] M. Murakami, Sci. Rep., 8, 11890 (2018)
[5] R. J. Hemley et al., Phys. Rev. Lett., 57, 747-750 (1986)
[6] S. Petitgirard et al., Geochem. Persp. Let., 9, 32-37 (2019)