

Water solution mechanisms in magmas: insights from *ex situ* piston-cylinder and *in situ* hydrothermal diamond anvil cell experiments.

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The global cycle of water on Earth involves different reservoirs in the solid Earth (Earth's crust, mantle, and potentially core) and its surface (atmo-, hydro- and biospheres). Exchanges of water between them occur in specific geological settings, such as subduction zones, mid-ocean ridges or intraplate volcanisms. Each of those settings implies magmatic activity, in which silicate melts play a determining role as they carry water from depth to surface. Furthermore, magmas rheological and thermodynamic properties are strongly affected by dissolved water that, hence, influences transfers of matter and heat in the crust and in superficial volcanic systems. However, the mechanisms of water incorporation in silicate melts are still not well understood, and, in particular, their dependence to melt chemistry, temperature and pressure remain to be fully assessed.

To address those issues, we investigated alkali (Li⁺, Na⁺, K⁺) silicate, and sodium and calcium aluminosilicate melts containing various quantities of dissolved water. The melts were hydrated with a given amount of water in piston-cylinder apparatus, and studied *ex situ* after quenching using ¹H, ²H and ²⁹Si Nuclear Magnetic Resonance, Infrared and Raman spectroscopy. Furthermore, using Raman spectroscopy, we probed *in situ* melts at equilibrium with aqueous fluids in hydrothermal diamond anvil cells (HDACs). While the *ex situ* data allow us to observe the melt structure and water environment at conditions corresponding to the glass transition, *in situ* data allow us to gather knowledge at higher P-T conditions, relevant to the Earth crust and upper mantle.

Results show that the environment of water in quenched melts is deeply affected by the melt composition. For instance, in silicate melts, increasing alkali ionic radius promotes water reaction with the silicate network, while the protons reside in environments with smaller volumes. The comparison of *ex situ* data for two sodium and calcium alumino-silicate melts further substantiates this dependence to melt composition, but also suggest that aluminum might help stabilizing the H⁺ local environment. At temperatures much higher than the glass transition, Raman data from HDAC experiments show that the O-H stretching vibration, and hence the O-O distances around H⁺, becomes more similar between the sodium and calcium alumino-silicate melts.

Deconvolution and peak fitting analysis of the Raman peaks assigned to T-O stretching in TO₄ tetrahedral units in the glasses and melts further show a strong depolymerisation of the melts, which depends on melt temperature and initial composition. This complex environment of water in hydrous silicate melts induces non-linear variations of their transport properties as a function of water content, major element composition and temperature. Such knowledge can be integrated in models of magma mobility in the Earth crust, and of possible extrusive mechanisms at the Earth surface.

Keywords: water, magma, silicate melt, diamond anvil cell, NMR spectroscopy, Raman spectroscopy