Experimental report

22/03/2019

Proposal: 6-05-975 **Council:** 4/2016

Title: Pressure as a probe of the network-forming versus network-modifying role of Mg in silicate glass

Research area: Materials

This proposal is a new proposal

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Samples: MgSiO3

Instrument	Requested days	Allocated days	From	To
D4	6	3	06/07/2016	10/07/2016

Abstract:

We will use pressure as a probe of the network-forming versus network-modifying role of Mg in MgSiO3 glass, one of a family of geophysically important silicate materials that form a significant component of the Earth's mantle. Motivation is provided by the capability of Mg to act as a network former in magnesium silicate glasses, a role that stems from the ability of Mg to form structural motifs with a small Mg-O coordination number of ~4 that can fit into a silicate network with minimal disruption. With increasing pressure, however, Mg will transform into a network-modifying role as the Mg-O coordination number increases. Pressure can therefore be used as a tuneable probe of the network-forming versus network-modifying role of this additive, an issue that is important for understanding the fundamentals of modified network glass-forming materials. The changes to the network topology will be modelled by using molecular dynamics simulations with a polarisable ion model.

Pressure as a probe of the network-forming versus network-modifying role of Mg in silicate glass

The results from the experiment have been submitted as a paper entitled "Pressure induced structural transformations in amorphous $MgSiO_3$ and $CaSiO_3$."

Abstract:

The pressure-induced structural transformations in metasilicate $MSiO_3$ glass (M = Mg or Ca) on cold-compression from ambient pressure to 17.5 GPa were investigated by neutron diffraction. The structure of the glass recovered to ambient conditions from a pressure of 8.2 or 17.5 GPa was also investigated by neutron or X-ray diffraction. The experimental work was complemented by molecular dynamics simulations using a newly-developed aspherical ion model. The results show network structures based predominantly on corner-sharing tetrahedral SiO_4 units. At pressures up to ~8 GPa, there is little change to the network connectivity as described by the Q^n speciation, where *n* denotes the number of bridging oxygen (BO) atoms per SiO_4 tetrahedron. On compression of the glass to 17.5 GPa, the Mg-O coordination number increases from 4.5(1) to 6.2(1), and the Ca-O coordination number increases from 6.15(17) to 7.41(7). In both cases, the increased M-O coordination numbers are accompanied by an increased fraction of M-BO versus M-NBO connections, where NBO denotes a non-bridging oxygen atom. The results give the fraction of triple-bridging oxygen atoms as ~0.5% at 17.5 GPa, which does not support the formation of a substantial fraction of oxygen triclusters in either glass. The M-O coordination number of the recovered glass is larger than for the uncompressed material, which originates from an increased fraction of M-BO connections, and increases with the pressure from which the glass is recovered. The results suggest that the measured decrease in viscosity of molten $MSiO_3$ on pressure increasing from ambient to ~8 GPa is not related to a large change in network polymerization, but to the appearance of higher-coordinated M-centred polyhedra that contain a larger fraction of weaker M-BO bonds.