

## ***The anomalous behavior of silicate glasses and liquids on densification: A combined X-ray tomography and diffraction study at high pressure***

A.N. Clark<sup>1</sup>, G. Morard<sup>1</sup>, Y. Le Godec<sup>1</sup>, N. Guignot<sup>2</sup>, A. King<sup>2</sup>

*1 Institut de minéralogie, de physique des matériaux et de cosmochimie, Univ. Pierre et Marie Curie, CNRS UMR 7590, Paris, France*

*2 Synchrotron Soleil, CNRS UMRUR1, Gif-sur-Yvette, France*

At modest pressures (<10GPa), the main structural feature of silicate glasses and liquids is the interconnected network of SiO<sub>4</sub> tetrahedra. Depolymerized compositions (MgSiO<sub>3</sub> – Mg<sub>2</sub>SiO<sub>4</sub> pseudo binary) have anomalously high proportions of interconnected SiO<sub>4</sub> tetrahedra compared to the predictions based on stoichiometry (NBO/T calculations)<sup>1</sup>. Polymerized silicate glasses (SiO<sub>2</sub>-MgSiO<sub>3</sub> pseudo binary) exhibit anomalous volumetric and elastic properties at high pressure and room temperature<sup>2-4</sup>. The unusual behavior of polymerized silicate glasses on compression, where the compressibility increases or is weakly pressure dependent, is consistent with densification being controlled by network flexibility, rather than the compression of interatomic bonds as exhibited in their crystalline counterparts<sup>2,5</sup>. Due to the lack of long range ordering for the glassy and liquid states, the SiO<sub>4</sub> tetrahedra are not fully constrained at every vertex allowing the silicate network flexibility that is not available to the crystalline counterparts. Because of the anomalously high proportion of interconnected SiO<sub>4</sub> tetrahedra for all compositions of silicate glasses, network flexibility is likely active, however the mechanism underlying the anomalous densification is active in these systems where the silicate network is depolymerized by network modifying cations has not been studied. To this end, we will present experimental results from the newly developed RoToPEC (Rotational Tomography Paris-Edinburgh Cell) large volume press combining *in situ* X-ray diffraction and microtomography datasets at elevated pressure and temperature at ESRF and Soleil synchrotron for a suite of silicate glasses spanning the SiO<sub>2</sub>-MgSiO<sub>4</sub> binary. Simultaneous acquisition of X-ray diffraction on a large Q-range and 3D tomography (to obtain the density of the amorphous sample) were performed under extreme conditions. We will use these results to investigate the role of network flexibility and potential polymorphism in the anomalous high pressure behavior and whether the densification mechanism that underlies the anomalous behavior in the glasses extends into the liquid state.

1. Sen S. et al., *Short-Range Structure of Invert Glasses along the Pseudo-Binary Join MgSiO<sub>3</sub>- Mg<sub>2</sub>SiO<sub>4</sub>: Results from 29Si and 25Mg MAS NMR Spectroscopy*, The Journal of Physical Chemistry B, 113, 15243-15248, 2009
2. Clark, A. N., et al., *Mechanisms of anomalous compressibility of vitreous silica*, Physical Review B, 90, 174110, 2014
3. Sakamaki, T., et al., *Contrasting sound velocity and intermediate-range structural order between polymerized and depolymerized silicate glasses under pressure: Earth and Planetary Science Letters*, v. 391, no. 0, p. 288-295.
4. Sanchez-Valle, C., Bass, J. D., *Elasticity and pressure-induced structural changes in vitreous MgSiO<sub>3</sub>-enstatite to lower mantle pressures*, Earth and Planetary Science Letters, 295, 523-530, 2010
5. Walker, A. M., et al., *The origin of the compressibility anomaly in amorphous silica: a molecular dynamics study*, Journal of Physics-Condensed Matter, 19, 9, 2007