ESRF	**Experiment title:** ***In situ* X-ray tomography and diffraction of amorphous and liquid silicates under extreme conditions**	**Experiment number**: ES-394
Beamline: ID27	**Date of experiment**: from: 02/06/2016 to: 07/06/2016	**Date of report**: 13/09/2016
Shifts: 15	**Local contact(s)**: MEZOUAR, Mohamed	*Received at ESRF:*

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Report:

The aims of our 2016-1 beamtime were 1) the development and calibration of a new experimental assembly, stable at elevated pressures and temperature, under rotation in the RotoPEC, and for extended periods of time (several hours at P-T) and 2) to collect microtomography and X-ray diffraction datasets on SiO_2 glass to measure *in situ* the volume reduction and changes in atomic arrangement, respectively. These data are fundamentally important in determining the densification mechanism(s) active in silicate glasses and melts at high pressure.

Development of experimental assembly:

During our 2106-1 beamtime, we successfully developed a composite experimental PE assembly (Figure 1). This cell assembly consists of a center disc of x-ray transparent boron epoxy (BE). Top and bottom of the BE disc are ZrO_2 caps. The sample is heated using a graphite furnace with Mo foil leads. The compostie BE/ZrO_2 experimental assembly design is optimized to provide higher temperautre range, increase the accessible window between the WC anvils at high P-T conditions, and longer cell stability at elevated pressure and presures compared to the standard BE gasket (cf. (Kono et al., 2014)). The sample is contained in a single crystal sapphire capsule, which provides a non-reactive, x-ray transparent sample chamber for the silicate glasses and liquids. Furthermore, insulation between the graphite heater and boron epoxy avoid reactions and increase stability over time (Kono et al., 2014). The pressures and temperatures reached with this cell during the 2016-1 beamtime (3.5 GPa and 1100K) were mainly limited by time data collection time and beamtime allotment constraints. The stability of the cell at pressure and temperatures for extended length of time (>8 hrs) indicates that this cell would likely reach higher P-T conditions. To significantly increase the pressure range (P >8 GPa), we plan on developing a toroidal experimental assembly that will be based on the general composite gasket design developed during the 2016-1 beamtime. The main difference for the toroidal experimental assembly will be that the outer diameter of the BE disc will be increased to 14 mm and the ZrO_2 caps will consist of an inner and outer ring. The toroidal anvil design has been shown to increase the pressure range for the PE cell above 8 GPa, which is typically the highest pressure reached for liquid samples in the

PE press (e.g. (Kono et al., 2014; Morard et al., 2014; Sanloup et al., 2013). The high compressibility of glass and liquid samples makes pressure generation more difficult than in crystalline samples.

Data collection and experimental procedure:

During the 2016-1 beamtime, we collected microtomography and x-ray diffraction datasets for SiO2 glass on compression at high temperature. Using our newly developed cell assembly (Figure 1), we did not observe any signals from the single crystal sapphire sample capsule in the tomography or diffraction data. Using the RotoPEC to collect volumetric data for our samples was problematic in two ways.

1) We discovered that the RotoPEC gears that rotate the sample turn at variable rates (1:2 power input for top and bottom motors, respectively). This lead to the shearing and subsequent collapse of several experiments before the issue was remedied (only late on Sunday, 36 hours before the end of the beamtime).

2) The physical geometry of the RotoPEC is such that we were unable to use the independent Soller slits developed by Morard et al. (2011). These recently developed Soller slits cannot be positioned in the appropriate distance to the sample in the RotoPEC due to the motor/anvil connection and support-post geometry. An old version of the Soller slits have to be used in our case, leading to strong deformation of the scattering signal (Figure 2). This would be a minor correction for the strong, localized diffraction signals in crystalline materials. However, removing these artifacts in a diffuse diffraction signal is nontrival. Correcting the diffraction datasets to remove these artifacts from the diffraction data is ongoing.

To address these issues we will modify our experiments for our next beamtime as follows. 1) We will use the panoramic PE press for our experiments. We plan to combine results from complementary tomography dataset performed on the Psiche beamtline at Soleil synchrotron with the diffraction data collected using the Soller slits on ID27 at ESRF to determine the atomistic mechanisms that control densification. 2) By using the panoramic PE press, we can use the recently developed Soller slits (Morard et al., 2011; Weck et al., 2013). This will provide the high quality, low noise diffraction data that is critical for measuring the atomic structure of amorphous materials. 3) To measure independently measure density while using the panoramic PE cell, we plan use the X-ray absorption method developed (Malfait et al., 2014; van Kan Parker et al., 2010), coupling our newly developed cell assembly with sapphire capsules.

References

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Figures

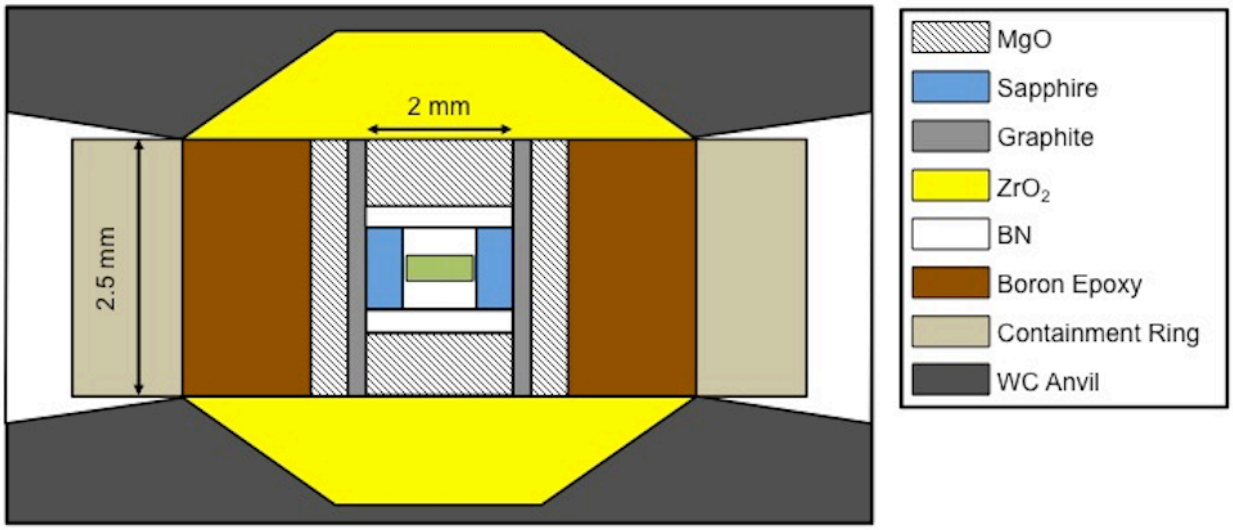

Figure 1. PE experimental assembly developed during the 2016-1 beamtime.

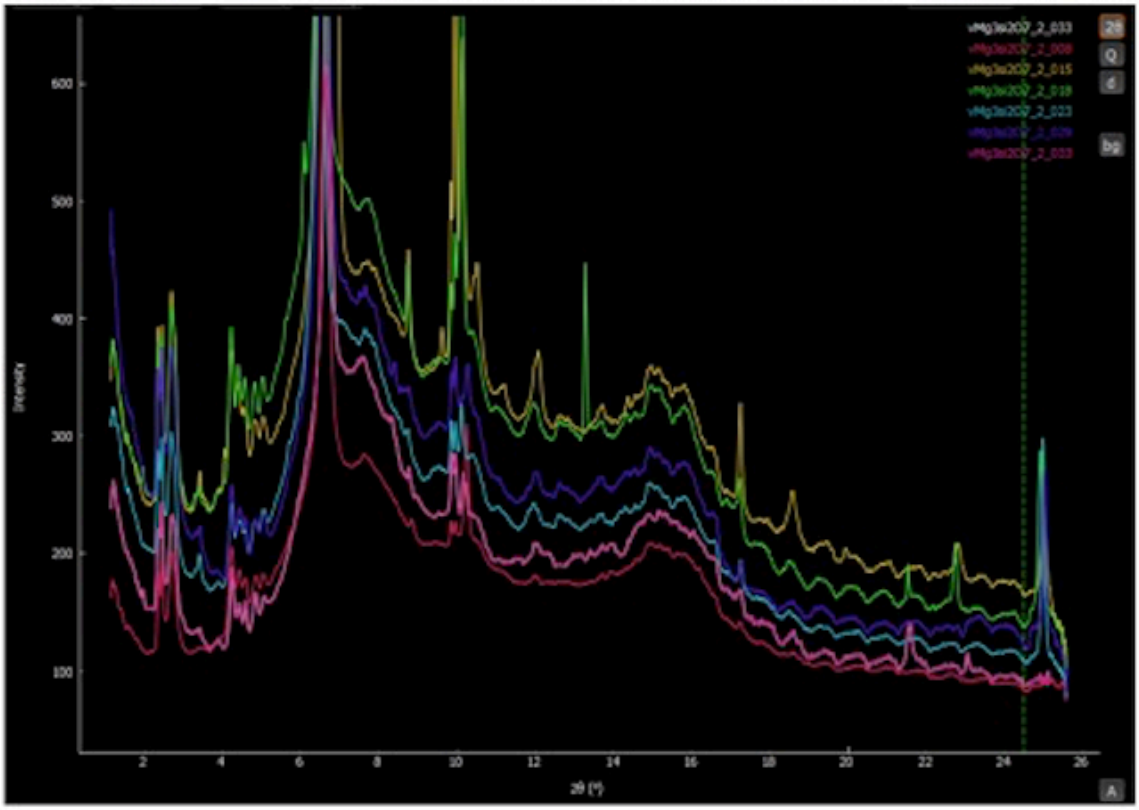

Figure 2. X-ray diffraction collected on the ID27 beamline during the 2016-1 beamtime. Data show increaseing temperature and pressure. Bottom (red) spectra: collected at 75 bars (1 GPa) and 300K. Temperautre is increased to 110 W (700K) and pressure to 145 bars (2.7 GPa). The peaks do not appear to move significantly during compression/heating. However, the edge artifacts in the spectra form the soller slits make data processing and interpretation difficult.