ESRF	**Experiment title:** Nanotomography of early mechanisms of phase separation in glasses	**Experiment number**: MA-2582
Beamline: ID16-A	**Date of experiment**: from: June 17th 2015 to: June 20th 2015	**Date of report**: 12/09/2015
Shifts: 9	**Local contact(s)**: Julio Cesar Da Silva, Yang Yang	*Received at ESRF:*

Names and affiliations of applicants (all experimentalists):

Ekaterina Burov, Jean-Thomas Fonné, Emmanuelle Gouillart, Hervé Montigaud,

Joint Unit CNRS/Saint-Gobain Surface of Glass and Interfaces, Aubervilliers, France

Damien Vandembroucq

PMMH, ESPCI, Paris, France

Background and context: using nanotomography on ID16-A, we study phase separation in barium borosilicate glasses, and in particular the morphology and topology of the two separated phases during isothermal coarsening, when the typical size of phase grows to decrease the interfacial energy [Mazurin84]. Our barium borosilicate compositions result in phases with good absorption and phase contrast (see Fig. 1), with domain sizes that can be tuned between a few nanometers to hundreds of microns, using different thermal treatments. Using in-situ microtomography on ID19, our team has characterized the evolution of the morphology of the phases during isothermal temperature treatments [HD501, Bouttes14, Bouttes15], showing in particular that topology changes could greatly affect coarsening kinetics and morphology. The aim of proposal MA-2582 is to extend this approach to smaller domain sizes, at which several physical phenomena at play (viscous flow, diffusion, capillarity) are competing.

Experiments on ID16-A: our team of 5 persons (a PhD student working on phase separation, and 4 permanent material scientists) got great help and assistance from our local contacts Yang Yang and Julio Da Silva (with the occasional and appreciated help of Peter Cloetens).
The first shift was spent on alignment and sample preparation. Acquisition parameters were chosen as follows: an energy of **33.6 keV** and voxel size of **25 nm** (with a few quick acquisitions at 50 nm), and holotomography mode, resulting in a total acquisition time of the order of **4 hours** for one sample, for the four distances.
A first batch of 4 samples was placed inside the vacuum chamber of the ID16-A chamber. These samples were 0.8-diameter cylinders that had been drilled out of larger-size pieces of phase-separated glass, following thermal treatments in the unstable temperature region.

Unfortunately, the first tomographies showed that the drilling process had tampered with the samples, and that samples had partially crystallized, resulting in a very different morphology than what we wanted to observe. This problem had never occurred before for the larger (2-mm diameter) samples used for the in-situ microtomography, probably because of a larger drilling speed for smaller samples. As a consequence, we had to prepare new samples at the beginning of the second day: using larger glass samples similar to the ones that had been drilled, we could wear away small parts to form rounded tips of diameter between 0.5 and 1 mm. These new samples were found to have a morphology similar to what had been observed in SEM imaging before. We could image **4 different satisfying samples**, because of the bad samples of the first day and of two beam losses and an air conditioning problem during the last day.

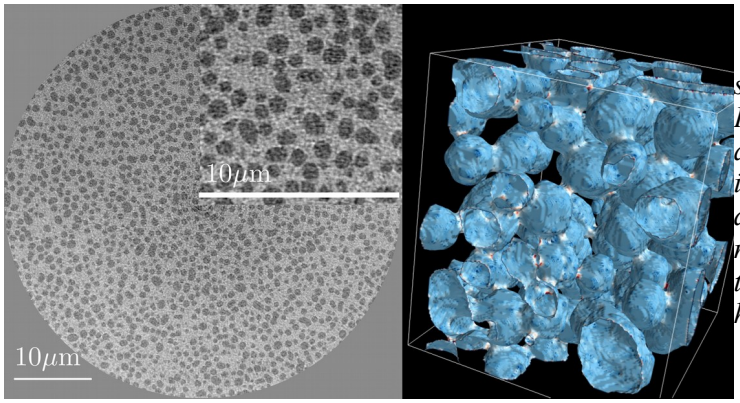

Fig. 2*: (Left) Phase reconstruction of phase-separated barium- borosilicate glass observed on ID16-A (pixel size 25 nm). Inset: silica-rich droplets are well contrasted, yet important noise corrupts the image. (Right) 3-D view of the droplets phase. Most droplets are connected to a percolating phase. Large necks are clearly identified, corresponding to droplets that are coalescing, however the thinnest bridges are hypothetical, because of possible segmentation errors.*

Data processing and expected results: after a training given by the ID16-A team, we were able to reconstruct the 3-D data (with responsive help by e-mail from our local contacts, which was very appreciated). We have started working on the segmentation of the two phases, using Python's scikit-image (our team contributing to the development of this tool). Although it is quite easy to segment the two phases, a heavy denoising step must be performed first, because of the important structured noise due to local tomography (our samples were more than 10x times larger than the reconstructed volume). Therefore, the effective resolution is not as good as the 25-nm pixel size. We expect that new experiments with smaller samples (say 0.1 or 0.2 mm diameter) could improve the quality of the images and the effective resolution.
The four volumes that we are working on will allow us to investigate
- the effect of the volume fraction, since we imaged three samples with a different volume fraction of the two phases, all heated for 2 hours at 950°C. One of them is shown in Fig. 1, demonstrating the importance of 3-D imaging: although droplet-like gray domains appear quite spherical, most belong to a percolating phase, because of the coalescence of droplets.
- the evolution of the morphology with coarsening time, since we have 2 images of the same composition after 2 and 4 hours at 950°C. In particular, we will test the invariance of the morphology with time, that we tested at larger scales using microtomography [Bouttes14].
We first need to finish to process the results, but we plan to integrate these results to a publication showing the variety of morphologies that can be observed in such glasses. This publication will present both SEM and nanotomography results. We will apply for more beamtime on ID16-A, in order to compare a larger number of samples and to gain a deeper understanding on topology changes such as the observed coalescence of droplets.

References

[Mazurin84] O. Mazurin and E. Porai-Koshits, Phase Separation in Glass (1984).
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[Bouttes14] D. Bouttes et al, Phys. Rev. Lett. **112**, 245701 (2014). ESRF14BO414
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