ESRF	**Experiment title:** Time resolved structural study of the glass transition in calcium aluminates and aluminosilicates;	**Experiment number**: HD-70
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Report:

Since many years the phenomena of supercooling and glass formation is the subject of large number of works. For a better understanding of the liquid structure and its behaviour during the glass transition, it is necessary to study the structure at the atomic scale in both stable liquid and solid states as well as during the glass transition, i.e. in a dynamical regime.

The aim of the project was to study the glass transition of liquid aluminates and calcium alumino silicates by making time resolved diffraction measurements from the liquid state to room temperature. Experiments have been carried out at the ID11 beamline in February 2007 and we present here some results.

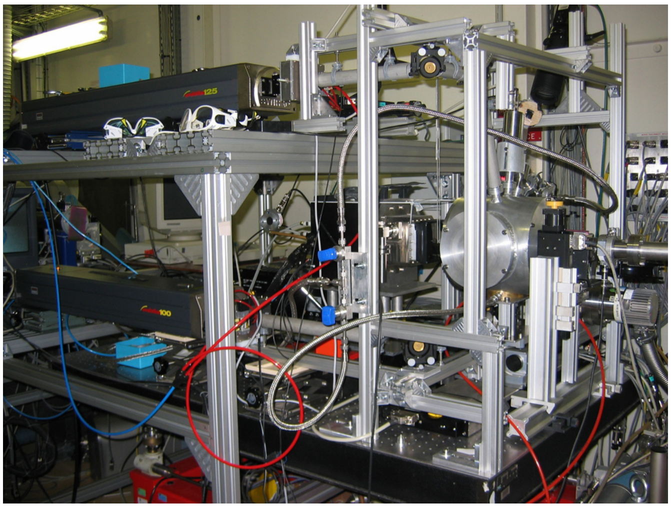

Figure 1: Levitation setup at the ID11 beamline

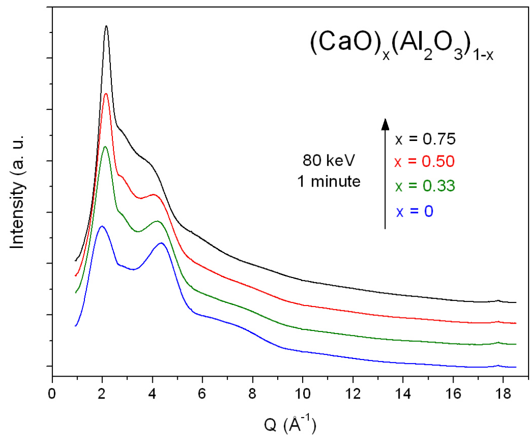

Figure 2: Scattered intensity from liquid Calcium aluminates with various compositions. The scattered intensity was measured at an energy of 80 keV during one minute using the ID11 FRELON detector

For this experiment, we used high energy x-rays (80 keV) and the fast Frelon CCD detector. The experimental setup is shown in figure 1. In order to avoid any problem with containers, the measurements have been performed using a containerless environment. Spherical samples with diameters between 2.5 and 3mm were aerodynamically levitated using an argon/oxygen gas jet and melted with two CO_2 lasers heating the sample from the top and from the bottom in order to access a deep supercooling. The laser beams are focused on the sample using a set of spherical copper mirrors. The temperature is measured using an optical pyrometer. The setup is entirely remotely controlled with a computer.

The 10cm detector was placed close to the chamber in order to have a maximal Q value about 19 Å^{-1}. In figure 2 we show the scattered intensity from various calcium aluminate compounds $(CaO)_x(Al2O3)_{1-x}$ with x in the 0-0.75 composition range. This curve shows an evident structural evolution with the CaO content. One can see the excelent signal/noise ratio even at large Q. Additional neutron diffraction have been recently performed at ILL and will be combined with the x-ray data.

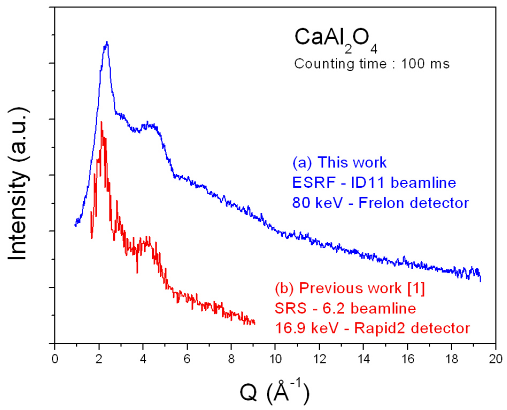

Figure 3: Scattered intensity from liquid $CaAl_2O_4$ measured with a counting time 100 ms. (a) using an energy of 80 keV and the ID11 FRELON detector. (b) using an energy of 16.9 keV and the SRS RAPID2 detector.

Time-resolved x-ray measurements require short counting times with good statistics over a wide range of scattering vector *Q*. By using 100ms acquisitions, we studied recently at the 6.2 beamline (SRS), the structural evolution of molten $CaAl_2O_4$ as it cooled from the stable liquid phase well above the melting point Tm (1878K) to the cold glass below Tg (1173K)[1]. The evolution is characterized by a sharpening of the first diffraction peak and a shortening of the average nearest-neighbour bond length at about 1467 K, indicating an increase in the degree of both intermediate-range and short-range order occurring close to the crossover temperature Tc (~1.2Tg).

Nevertheless, these measurements were performed using a 60° curved detector which gave a limited maximal Q value of 9. Å^{-1} (see figure 3 and consequently, a low resolution in r-space

Compared to our previous study, the use of the Frelon detector makes it possible to double the Q range and consequently the r-space resolution. The statistics are also very good over the full Q range. This has enabled good measurements with even with shorter counting times (30ms).

The data treatment is in progress and takes some time due to the number of files to analyze (120 files for a cooling with 100ms measurements), but the first results are very promising.

This experiment was the first application of the levitation setup at the ID11 beamline and various aspects can be optimised in order to get better results.

[1] L. Hennet et al, J. Chem. Phys. 126 074906 (2007)