ESRF	**Experiment title:** In situ study of the structure and nucleation of strontium aluminosilicate melts.	**Experiment number**: HD-517
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Report:

Aim of the experiment.

Nucleation is the step between amorphous and crystalline phases and represents one of the most important effects taking place in earth or material science (magmatic processes, phase differentiation and segregation...) and in various industrial processes (ceramics, vitroceramics…). However, nucleation processes are still not very well understood.

The relation between atomic structure of supercooled liquids and the ability to nucleate (in bulk or at the surface) is therefore of major importance to understand the first stages of nucleation. In particular, from a fundamental point of view, it is when important structural rearrangements occur. From a technological point of view, it is also during this stage that appears the possibility to control the growth of specific crystals, enabling the synthesis of nanocomposite glass-ceramics of new generation.

X-ray diffraction (XRD) with the possibility of performing time resolved analysis, is a very useful tool for the study of nucleation processes, before and during the the appearance of the first crystal. ID11 is particularly well adapted for such experiments. We have chosen to work with the SrO-Al_2O_3-SiO_2 system for several reasons: -

- This system gives one of the best ceramic material: the celsianite $SrAl_2Si_2O_8$ is one of the hardest ceramic and is readily synthesised from the glass and glass-ceramic process;
- This system nucleates and crystallizes easily without nucleating agent;
- The absence of nucleating agent implies that all the Sr participates to the nucleation process. This can give interesting and visible structural information on the melts before and during nucleation.

Experimental details.

During this experiment, 2 high temperature environments have been used :
1/ Aerodynamic levitation combined with laser heating [1] for studying the liquid and supercooled states [2].
2/ Micro-heating furnace developed by Neuville [3] which can be used from ambient up to temperatures above 2000°C and in particular in the glass transition domain where nucleation process can occur.

We worked at an energy of 100 keV and we used the Frelon CCD camera placed at a distance of 200 mm from the sample enabling measurements over a wide Q-range (up to 25 $Å^{-1}$).

Results.

The figure below shows the structure pair distribution function obtained with the composition SA33.33 (SrAl2SiO6).

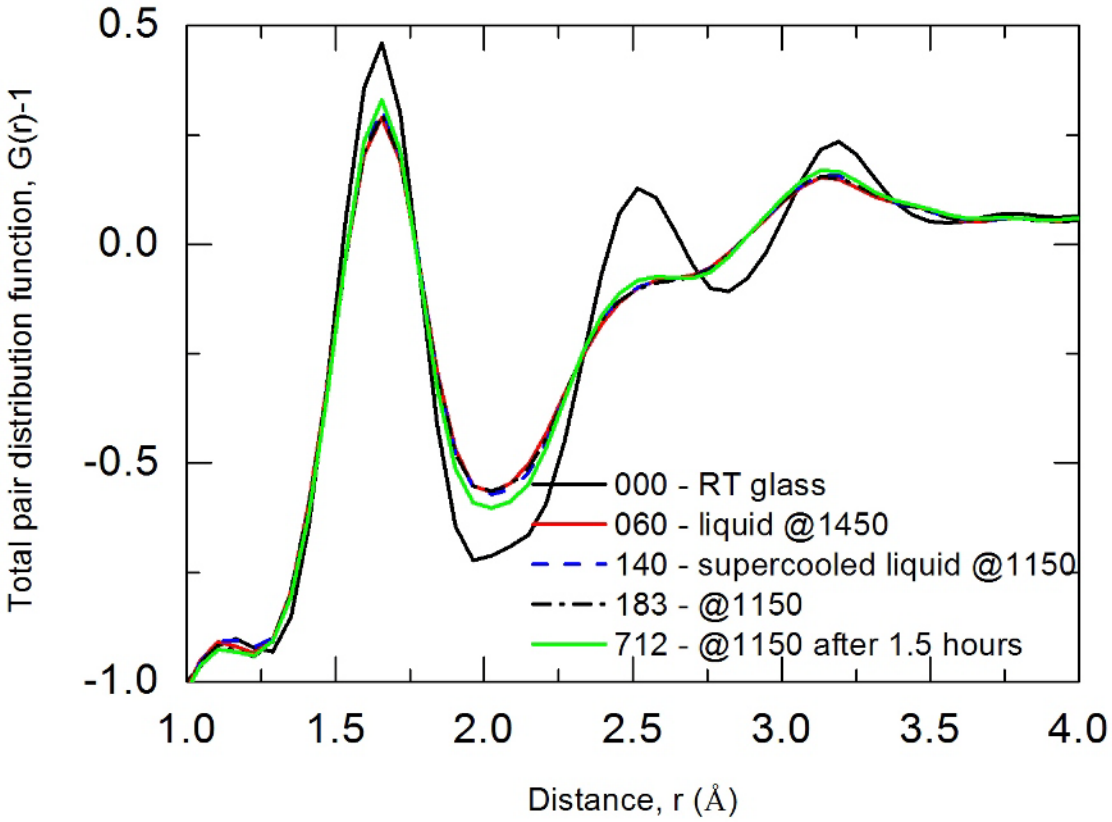
Measurement in the liquid state at 1450 and 1150°C have been performed using the levitation setup. The measurements on the the hot glass (supercooled) at 1150°C at t=0 and after 90 min have been perfomed using the furnace. The measurement performed on the glass at room temperature is also presented.

As expected, the liquid structure in the normal and supercooled states is very similar and the two measurements performed at 1150°C using the two heating device are identical.

The main differences are observable on the measurements performed at 1150°C after 90 min when the liquid starts to nucleate.

The signal is modified and the evolution goes in the direction of the glass curve: Higher maxima and deeper mimina, corresponding to a more ordered stucture.

With time the signal continues to evolve with increasing structural ordering and finally the appearance and growth of Bragg peaks in the structure factor corresponding to the crystallisation.



The data treatment is still ongoing. These XRD measurements as a function of time and temperature in the annealed melt before and during nucleation processes will give important parameters to understand structural rearrangement during crystallization processes and will be completed by x-ray absorption spectroscopy at the Sr K-edge at high temperature that will give information on the short range order evolution about Sr atoms.

Reférences.

[1] L.Hennet et al *J. Non-Cryst. Sol,* 354 5104–5107 (2008)
[2] L. Hennet et al, J. Phys.: Condens. Matter 19, 455210 (2007)
[3] D.R. Neuville et al, American Mineralogist, 93, 228-234. (2008)