Experimental Report

Proposal: **6-05-919** **Council:** 10/2012

Title: Structural heterogeneity in sodiumsilicate glasses near the glass transition.

This proposal is a new proposal

Researh Area: Materials

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Samples: Na2Si3O7

Instrument	Req. Days	All. Days	From	To
D16	5	3	02/04/2013	05/04/2013
D33	5	3	08/03/2013	11/03/2013

Abstract:

The aim of this proposal is to use Small Angle Neutron Scattering (SANS) to get information on the structural heterogeneities that would influence the dynamics in our sodium silicate glasses at high temperature.
The measurement of the SANS signal up to a Q value including the first sharp diffraction peak will give us information at short, medium and long distances.
We will get valuable data on the dynamics of structural relaxation that will help us to understand the observation made by Brillouin Light Scattering.

-I- Introduction

Understanding the nature of the glass transition has been a subject of interest for over half a century, especially the origin of the slowing of the dynamics during supercooling. Various theories for the glass transition focus on microscopic dynamical mechanisms. The underlying concept of many of these theories is the Adam and Gibbs hypothesis, which states that the flow in a supercooled liquid involves cooperative motion of molecules and that the structural arrest at the glass transition is due to a divergence of size of cooperating regions.

In this work, we studied the glass composition $Na_2Si_3O_7$ (Tg ~ 710K). From Brillouin Light Scattering (BLS) observations aimed to study the relaxation dynamics below, across and above the glass-transition, we have shown that the aging dynamics are heterogeneous. In particular, we observed two distinct time scales including a fast and a slow component in this temperature range

The aim of this proposal was to use Small Angle Neutron Scattering (SANS) to get information on the structural heterogeneities that would influence the dynamics and that would explain our BLS observations. In order to have a wide range of Q-values, measurements have been performed at the two instruments D16 and D33. The high temperatures were achieved by using the ILL furnaces (Figure 1 presents the setup used at D16).

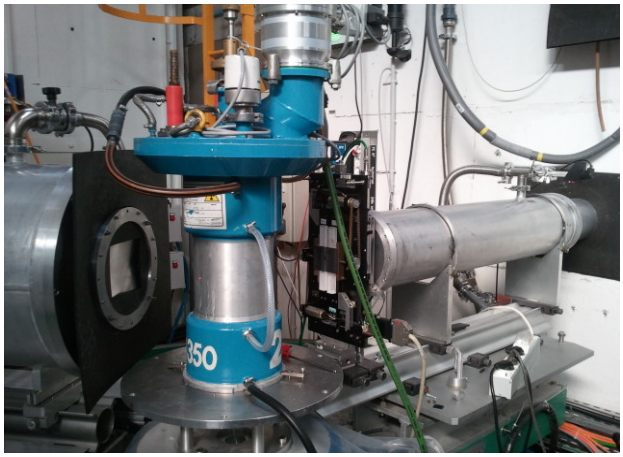

Fig1. Experimental setup at D16.

-II- Experiment at D16

The experiment was performed at a wavelength of 4.7Å at several positions of the detector up to 95°, giving a usable Q-range of 0.01-2.15 $Å^{-1}$.

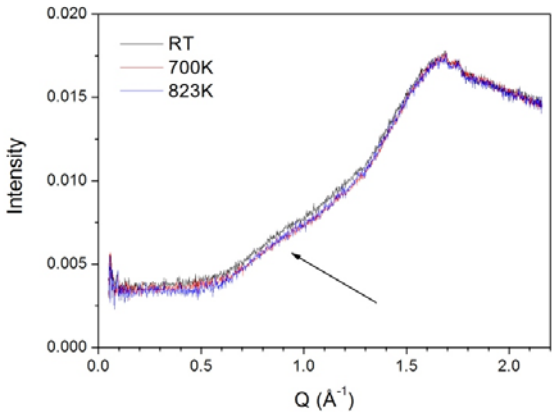

Fig.2. SANS signal obtained at several temperatures.

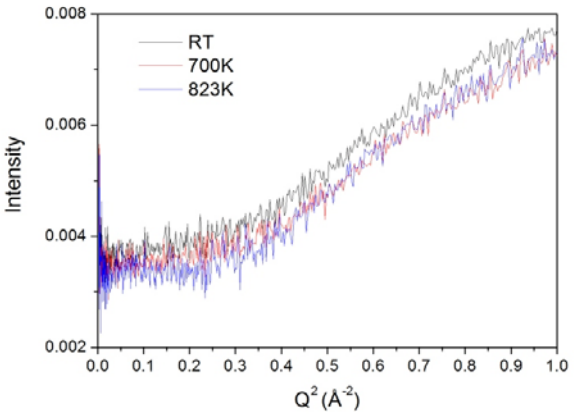

Fig. 3. Expanded region as a function of Q^2.

Fig 2 shows the intensity measured at several temperatures (RT, 700 and 823 K). All curved are very similar except an evolution of the medium range order characterized by a decrease of the intensity of the shoulder on the low-Q side of first peak (Fig 3). This Q value, about 1 $Å^{-1}$, corresponds to distances of about 6 Å in the r-space. A variation of the slope at small Q is also observed in Figure 3, at Q^2 values around 0.3 $Å^{-2}$.

-III- Experiment at D33

In order to study the low Q-region, we performed complementary measurements at the D33 instruments.
We used the TOF mode with the 2 groups of detectors at distances of 2m and 5.3m? The aperture was 8mm.
We used a furnace from the ILL, almost identical to the one used at D16.

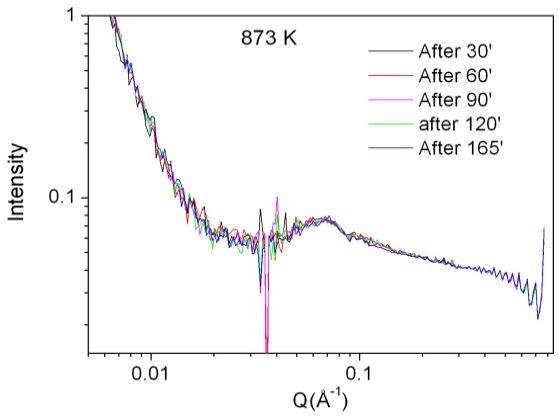

Fig.4. SANS signal obtained during the sample annealing at 873K after several durations.

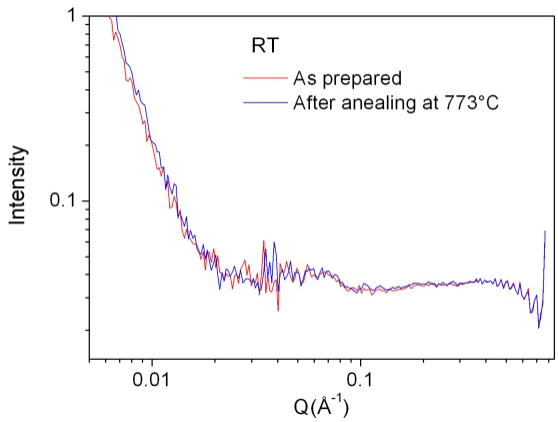

Fig. 5. SANS signal measured before and after annealing the sample at 773K.

Fig 4 shows the intensity measured during the sample annealing at 873K after several durations. At this temperature above Tg, the relaxation is supposed to occur rapidly but we didn't observe any evolution of the signal as a function of time. Fig 5 shows the measurements performed at room temperature recorded before and after the sample annealing at 773K. This temperature is also above tg and as previously, we didn't observed any evolution.
This seems to say that the signal expected from the heterogeneities is too week to be evidenced using neutron scattering.

-IV- Conclusion

The objectives of these experiments were to study the evolution of the medium range (MRO) and the structural relaxation dynamics during the annealing of a sodium silicate glass. If an evolution of the MRO is observed, it seems difficult to evidence any evolution of the dynamics at small angles. A comparison with data recently acquired with x-rays at small angles at ESRF will help to better understand what we observed during this neutron experiment.