ESRF	**Experiment title:** Evolution of the fragility in Calcium aluminosilicate melts	**Experiment number**: HD-494
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Report:

Introduction

This study is a continuation of the experiments HD-190 and HD-395 during which we studied the liquid glass former CaAl2O4 above and below the melting point at temperatures around 1.6Tg and 1.4Tg. The scientific purpose of this work is to use the IXS technique for studying the fragility of liquid calcium aluminosilicate (CAS) glasses: $(CaAl_2O_4)_{1-x}(SiO_2)_x$ as a function of the silica content x.

In particular, contrary to SiO_2 which is a strong glass former, melts in the $CaO\text{-}Al_2O_3$ system are extremely fragile. Consequently the fragility of CAS melts decreases with increasing the SiO_2 content and the longitudinal viscosity that can be derived from a memory function fit to the scattering function $S(Q,\omega)$ is expected to increase.

Experiment

The IXS experiments were performed on the high resolution spectrometer at the ID16 beamline under containerless conditions using the levitation apparatus developed by the CEMHTI. We worked with an incident energy of 23.725 keV using the reflection Si (12,12,12). The global resolution was about 1.3 meV. The high temperature setup and the data treatment are described in details in ref [1]. The configuration of the spectrometer makes it possible to measurement the scattered beam at 9 equidistant *Q*-values (*Q*-set). Different *Q*-sets are obtained by rotating the detection arm.

Since our samples required a relatively long acquisition time, we performed the measurements at only one *Q*-set (Q_1 = 1, 2.6, 4.3, 5.8 nm^{-1}…) that was sufficient to provide a good interpretation of the data. It can be noted that for *Q* values above 5.8 nm^{-1}, the signal was too damped and Brillouin peaks were difficult to distinguish.

We studied 3 compositions CASx.y (12.44, 19.40 and 33.33) where x is the silica content and y~(100-x)/2 corresponds to the Al_2O_3 and CaO contents (equal). The IXS spectra measured at 2073 K are shown in Fig. 1. Considering the results obtained earlier for liquid alumina [2], a model involving two relaxation times as

described in ref [1] was used to model the experimental data and we obtained a good agreement between the experimental and calculated spectra (Fig1).

In this model, thermal fluctuations are neglected and the density fluctuations are described by a memory function given by the sum of two contributions: a slow component following a Debye law (exponential decay) and a fast component that is effectively instantaneous:

$$M(Q,t) = \Delta_\alpha{}^2(Q)e^{-t/\tau_\alpha(Q)} + 2\Gamma_S(Q)\delta(t) \qquad (1)$$

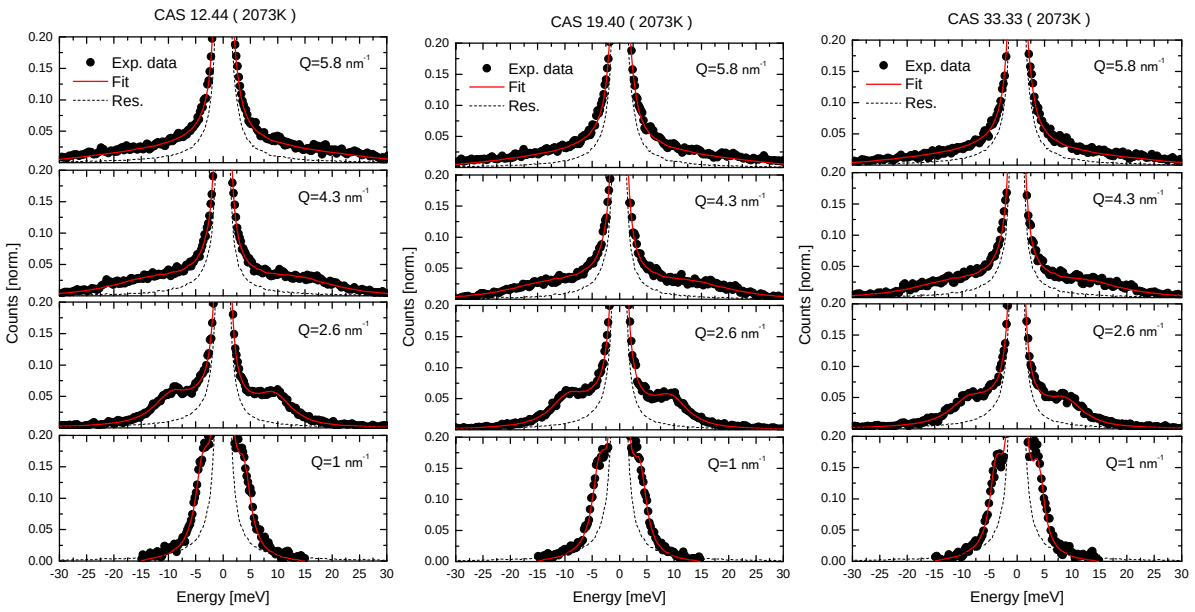

Fig. 1. IXS spectra of the 3 liquid CAS at 2073K and fit using eq. (1).

The longitudinal viscosity is calculated using the parameters derived from the fit using Eq. (1) to the experimental data and using the eqution $\eta_l(Q) = \rho(\Delta_\alpha^2\tau_\alpha + \Gamma_S)/Q^2$.

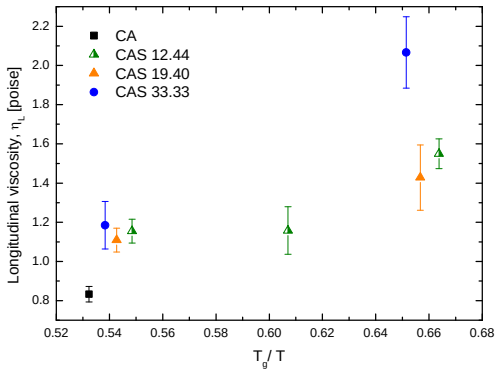

Fig. 2. Angell representation of the longitudinal viscosity for all samples studied.

Fig. 2 shows an Angell representation of the obtained viscosities where η_l is plotted as a function $1/T$ and normalized to the corresponding glass transition temperature T_g for each composition. One can observe an increase of the viscosity when the temperature decrease or when the silica content increase. It is also important to point that the glass with the highest silica content CAS33.33 shows a large deviation from the other two CAS compositions indicating a pronounced decrease of its fragility. Then, more effect should be obtained by increasing the silica content in the glass that could be the purpose of a further proposal.

Further analysis

To go further in the analysis, we also performed Quasi-Elastic Neutron scattering experiments on the same compositions and Molecular Dynamics simulations are in progress.

Reference

[1] I. Pozdnyakova, L. Hennet, J. Brun, D. Zanghi, S. Brassamin, V. Cristiglio, D. Price, F. Albergamo, A. Bytchkov, S. Jahn, and M. Saboungi, J. Chem. Phys. 126, 114505 (2007).

[2] H. Sinn, B. Glorieux, L. Hennet, A. Alatas, M. Hu, E. E. Alp, F. J. Bermejo, D. L. Price, and M.-L. Saboungi, Science 299, 2047 (2003)