Aberystwyth University

In-situ X-ray structure measurements on aerodynamically levitated high temperature liquids

Weber, Richard; Benmore, Christopher; Mei, Qiang; Wilding, Martin

Published in:
Synchrotron Radiation in Materials Science

DOI:
10.1063/1.3086241

Publication date:
2009

Citation for published version (APA):
Weber, R., Benmore, C., Mei, Q., & Wilding, M. (2009). In-situ X-ray structure measurements on aerodynamically levitated high temperature liquids. In R. Magalhaes Paniago (Ed.), *Synchrotron Radiation in Materials Science: Proceedings of the 6th International Conference on Synchrotron Radiation in Materials Science* (Vol. 1092, pp. 79-83). (AIP Conference Proceedings; Vol. 1092). American Institute of Physics Publishing. https://doi.org/10.1063/1.3086241

In-situ X-ray structure measurements on aerodynamically levitated high temperature liquids.

Richard Weber*[1,2], Christopher Benmore[2], Qiang Mei[2] and Martin Wilding[3]

1. Materials Development, Inc., Arlington Hts., IL 60004, USA, rweber@matsdev.com
2. Argonne National Laboratory, benmore@anl.gov, qiang.mei@hpcat.aps.anl.gov,
3. University of Wales, mbw@aber.edu

Abstract. High energy, high flux X-ray sources enable new measurements of liquid and amorphous materials in extreme conditions. Aerodynamic levitation in combination with laser beam heating can be used to access high purity and non-equilibrium liquids at temperatures up to 3000 K. In this work, a small aerodynamic levitator was integrated with high energy beamline 11 ID-C at the Advanced Photon Source. Scattered X-rays were detected with a Mar345 image plate. The experiments investigated a series of binary in the CaO-Al_2O_3, MgO-SiO_2, SiO_2-Al_2O_3 metal oxide compositions and pure SiO_2. The results show that the liquids exhibit large changes in structure when the predominant network former is diluted. Measurements on glasses with the same compositions as the liquids suggest that significant structural rearrangement consistent with a fragile-strong transition occurs in these reluctant glass forming liquids as they vitrify.

Keywords: Materials Science, Synchrotron radiation
PACS: 81.00.00

INTRODUCTION

Extreme sample environments provide unique opportunities when they are used in combination with high flux x-ray sources. The use of aerodynamic levitation with laser beam heating provides a convenient way to access very high temperature liquids in conditions that avoid contamination and heterogeneous nucleation by container walls [1,2]. This enables (i) the study of metastable high temperature liquids, and (ii) formation of new glasses. The aerodynamic levitation technique is extremely versatile and compact and it has been used to study liquid metals, oxides and integrated with NMR, neutron, and x-ray facilities [3,4].

The present research is focused on investigation of fragile binary oxide liquids. These liquids exhibit a highly non-Arrhenius temperature dependence of viscosity [5]. In many cases fragile liquids can be vitrified using levitation methods. The study of

CP1092, *Synchrotron Radiation in Materials Science: 6th International Conference,* edited by R. Magalhaes-Paniago

the high temperature liquid and the corresponding glass structure provides insights in structural evolution as the liquid cools, its viscosity increases, and ultimately the glass transition occurs.

EXPERIMENTAL METHODS

A photograph of the laboratory-based aerodynamic levitation facility at the Advanced Photon Source is shown in Fig. 1. This instrument is used in the laboratory to synthesize glasses and investigate undercooling of liquids. The instrument can be installed at the high energy beamline 11 ID-C for *in-situ* measurements on liquids. The sample temperature is measured using an optical pyrometer and progress of the experiments is followed from outside the beamline hutch using video cameras that view the sample. Samples approximately 3 mm in diameter can be levitated in process gases (oxygen or argon, which is necessary for metals).

In the x-ray experiments, scattered x-rays are detected using a Mar-345 image plate that is located approximately 50 cm from the sample. The x-ray beam is 1 x 1 mm and intersects the top part of the sample in the region where it is heated and where the temperature is measured. The instrument is calibrated using a standard CeO_2 sample and then the materials to be investigated are introduced into the levitator.

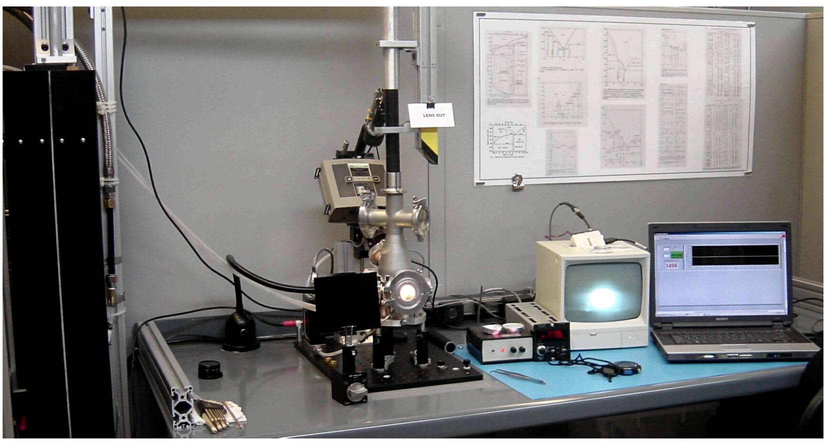

Figure 1. Image of the aerodynamic levitation facility at the Advanced Photon Source. Binary oxide compositions are made by fusing high purity metal oxide powder mixtures.

A schematic layout of the experimental set up is illustrated in Fig. 2. A tungsten pin loc ated at the center of the image plate blocks the direct x-ray beam. Scattered x-ray data is acquired by computer and analyzed using procedures established in prior work [6].

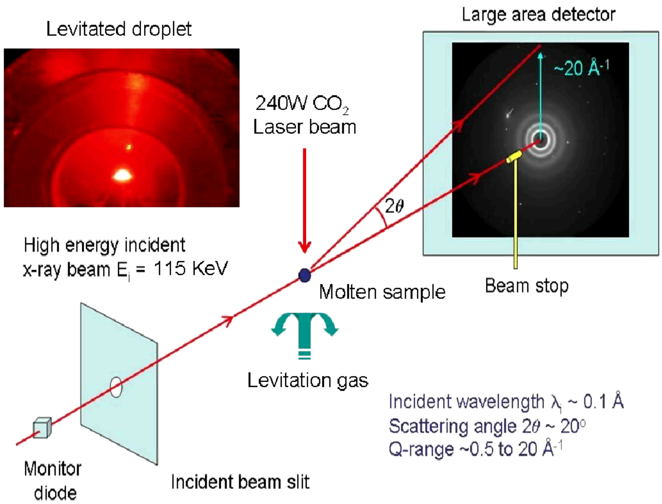

Figure 2 - Illustration of the experimental set up at the beamline. By using an incident x-ray energy of 115 keV a Q-range of *ca*. 20Å^{-1} is obtained

Silica. High purity silica glass and the corresponding liquid were investigated at temperatures from 25-2100°C [6}. The strong liquid exhibits very minor structure changes with temperature: (i) ~2% increase in Si-O bond length from 25 to 1600°C, and (ii) the average bond angle decreases ~9° at high temperature, indicating small changes in polymer ring size

CaO-Al$_2$O$_3$ binaries. The materials melt to form fragile liquids that can be vitrified over a range from approximately 50-70 mole % CaO. The derived D(r) for liquids at 2000°C and glass at ambient temperature are presented in Fig. 3.

While the Al-O first nearest neighbor distance is essentially the same in the glass and liquid, there are significant changes in the Ca-O bonding and the correlations at longer distances. NMR measurements on similar glasses indicated that AlO_4 triclusters are present in compositions containing 50% CaO but not in those with higher CaO contents [8,9]. MD simulations on the liquid [10] indicate a substantial concentration of triclusters in the liquid.

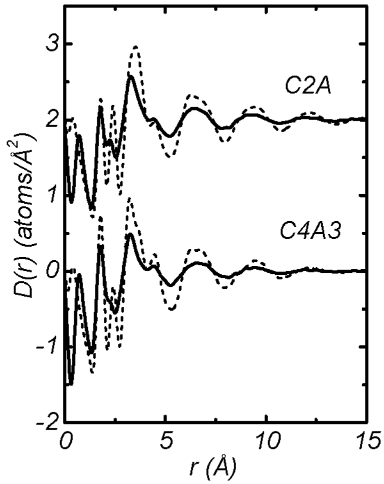

Figure 3. Comparison of the structures of liquid and glass for two compositions in the CaO-Al_2O_3 binary system shown out to long distances, glass is the dashed line – see ref [7].

MgO-SiO_2 binaries. Detailed structure measurements across the $MgSiO_3$-Mg_2SiO_4 region show that there is a transition from predominantly SiO_2 network to MgO network at approximately 38 mole % SiO_2 in the glass. Measurements on the corresponding high temperature liquids suggest that the transition in network behavior occurs at approximately 48 mole % SiO_2 in the liquid.

Al_2O_3-SiO_2 binaries. Measurements were made on liquids containing 30-90 mole% Al_2O_3. The melt structure is remarkably constant over this range comprising predominantly 4-coordinated Si-O and Al-O species. Simulations indicate a high population of AlO_4 triclusters in the liquids. In contrast, glasses formed in this system show high populations of 5 and 6-coordinated aluminum ions [11].

DISCUSSION

For the three binary oxide systems investigated, there are substantial differences in structure between the glass at ambient temperature and the high temperature liquid. In the case of the aluminates, the presence of triclusters of AlO_4 species appears to be common in the high temperature liquids. On cooling, the concentration of triclusters decreases and the glasses frequently contain higher coordinated aluminum species. In the magnesium silicates, changes in Mg-O bonding occur as the liquid is cooled.

The high and low temperature structural 'snapshots" enabled by the high energy x-ray experiments provide a new insight into the structural evolution of the liquids with temperature. Ongoing research is targeted towards understanding the temperature dependence of structure as the liquids approach the glass transition. One key goal of this work is to determine if the fragile-strong transition exhibits the character of a phase transition or a slow evolution of structure with changing temperature.

ACKNOWLEDGMENTS

This work was supported by the U.S. DOE, at Argonne National Laboratory under contract number DE-AC02-06CH11357.

REFERENCES

1. S. Ansell. S. Krishnan, J.K.R. Weber, J.J. Felten, P.C. Nordine, M.A. Beno, D.L. Price and M-L Saboungi, P*hys. Rev. Lett.*, 1997, 78, 464.
2. J. J. Wall, R. Weber, J. Kim, P. K. Liaw, and H. Choo, *Mater. Sci. Eng. A*, 2007, 445-446, 219.
3. C. Landron, L. Hennet, J.-P. Coutures, T.E. Jenkins, C. Aletru, G.N. Greaves, A.K. Soper and G. Derbyshire, *Rev. Sci. Instrum.*, 2000, 71, 1745.
4. J.K.R. Weber, C.J. Benmore, J.A. Tangeman, J. Siewenie and K.J. Hiera, *J. Neutron Res.*, 2003, 11, 113.
5. F. H. Stillinger, *Science,* 1995, 267, 1935..
6. Q. Mei, C.J. Benmore and J.K.R. Weber, *Phys. Rev. Lett,* 2007, 98, 057802.
7. Q. Mei, C.J. Benmore, J. Kim, J.K.R. Weber, J.E. Rix and M.C. Wilding, *J. Phys. Cond. Matt.*, in press.
8. D. Iuga, C. Morais, Z. Gan, D.R. Neuville, L. Cormier, and D. Massiot,, *J. Am. Chem. Soc*. 2005, 127, 11540.
9. J.R. Allwardt, S.K. Lee and J.F. Stebbins, *Am. Miner.*, 2003, 88, 949.
10. S. Kohara, Spring-8, private communication.
11. S. Sen, R.E. Youngman. *J. Phys. Chem. B,* 2004 108, 7557.