Structure and Polyamorphism in Alumina-based Glasses

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Introduction

Polyamorphic phase transitions, where liquid or amorphous phases restructure to a more stable configuration via a first-order process, occur in a variety of systems. Evidence of such transitions has been seen in water, pure elemental phosphorus, SiO_2, triphenyl phosphite, and binary aluminum-yttrium (AY) oxide liquids [1, 2]. Computer simulations support the idea that either of two liquid states with different densities and the same composition can be stabilized through changes in pressure or temperature. Binary AY oxide compositions form ionic liquids that can be quenched to form two-phase glass mixtures with identical compositions by arresting a liquid-liquid transition that occurs as the glass transition is approached. The supercooled $Y_3Al_5O_{12}$ (YAG) composition liquid contains mainly 4-coordinate Al^{3+} and 6-coordinate Y^{3+} ions, and conditions that favor the L-L transition tend to increase the Al^{3+} coordination.

A structural investigation of the liquid-liquid phase transition AY oxides has been performed through a detailed study of their associated glasses (see Fig. 1).

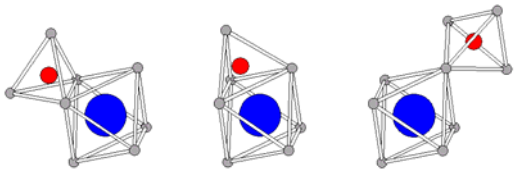

FIG. 1. This schematic diagram represents typical local structure arrangements of the Y or La and Al ions. It shows some of the various connectivity arrangements that can occur in the aluminum-rare-earth glass systems.

Methods and Materials

AY and aluminum-lanthanum materials were prepared by fusing and quenching levitated mixtures of high-purity metal oxides into about 3.5-mm spheroids in a laser beam. In this study, we report on a combination of x-ray, neutron, and simulation methods used to describe the structure of the two-phase YAG glass.

Results

The high-energy x-ray data taken on the Basic Energy Sciences Synchrotron Radiation Center Collaborative Access Team (BESSRC-CAT) 11-ID-C beamline were analyzed by using the ISOMer-X software package and are shown in Fig. 2.

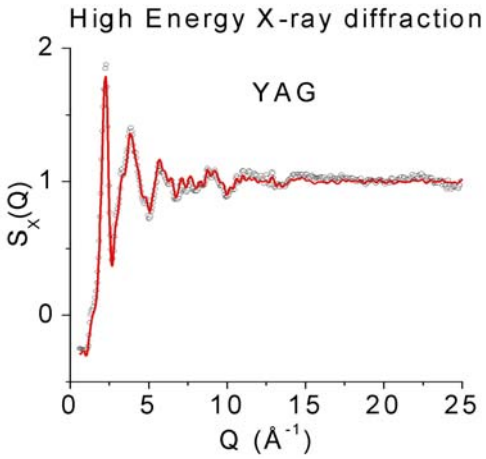

FIG. 2. The measured x-ray structure factor for laser-quenched glassy YAG. This sample is composed of two phases. The circles represent the measured data, and the red line is a reverse Monte Carlo model fit simultaneously to the x-ray data and pulsed neutron diffraction data taken from the same sample.

Discussion

Since x-ray scattering is most sensitive to the positions of the heavier rare earth and aluminum component interactions, and because neutron scattering is particularly sensitive to the positions of the lighter oxygen atoms, these two techniques are highly complementary in the case of oxide glasses. When combined with Monte Carlo modeling techniques, a detailed 3-D picture of the glassy network that is consistent with the data can be obtained (see Fig. 3). Corner-sharing units are found to dominate the structure of these glasses.

Acknowledgments

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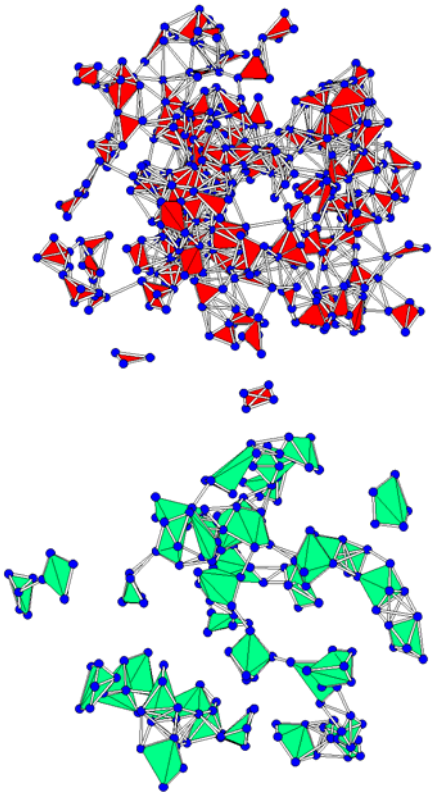

FIG. 3. Typical example of the local and intermediate-range structure of the AlO_4 (red) and YO_6 (green) polyhedra. The oxygen atoms are shown as blue spheres. The connectivity between units is shown to be mainly corner-sharing on the basis of the results obtained from the reverse Monte Carlo simulations taken on alumina-based glasses.

References

[1] S. Aasland and P. F. McMillan, Nature (London) **369**, 633 (1994).

[2] J. K. R. Weber, A. D. Hixson, J. G. Abadie, P. C. Nordine, and G. A. Jerman, J. Am. Ceram. Soc. **83**, 1868 (2000).

[3] J. Urquidi, C. J. Benmore, B. Tomberli, J. Neuefeind, and P. A. Egelstaff, "*ISOmer-X: Intermediate and Short Range Order Analysis of Measurements Using High Energy X-rays* (submitted).