Experimental report

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Proposal: **6-05-1002**

Council: 10/2018

Title: Structure of magnesium aluminosilicate glass by neutron diffraction with isotope substitution

Research area: Materials

This proposal is a resubmission of 6-05-997

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Samples: $(MgO)x(Al_2O_3)50-x(SiO_2)50$, x = 15, 25, 35

Instrument	Requested days	Allocated days	From	To
D4	4	5	17/07/2019	22/07/2019

Abstract:

Magnesium is an essential element in vitreous aluminosilicate materials that have a variety of applications, ranging from commercial display glass to the proxies for dry basaltic melts. The structural role of Mg in oxide glasses is, however, largely unknown because of (i) its ability to change the Mg-O coordination number from 4 to 6, and (ii) the absence of definitive structural information from experiment. We will therefore employ the method of neutron diffraction with isotope substitution to give site-specific information on the coordination environment of Mg in three different glasses along the $(MgO)x(Al_2O_3)50-x(SiO_2)50$ tie-line, with x = 15, 25 and 35. These compositions are chosen in order to highlight the response of the coordination environment of Mg to significant changes in the network structure that emerge, e.g., from the availability of bridging versus non-bridging oxygen atoms. The results will complement those obtained from 27Al and 29Si NMR, thus providing a complete structural picture for an important model system.

Structure of magnesium aluminosilicate glass by neutron diffraction with isotope substitution

The method of neutron diffraction with Mg isotope substitution was used to measure the structures of glassy $(MgO)_{25}(Al_2O_3)_{25}(SiO_2)_{50}$, $(MgO)_{37.5}(Al_2O_3)_{12.5}(SiO_2)_{50}$ and $(MgO)_{20}(Al_2O_3)_{20}(SiO_2)_{60}$. Specifically, let ${}^{\mathrm{Nat}}F(q)$ and ${}^{25}F(q)$ represent the total structure factors measured for two of these glasses that are identical in every respect, except that one contains magnesium of natural isotopic abundance ${}^{\mathrm{Nat}}$Mg with scattering length $b_{{}^{\mathrm{Nat}}\mathrm{Mg}}$ = 5.375(4) fm and the other contains the isotope ^{25}Mg with scattering length $b_{{}^{25}\mathrm{Mg}}$ = 3.62(14) fm. Site specific information on the Mg coordination environment was obtained from the first-difference function

$$\begin{aligned}\Delta F_{\mathrm{Mg}}(q) &\equiv {}^{\mathrm{Nat}}F(q) - {}^{25}F(q) \\ &= 2c_{\mathrm{Mg}}\left(b_{{}^{\mathrm{Nat}}\mathrm{Mg}} - b_{{}^{25}\mathrm{Mg}}\right)\sum_{\mu\neq\mathrm{Mg}} c_\mu b_\mu \left[S_{\mathrm{Mg}\mu}(q)-1\right] + c_{\mathrm{Mg}}^2\left(b_{{}^{\mathrm{Nat}}\mathrm{Mg}}^2 - b_{{}^{25}\mathrm{Mg}}^2\right)\left[S_{\mathrm{MgMg}}(q)-1\right]\end{aligned}$$

where $\mu \neq \mathrm{Mg}$ denotes a matrix atom (i.e., Al, Si or O), c_{Mg} denotes the atomic fraction of Mg, and c_μ and b_μ denote the atomic fraction and coherent scattering length of matrix atom μ, respectively [1]. The Mg-μ correlations were then removed by taking the weighted difference function

$$\begin{aligned}\Delta F(q) &\equiv \left[b_{{}^{\mathrm{nat}}\mathrm{Mg}}\,{}^{25}F(q) - b_{{}^{25}\mathrm{Mg}}\,{}^{\mathrm{Nat}}F(q)\right]\Big/\left(b_{{}^{\mathrm{nat}}\mathrm{Mg}} - b_{{}^{25}\mathrm{Mg}}\right) \\ &= \Delta F_{\mu\mu'}(q) - c_{\mathrm{Mg}}^2 b_{{}^{\mathrm{nat}}\mathrm{Mg}} b_{{}^{25}\mathrm{Mg}}\left[S_{\mathrm{MgMg}}(q)-1\right]\end{aligned},$$

where $\Delta F_{\mu\mu'}(q) = \sum_{\alpha\neq\mathrm{Mg}}\sum_{\beta\neq\mathrm{Mg}} c_\alpha c_\beta b_\alpha b_\beta\left[S_{\alpha\beta}(q)-1\right]$ contains information only on those pair-correlation functions describing the matrix atoms. Examples of the measured $\Delta F_{\mathrm{Mg}}(q)$ and $\Delta F(q)$ functions for glassy $(MgO)_{25}(Al_2O_3)_{25}(SiO_2)_{50}$ are shown in Fig. 1. A complete analysis of the results for the different glasses is underway.

[1] Salmon P S & Zeidler A 2013 Phys Chem Chem Phys **15** 15286

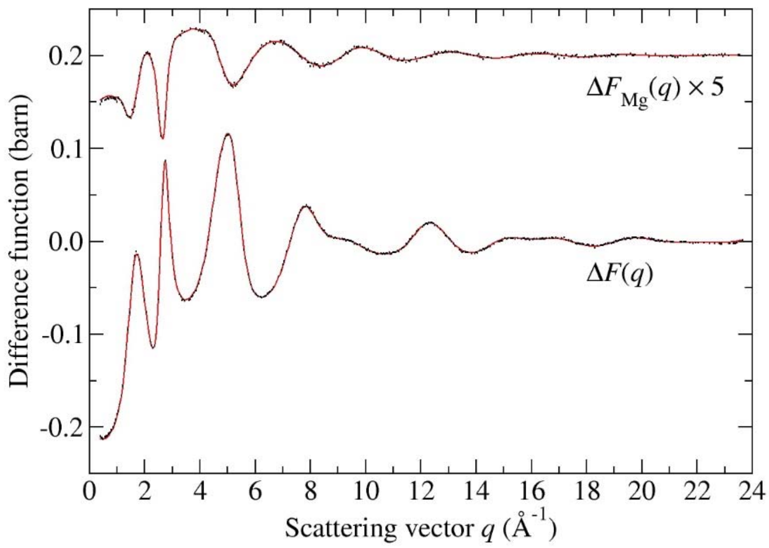

Fig. 1. The measured difference functions for glassy $(MgO)_{25}(Al_2O_3)_{25}(SiO_2)_{50}$. The $\Delta F_{\mathrm{Mg}}(q)$ function has been shifted vertically for clarity of presentation.