Experimental report

30/05/2022

Proposal: 6-05-1051 **Council:** 4/2021

Title: Structure of lithium silicate glass by isotope substitution

Research area: Materials

This proposal is a resubmission of 6-05-1024

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Samples: (Li2O)x(SiO2)1-x, x = 0.29, 0.33 or 0.36

Instrument	Requested days	Allocated days	From	To
D4	4	4	25/06/2021	29/06/2021

Abstract:

The Li2O-SiO2 system is at the heart of many technologically important glass-ceramic materials. However, little is known about the coordination environment of Li because of its inaccessibility from both x-ray diffraction and NMR experiments. The structure of Li2O-SiO2 glasses will therefore be investigated by using the method of neutron diffraction with Li isotope substitution. The results will reveal how the Li coordination environment changes with composition. They will also reveal the accompanying changes to the silicate network. The results will be combined with those obtained from 29Si NMR and high-energy x-ray diffraction in order to (i) establish how Li affects the composition-dependent connectivity of the silicate network, (ii) build realistic atomistic models of the glass structure in order to establish the origin of the increased rigidity of the glass with increased Li2O content and to elucidate the role played by voids in promoting fast-ion conductivity, and (iii) provide benchmarks for investigating the structural origin of the mixed alkali effect and the zero coefficient of thermal expansion in commercially important glass-ceramics.

Structure of lithium silicate glass by isotope substitution

The structure of the lithium silicate glasses $(Li_2O)_x(SiO_2)_{1-x}$ with x = 0.29, 0.33 and 0.36 was measured using neutron diffraction with ^{0}Li and ^{7}Li isotope substitution, where the zero denotes a null scattering length. For each composition, the first order difference function $\Delta D_{\mathrm{Li}}(r) = {}^0D(r) - {}^7D(r)$ was successfully obtained from the total pair-distribution functions ${}^0D(r)$ and ${}^7D(r)$ measured for the samples of $(^0Li_2O)_x(SiO_2)_{1-x}$ and $(^7Li_2O)_x(SiO_2)_{1-x}$, respectively. The $\Delta D_{\mathrm{Li}}(r)$ functions reveal the nearest-neighbour coordination environment of the mobile Li^+ ions. The results are being combined with those from high-energy x-ray diffraction and ^{29}Si magic angle spinning NMR experiments to build a complete picture of the glass network and find the effect of its rigidity on the ion mobility.

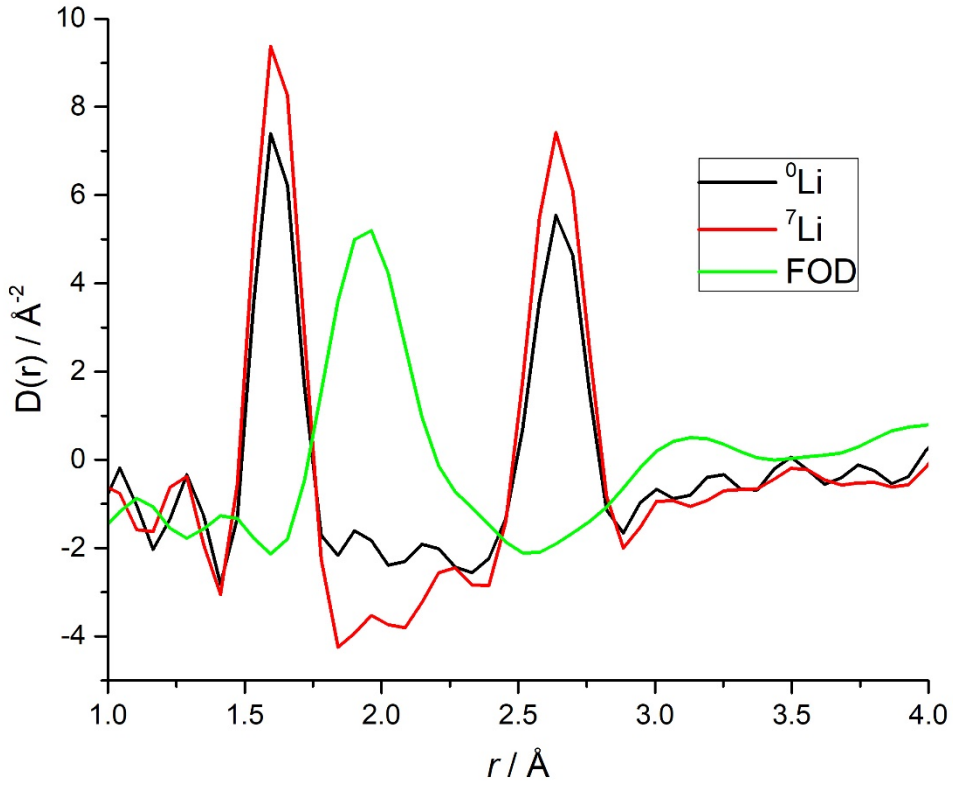

Fig. 1. The total pair-distribution functions ${}^0D(r)$ and ${}^7D(r)$ measured for samples of glassy $(^0Li_2O)_{1/3}(SiO_2)_{2/3}$ and $(^7Li_2O)_{1/3}(SiO_2)_{2/3}$, respectively, and the first order difference (FOD) function $\Delta D_{\mathrm{Li}}(r) = {}^0D(r) - {}^7D(r)$.