Structural study of novel alkali metal intercalated silica gel materials

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Introduction

Recently, candidate materials with good air stability for producing hydrogen were developed by doping liquid alkali metals into silica gels.[1]. In this work we attempted to investigate the structure of these new materials.

Methods and Materials

Three powder samples were prepared for room temperature X-ray scattering experiments: (1) NaK2-SG0: In this case the alkali metal, M, was the NaK2 alloy which is liquid at room temperature. (2) Na-SGI: Pure sodium was used as the metal and heated with the silica-gel to 150 ^{o}C (3) Na-SGII: Pure sodium was heated with silica gel, while agitating, to 400 ^{o}C.
The atomic pair distribution function (PDF) method has been used to study the structure of these materials.

Results

The atomic PDF, G(r), was calculated by Fourier transformation of the reduced structure function, F(Q) = Q[S(Q)-1], as follows:

$$G(r) = \left(\frac{2}{\pi}\right)\int_{Q_{min}}^{Q_{max}} Q\left[S(Q)-1\right]sin(Qr)\,dQ.$$

Here, $S(Q)$ represents the total scattering functions. $F(Q)$, were truncated at a Q_{max} of 20 Å^{-1} before the PDF was calculated. The PDFs obtained are shown in Fig. 1.

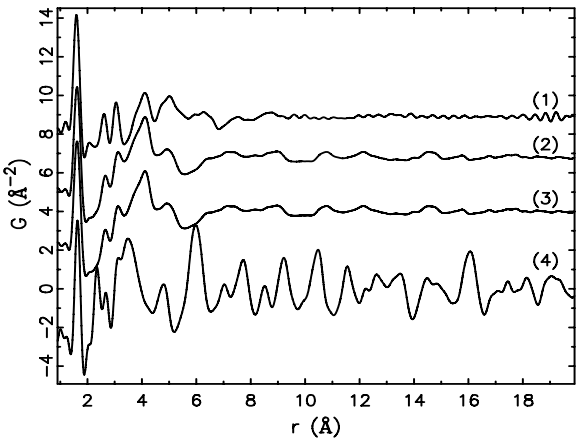

Fig. 1. Experimental PDFs for (1) pure silica, (2) NaK2-SG0, (3) Na-SGI and (4) Na-SGII.

Discussion

From the similarity in the data it is concluded that NaK2-SG0 and Na-SGI retain most of the features of the pure silica structure, with the existence of some inclusions, indicated by the broad oscillations on the nanometer scale.
For the case of Na-SGI, the PDF was calculated from the structure of metallic sodium and compared to the PDF. As evident in Fig. 2. the broad oscillations in the PDF of Na-SGI matched with peaks from the Na PDF.

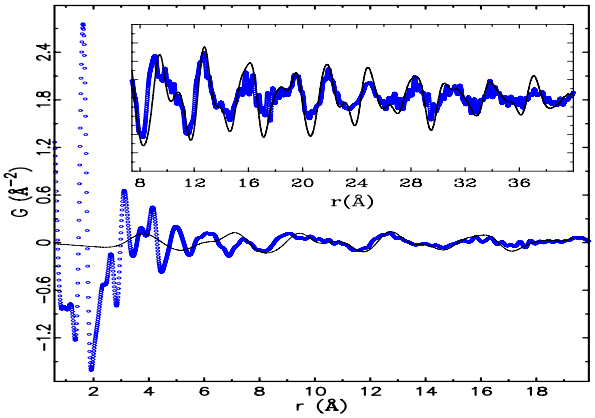

Fig. 2. Na-SGI Experimental PDF (circles) and the calculated PDF for metallic sodium (line). The inset shows the same data and fit curves in the high-r region.

In NaK2-SG0, attempts to fit the ripples using bcc simple metallic structures were unsuccessful. More work is in progress to investigate the origin of these oscillations.
Fig. 3a. shows single phase refinement with Na_2SiO_3 as the initial structure while Fig.3b shows a two phase refinement Na_2SiO_3 and NaSi. It can bee seen that the two phase refinement gave a better agreement from which we conclude that this material is composed from NaSi and Na_2SiO_3.

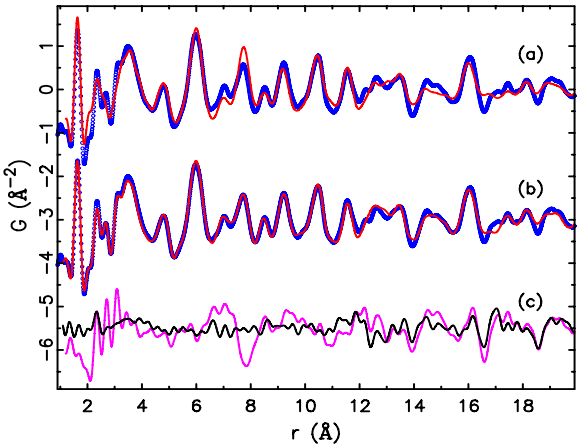

Fig. 3. Experimental (circles) and calculated (solid lines) PDFs for the Na-SGII sample, using (a) a single phase Na_2SiO_3 model and (b) (offset) a dual-phase model containing Na_2SiO_3 and NaSi . (c) the residual plots are shown displaced below the fits overlaid on each other; single phase fit (magenta) and for the dual-phase fit (black).

References

[1] J. L. Dye, K. D. Cram, S. A. Urbin, M. Y. Redko, J. E. Jackson, and M. Lefenfeld, J. Am. Chem. Soc. 127(26) (2005).