MS18-P09

High pressure pair distribution function study of amorphous silica in helium

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Glasses are used for a large number of technological and everyday applications. Among them, the system that has been most widely investigated is amorphous silica (a-SiO_2). The structure of aSiO_2 consists of corner-sharing SiO_4 tetrahedra which form *n*-fold rings (with $n \geq 2$, in a-SiO_2 mostly $n = 4, 5, 6, 7, 8$). The absence of Bragg reflections in diffraction experiments precludes the use of traditional crystallographic techniques to determine their atomic structure. However, it is clear that these materials possess well-defined local structure on the nanometer scale, which is often linked with their physical properties. Local structure can be obtained using x-ray total scattering from which the distribution of interatomic separations can be measured via the pair distribution function (PDF). High-quality PDFs require high-energy photons and access to large scattering angles to measure data to high values of momentum transfer ($Q = 4\pi \sin\theta/\lambda$).

It was recently discovered that when helium or argon are used as pressure transmitting medium, a-SiO_2 exhibits a completely distinct compression behavior[1]. The compressibility of a-SiO_2 drastically decreases in a helium medium which can easily be understood by the penetration of helium atoms (r_{He}=1.3 Å) in *n*-fold rings (with $n \geq 6$, $r_{6\text{-fold}}$ ring =1.5 Å) which prevents collapse of the rings, contrary to argon (r_{Ar}=1.88 Å), and therefore the densification of the material; based on a poromechanics model [2], up to 0.53 mole He can be incorporated under pressure. In order to determine the structural change dependence of helium adsorption on a-SiO_2, high pressure PDF measurements were undertaken.

High-pressure high-quality data were obtained using 61 Kev photons up to 22 $Å^{-1}$. The total scattering data was refined by the reverse Monte Carlo (RMC) method [3] in order to obtain changes to bond-angle distributions and network topology of a-SiO_2 in helium as a function of pressure. Partial PDFs were obtained and permitted to understand the structural modifications induced by the helium adsorption at high pressure which consist in: i) a broadening of the Si-Si bond distribution(inter-tetrahedra distances) in the first and second coordination shells; as a consequence appearance of a new contribution is also observed above 3 Å for the Si-O PDF, ii) a decrease in the O-He distance distribution with increasing pressure which becomes more ordered as a result of helium confinement.

References:

[1] T. Sato, N. Funamori, and T. Yagi, Nat. Commun. 2 (2011).
[2] M. G. Tucker, D. A. Keen, M. T. Dove, and K. Trachenko, Journal of Physics-Condensed Matter 17, S67 (2005).
[3] B. Coasne, C. Weigel, A. Polian, M. Kint, J. Rouquette, J. Haines, M. Foret, R. Vacher, and B. Ruffle, J Phys Chem B 118, 14519 (2014).

Keywords: PDF study, amorphous silica, helium asorption