Investigation on structures of simple glasses using PDF-analysis

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Abstract

X-ray diffraction techniques are frequently used to investigate structural aspects of crystalline phases due to their ordered structure and the presence of a long-range order (LRO). Glasses-like any other amorphous material-unfortunately lack this long-range order and thus the investigations on the structure of glasses is an arduous and time consuming task. Nowadays, concepts about the structure of glasses are mostly dealing with the immediate surrounding of constituting atoms leading to a short-range order (SRO), which for alumosilicate systems is mainly due to the SiO4-tetrahedra of the silicate network and the charge deficit tetrahedra emerging from Al3+-ions. On top of this short-range order, glasses also feature some kind of mid-range order (MRO) that separates glasses from the truly amorphous materials.

In this study we are using total scattering techniques, e.g. pair distribution function (PDF) analysis to assess the structure of simple glasses with special emphasis on the degree of polymerization and it's impact on the SRO as well as the MRO. To this end, we are using a laboratory X-ray diffractometer (Empyrean 3rd Gen.) equipped with an Ag-tube to allow for measurements up to high q-values (qmax 23 Å–1). Additionally, the system is equipped with a state-of-the-art hybrid pixel detector (GaliPIX3D) that features a CdTe sensor with 100% efficiency even at high energy radiations like that of the used Ag-tube. The powdered samples are prepared in quartz capillaries that are then attached to a standard capillary spinner. For measurements at elevated temperatures a HTK 1200N oven chamber from Anton Paar with a capillary extension is available.

The range of analyzed glasses comprises pure quartz (crystalline and amorphous), sodium silicate glasses, calcium silicate glasses, sodium aluminosilicate glasses as well as calcium aluminosilicate glasses of different degrees of polymerization (Q4-, Q3- and Q2-structures). The measured diffraction patterns are then converted to the reduced structure factor functions $S(Q)$ and the radial distribution functions $G(r)$ by using the program PDFgetX3 from P. Juhás et al.

To further evaluate the structure of these glasses, Reverse-Monte-Carlo simulations using the program Dissolve-provided by the Disordered Materials Group of ISIS-are conducted. To this end, cells with up to 12000 atoms are generated based on the general composition of the analyzed glasses. The attractive and repulsive interactions between the different atoms are approximated by using a 12-6-Lennard–Jones potential in addition to basic Coulomb interactions. Besides coordinates and visualization of atoms and their movement it also provides internal calculations resulting in a radial distribution function which can directly

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be compared to the measured distributions. Using the measured reduced structure factor functions and the radial distribution functions from the X-ray diffraction experiments as reference, the program Dissolve can also try to refine the structure of the analyzed simple glasses until the difference between the calculated and measured functions reaches a global minimum (provided the general solution just from given potentials is already in close enough agreement).

Keywords: pair distribution function, glass, structure, total scattering, X, Ray