Combined Solid-State NMR and Molecular Dynamics Study of the Structure of Strontium-Aluminosilicate glasses (a) Science and Technology of Glass

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Abstract

Aluminosilicate based glasses are widely used in glass industry and their atomic-scale structural and dynamical properties have been thoroughly investigated using various spectroscopic methods. Among them, solid state NMR has firmly established itself as a method of choice for providing key information for the elucidation of their atomic-scale structure. Recently, a methodology based on the combination of DFT-NMR calculations with molecular dynamics simulations has emerged as a significant step for the improvement of the detailed interpretation of experimental NMR spectra.
Using this approach, we have investigated the structure of aluminosilicate SiO_2-Al_2O_3-SrO based glass compositions which are largely unexplored systems. Glasses on the compensation line Al_2O_3 = SrO, were studied with ^{17}O, ^{29}Si and ^{27}Al solid state NMR at high (11.7 T) and very-high (20.0 T) magnetic fields, together with neutron diffraction spectroscopy. Classical and ab-initio molecular dynamics (MD) simulations were performed and combined with calculations of NMR parameters with the DFT-GIPAW method. Concerning MD, two analytical forms of the force-fields (Morse and Buckingham) were compared for the description of the short-range interatomic interactions. Computed NMR parameters were linked to local structural features to establish relationships between experimental NMR spectra and the underlying topological disorder (in terms of chemical and geometrical disorder) . NMR fingerprints of debated units such as tricoordinated oxygen atoms could be predicted with the aims to assess their existence from experimental data.
In agreement with experimental NMR data, MD simulations predict that aluminium is predominantly tetrahedrally coordinated for all the studied compositions with a small fraction of AlO_5 units ranging from 2-5%. Variations of the ^{29}Si NMR spectra, and to a less extent of ^{27}Al spectra, could be quantitatively correlated to the Al/Si mixing. In parallel, the Al/Si connectivities were investigated using advanced NMR techniques enabling the resolution of the ^{29}Si NMR spectrum in terms of $Q^n(mAl)$ units (i.e., Q^n connected to m Al units). Simulations of ^{17}O NMR experiments from our first-principles methodology combined to ^{17}O-^{27}Al correlation experiments allowed extractions of Al-O-Si, Al-O-Al and Si-O-Si peaks which were found to be strongly overlaping in experimental 1D and 2D ^{17}O MAS NMR spectra.
This study illustrates well this novel methodology which allows quantifying the medium-range order of amorphous materials by comparison of NMR results with first-principle NMR parameters calculations performed on Molecular Dynamic derived structures.

Brief Biographical Notes

(Please include some brief biographical notes, together with a head and shoulders photograph of the presenting author, to introduce the author to potential delegates.)

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