Combining through-space and through-bond interactions to visualise intermediate range structures with solid-state NMR spectroscopy

Laura Piveteau*[1], Franck Fayon[2], and Dominique Massiot[3]

[1]Conditions Extrêmes et Matériaux : Haute Température et Irradiation (CEMHTI) – Centre National de la Recherche Scientifique : UPR3079, Université d'Orléans : UPR3079 – Site Cyclotron, CS 30058, 3A rue de la Férolerie, 45071 Orléans Cedex 2 Site Haute Température, CS 90055, 1D avenue de la Recherche Scientifique, 45071 Orléans Cedex 2, France

[2]CNRS (CEMHTI) – Centre National de la Recherche Scientifique - CNRS – 3D avenue de la recherche scientifique, 45071 Orléans cedex 2, France

[3]Conditions Extrêmes et Matériaux : Haute Température et Irradiation (CEMHTI) – Université d'Orléans, Centre National de la Recherche Scientifique : UPR3079 – Site Cyclotron, CS 30058, 3A rue de la Férolerie, 45071 Orléans Cedex 2 Site Haute Température, CS 90055, 1D avenue de la Recherche Scientifique, 45071 Orléans Cedex 2, France

Résumé

In the range of spatial distances of the order of nanometers, the so-called intermediate range, chemical, geometrical and topological heterogeneities are suspected to be the origin for numerous structural rearrangements in glassy materials, such as polyamorphism, crystallization of stable and metastable phases, solid/solid phase separation (*e.g.* spinodal decomposition), and many more. However, in this size range, the continua of compositions or coordination geometries are difficult, if not impossible, to describe by phases and interfaces. Solid-state NMR spectroscopy is a powerful tool to determine chemical species and their distribution within amorphous materials. Nevertheless, structural understanding beyond the coordination shell remains challenging. We propose to combine through-space and through-bond interactions in solid-state NMR experiments to assess the intermediate range structure of glasses. We investigate the possibilities offered by the dipole-dipole-chemical-shift-anisotropy cross-terms in triple-quantum experiments to visualize the proximity of molecular units and combine with spin-diffusion after the selection of molecular units by multiple quantum filters. The functioning and validity of these approaches are illustrated on PbO-P2O5 glasses.

*Intervenant