Drivers of water transport in glass: chemical or topological effect of the glass network?

DIANE RÉBISCOUL[1], CLÉMENTINE MANSAS[1], JEAN-MARC DELAYE[2], THIBAULT CHARPENTIER[3], FLORENCE BRUGUIER[2], OLIVIER BOUTY[2] , BRUNO PENELON[2] , HÉLÉNE ARENA[1]

[1] CEA, ICSM – UMR 5257 CEA-CNRS-UM-ENSCM, 30207 Bagnols-sur-Cèze Cedex, France
[2] CEA, DEN, DTCD/SECM, Marcoule, F-30207 Bagnols sur Cèze, France
[3] NIMBE, CEA, CNRS, Université Paris-Saclay, CEA Saclay, 91191 Gif-sur-Yvette cedex, France.

For the first time, the relation existing between water transport in glass, topology and chemical elements (network formers and charge compensators) and their structural role in glass were investigated through a new approach : atomistic modelling of glass topology, glass and water structure analysis, and water diffusion characterization.

Two series of aluminosilicate glasses without and with boron and having various ratios of charge compensators CaO/Na_2O were used. The glass structure was characterized using Raman spectroscopy and NMR and the glass topology, i.e. the density of the bottlenecks and the interstitial sites, were obtained by molecular dynamics The results highlight that the presence of boron and the CaO/Na_2O ratio drive the role of Ca inside the glass structure. Ca strengthens the glass network except when a part of Ca is required to compensate the charge of AlO_4 and BO_4 units.

These glass properties were closely related to water transport characterized by the duration of the predominance of the hydration/interdiffusion processes, the apparent water diffusion coefficient and the water structure in the hydrated glass. These properties were studied and determined using X-Ray Reflectivity and ATR-FTIR. For aluminosilicate glasses, we show that water transport is mainly driven by glass topology through the role of Ca and its amount in glass. Indeed, Ca strengths the glass network decreasing the density of bottlenecks allowing the diffusion of water molecules. When boron is added to the glass, water transport is mainly driven by the chemical interactions of water molecules with Ca and the network formers (Al and Zr) of glass matrix.

These results show that the boron presence in glass and the CaO/Na_2O ratio drive the water transport in glass either by a topologic or a chemical effect.