IODINE DISSOLUTION MECHANISMS IN HIGH-PRESSURE ALUMINOBOROSILICATE GLASSES AND ITS RELATIONSHIP TO OXYGEN SPECIATION

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Incorporation of iodine (I) into high-pressure vitrified glasses appears to be a potential solution for the immobilization of ^{129}I radioisotopes. Under those conditions, I dissolution is strongly enhanced, however, the impact I dissolution has on the glass structure remains to be determined to assess the matrix durability.

We have studied experimentally the change in I solubility and speciation in a series of sodium aluminoborosilicate glasses (Na_2O ranging from 10 to 40 mol.%) held at 0.25 and 1.0 GPa and 1250°C. As expected, the I solubility increases with pressure conditions, with increasing Na_2O and is positively correlated to the glass optical basicity. The I speciation determined by XPS is changing with the initial loaded source of iodine (either I_2 or I_2O_5) with a predominant iodide form (I^-) in the glass structure.

The investigation of the oxygen environment in the I-bearing glasses using O 1s XPS revealed that I dissolution induces an apparent oxygen loss within the glass structure. This result is consistent with our current view on I dissolution mechanisms. Furthermore, the subsequent simulations of the O 1s XPS spectra suggest that I dissolution consumes non-bridging oxygen to form bridging oxygen. This change in the oxygen speciation points toward an increase in the glass durability that is an important aspect for nuclear waste immobilization.

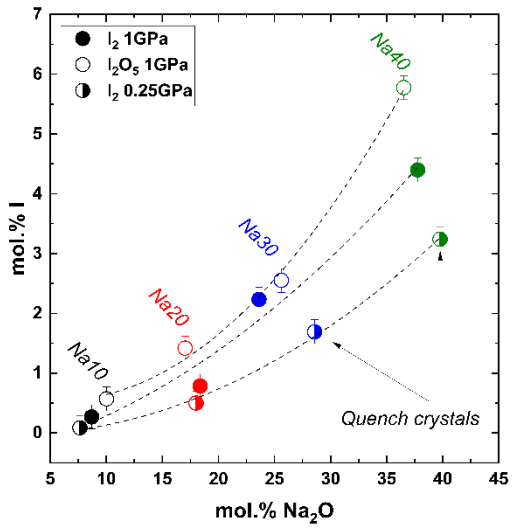

Figure 1: Evolution of the I solubility as a function of the glass Na_2O content expressed in mol.%. There is an increase in the I solubility with 1) increasing Na_2O content, 2) increasing pressure conditions from 0.25 to 1.0 GPa and 3) when I is loaded as I_2O_5 as compared to I_2.