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Effect of Nitridation on Structure, Glass Transition and Fragility of Phosphate Glasses

Georgiana-Laura Paraschiv[a], Francisco Muñoz[b], Yuanzheng Yue[a], Morten M. Smedskjaer[a]

[a]*Department of Chemistry and Bioscience, Aalborg University, DK-9220 Aalborg, Denmark*

[b]*Instituto de Cerámica y Vidrio (CSIC), Kelsen 5, 28049 Madrid, Spain*

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Oxynitride glasses are mixed-anion glasses in which the oxygen atoms have been partially substituted by either two-fold (N_d) or three-fold (N_t) coordinated nitrogen atoms, introducing additional bonds and thereby constraining the glass network and enhancing the atomic packing density. Phosphate glasses are more prone to undergo nitridation via ammonolysis at relatively low temperature in comparison to most other oxide glass systems and the nitridation improves the chemical durability of these otherwise hygroscopic samples, as compared with the parent glasses. However, the correlations among the glass transition, structure and dynamics in oxynitride glasses have not been well understood. In this work, we have studied these correlations for the sodium, lithium, and sodium-lithium phosphorus oxynitride glasses with 0-10 mol% SiO_2. Using differential scanning calorimetry (DSC), we investigated the effect of the N/P ratio on the glass transition temperature (T_g), jump in isobaric heat capacity at the glass transition (ΔC_p), glass transition width (ΔT_g), and liquid fragility index (m) of the various phosphorus oxynitride glasses. We have also determined the chemical durability and micromechanical properties of the samples. The changes in the various properties as a function of N/P ratio and base glass composition are correlated with the underlying changes in the short- and intermediate-range network structures, as obtained from ^{31}P and ^{29}Si NMR and Raman spectroscopy. Our results provide new insights into the structure-property relationships of oxynitride glasses, and also into the structural origin of the thermodynamic liquid fragility.