Phase Separated SiO2-B2O3-Na2O Glasses: Part I - Structure

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Abstract

Sodium borosilicate, coined SBN glasses, is of major interest, as the three constitutive oxides (NaO, B2O3 and SiO2) are commonly used in industrial glasses. Theoretical studies evidence the existence of a large phase separation region in the ternary phase diagram. The amorphous phase separation (APS) induces structure evolutions that influence physical and mechanical properties. Oxide glasses, including phases separated glasses, have many uses; however, a major weakness of glass is its fragility. Relatively moderate stresses can cause a sudden rupture without any warning. Glass is also sensitive to stress corrosion cracking (SCC): sub-critical cracking aided by environmental conditions (relative humidity, temperature, etc.). The SCC behavior of SBN glasses with APS is seldomly studied, in particular the link between meso-structure material and mechanical properties that can lead to a beneficial effect. The aim of this study is to characterize the structural evolution of APS-SBN glasses prior to stress corrosion cracking tests (C. L. Rountree will present the SCC behavior). Two compositions in the APS region were studied: SBN42 (67SiO2-26B2O3-7Na2O, wt %) and SNB96 (60SiO2-33B2O3-7Na2O, wt %). Glass samples were annealed to initiate the APS at different annealing times (ranging from 2 to 48 hours) and different annealing temperatures (range from 550 to 700∘C). Opalescence was tracked as an indicator of phase separation. Naked eyed sample observations after heat treatment evidenced opalescence is more pronounced with increasing annealing temperature and annealing time. Several techniques are used to characterize the structure evolution in annealed samples along with their pristine counterparts: XRD, Raman, NMR and AFM (Atomic Force Microscope). A specific procedure based on leaching (to remove the leachable B-rich phase) is used to reveal the structure of APS-SBN glasses analyzed by AFM. It has been shown that Si-rich phase constitutes the backbone of the structure and a spinodal decomposition is evidenced in annealed SBN42 samples and annealed SBN96 samples. The Si-rich phase size increases with annealing temperature and annealing time. Crystallization is not detected by XRD, except in SBN42 annealed at 700∘C. For SBN42, MAS-NMR spectroscopy shows an increase of BO3 ring units along with a decrease of BO3 non-ring units in annealed samples. This is

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consistent with the formation of B-rich phases due to APS. For SBN96, phase separation occurs even in pristine samples. Raman spectra indicate a significant structure evolution of the borate network. All the characterizations carried out highlight an important structural evolution, which should have a significant impact on the mechanical properties.

Keywords: Amorphous phase separation, structure, oxyde glass