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Enthalpy relaxation and microstructure evolution in hyperquenched SiO_2–Al_2O_3-ZrO_2 system

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A study of the sub-T_g enthalpy relaxation in glass far from equilibrium is crucial for understanding nature of glass. Recently, considerable progress has been made towards understanding the sub-T_g enthalpy relaxation in glass by using the hyperquenching-annealing calorimetry (HAC) method [1,2)] as well as modeling. However, most of the sub-T_g enthalpy relaxation studies have been done on stable oxide glass systems, i.e., those with low tendency to crystallization. Only a very few studies have been carried out on extremely unstable glasses.[1)] In order to get a broad picture about glass relaxation and glass transition, it is also necessary to investigate those extremely poor glass formers. Such investigation can provide valuable information on the structural heterogeneity in glass and liquid.

In this work, we study sub-T_g enthalpy relaxation and microstructure evolution of an extremely unstable glass former, i.e., a ternary oxide system SiO_2–Al_2O_3-ZrO_2. This special glass system arouses our interests mainly from three aspects. First, it is a material with broad applications in high temperature insulation. Second, it is obtained by hyperquenching (~10^6 K/s) the liquid during which high potential energy is frozen in. In the subsequent sub-T_g annealing process, the frozen-in energy will be gradually released, providing opportunities to study its microstructure evolution and sub-T_g enthalpy relaxation. The amorphous nature of the as-produced glass is verified by the X-Ray Diffraction (XRD) pattern as shown in Figure 1. Thirdly, it is an extremely unstable glass against crystallization since its crystallization peak overlaps with the glass transition peak in the isobaric heat capacity (C_p) curve. The results show that the structural ordering can take place during multiple DSC upscans to a temperature just above the glass transition temperature. Strikingly, the structural ordering is not just in nanoscale as our previous findings in a binary SiO_2-Al_2O_3 glass system[3)], but forms the zirconia crystal phase which can be verified by XRD (see Figure 1). Figure 2 shows the C_p curves of the hyperquenched (HQ) glass. The numbers near the C_p curves refer to the sequence of DSC upscans from 333 to1260 K at 20 K min^{-1}. In addition, the dynamics of microstructure evolution during sub-T_g annealing is also studied by XRD, high resolution transmission electron microscopy (HRTEM) and nuclear magnetic resonance (NMR). This work provides insight into both the thermal and structural origin of the extremely poor glass forming ability in SiO_2–Al_2O_3-ZrO_2 glass.

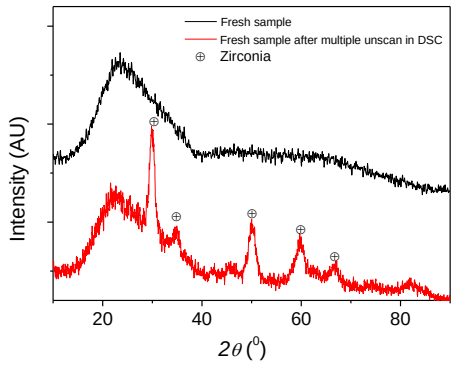

Figure 1. XRD patterns of both the fresh sample (HQ SiO_2–Al_2O_3-ZrO_2 glass, in black) and the sample after multiple (5) DSC upscans (in red).

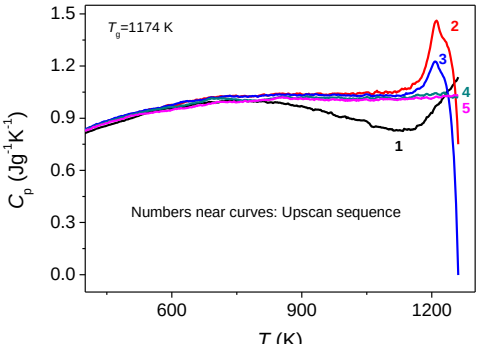

Figure 2. Isobaric heat capacity (C_p) against temperature for the HQ SiO_2–Al_2O_3-ZrO_2 glass subjected to multiple DSC upscans. Structural ordering could take place during the multiple upscans in DSC.

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