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An Invited Talk

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Sub-Tg enthalpy relaxation in unstable oxide glass formers: insights into the structural heterogeneity

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The structural heterogeneity in liquid has a strong impact on both glass stability and functionality of glass. Exploring the structural heterogeneity is crucial for understanding the glass transition and glass formation. However, one of the challenges for such exploration is lack of an effective method for detecting the structural heterogeneity in liquid. Here, we introduce a promising approach for detecting the structural heterogeneity in unstable oxide glass formers. This approach takes advantage of abundant features of the sub- $T_{\rm g}$ enthalpy relaxation in hyperquenched (HQ) unstable oxide glasses. We conduct the present study on the two unstable model glasses (towards crystallization): 65SiO_2 - $35 \text{Al}_2 \text{O}_3$ (SA) and 57SiO_2 -21 CaO-22 MgO (SCM) (mol%). This is done by hyperquenching the liquids via fiberization, annealing the HQ samples below $T_{\rm g}$, and then upscanning the annealed samples in a differential scanning calorimeter (DSC). For SA, we find that both sub- $T_{\rm g}$ annealing and repeated dynamic heating to $T_{\rm g}$ can greatly enhance structural ordering, and facilitate crystal growth upon further heating to well above $T_{\rm g}$. The results indicate that highly heterogeneous structure already exists in the liquid state. For SCM, we observe two exothermic sub-peaks in sub- $T_{\rm g}$ relaxation region during the first DSC upscan, implying that two types of structural domains occur already in the liquid state before HQ. The existence of the two types of structural domains is verified by direct transmission electron microscopy imaging. By performing sub- $T_{\rm g}$ annealing, we find that one type of structural domain is fragile, whereas another is strong. In addition, the sub- $T_{\rm g}$ annealing can be used as a sensitive tool for determining the glass forming ability.