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# Sub- $T_g$ Enthalpy Relaxation in a Hyperquenched $\text{SiO}_2\text{-Al}_2\text{O}_3$ System

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Abstract: We study the sub- $T_g$  enthalpy relaxation in an extremely unstable binary  $69\text{SiO}_2\text{-}31\text{Al}_2\text{O}_3$  (SA) glass against crystallization.<sup>1</sup> We reveal the structural origin of the extremely unstable feature of the studied glass by exploring the change of the medium-range structure during the relaxation process. This is done by using hyperquenching-annealing-differential scanning calorimetry (DSC), nuclear magnetic resonance (NMR) and high-resolution transmission electron microscopy (HRTEM). The DSC results show a non-symmetric energy release peak with a long tail at the lower temperature starting from the onset temperature for the hyperquenched SA glass. With increasing the degree of sub- $T_g$  annealing, the tail gradually evolves into a peak and the peak becomes broader. This phenomenon is quite different from that of many other hyperquenched glass systems, in which the long tail gradually disappears with increasing sub- $T_g$  annealing. Using NMR, we find that the contents of both the 5- and 6- fold coordinated Al species in the hyperquenched SA glass decrease with sub- $T_g$  annealing, while the 4-fold coordinated Al species increases. This leads to the formation of the oxygen triclusters, which can act as the mullite nucleation sites and facilitate the formation of mullite. This is confirmed by the ordered structural domains at nano-scale as shown in the HRTEM images. These ordered domains can lower the activation energy for further structural ordering during the sub- $T_g$  annealing, and thereby lowers the energy barrier for crystal growth. In addition, the extremely unstable behavior of the studied glass could also be attributed to the higher content of the 5- fold coordinated Al (~ 63%). The triclusters also indicate the structural heterogeneity of the supercooled liquid. This work provides insights into not only the structural evolution during the sub- $T_g$  relaxation but also reveal the structural origin of an unstable glass system in terms of the structural heterogeneity.

## REFERENCES

1. Zhang Y, Zhao D, Yue Y. Phase transitions and glass transition in a hyperquenched silica-alumina glass. *J. Am. Ceram. Soc.* (2017) DOI: 10.1111/jace.14880.