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Enthalpy Relaxation and its Correlation to the Medium-Range Structural Evolution in a Hyperquenched SiO₂–Al₂O₃ System

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We study the sub- T_g enthalpy relaxation in an extremely unstable binary SiO₂–Al₂O₃ (SA) glass and its correlation to the change of the medium-range structure. The results show that the energy release peak for the hyperquenched SA glass over temperature is not symmetric, and a long tail is observed at the lower temperature starting from the onset temperature of the enthalpy release. With increasing annealing, the long tail gradually evolves into a peak and becomes broader. This phenomenon is quite different from that of many other hyperquenched glasses, in which the long tail gradually disappears with annealing. We find that the contents of both the 5- and 6-fold coordinated Al species in the hyperquenched SA glass decrease with 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. This is confirmed by the ordered structural domains at nano-scale as shown in the HRTEM images. The extremely unstable behavior of the studied glass against crystallization is attributed to both the higher content of the 5-fold coordinated Al and the easily formed oxygen triclusters in the hyperquenched glass during sub- T_g annealing. This work provides insights into both the structural evolution during relaxation and the structural heterogeneity of an unstable glass system.