Origin of empirical Vogel temperature emerging from Molecular dynamics simulations

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Abstract

Vitreous silica was modelled by Molecular dynamics using BKS potentials and the obtained snapshots of glass structure were transferred into undirected graphs and decomposed into disjoint structural units that are ideally mixed to calculate configuration entropy. A good agreement with experimental heat capacity drop at Tg is demonstrated. Entropy is related with structural evolution of the obtained units; among them dangling oxygen dominantly effects low-temperature course of entropy that is fitted by two parabolas. The parabola corresponding to lower temperatures introduces temperature T^*, at which structure is completely frozen. It is proposed T^* stands as a counterpart of Vogel temperature in VFT equation. AGE model is introduced as the combination of the quadratic dependence of configuration entropy and Adam and Gibbs equation. AGE model not only removes the singularity at Vogel temperature but predicts existence of strong and fragile glasses as two limiting cases. Fragility is associated with quickness of structural response to temperature changes and distance between Tg and T^*. The model is tested on viscosity curves of SiO2 and B2O3 glasses and compared with VFT equation; fits obtained by AGE are better than or at least as good as those by VFT.

Keywords: Molecular dynamics, configuration entropy, Vogel temperature, glass transition

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