

Abstract Submitted
for the MAR09 Meeting of
The American Physical Society

Cooperativity and fragility in glass forming systems: not a simple relationship¹ LIANG HONG, ALEXANDER KISLIUK, ALEXEI SOKOLOV, Department of Polymer Science, The University of Akron, Akron, Ohio 44325-3909, USA, DR. SOKOLOV'S TEAM — Understanding the sharp increase of the main structural relaxation time τ on approaching the glass transition temperature (T_g) remains a great challenge. Traditionally this relaxation is considered as a cooperative process, with larger cooperativity leading to a steeper temperature dependence of τ around T_g , i.e. higher fragility. On the other hand, the boson peak, a collective vibration in the pico-second time region, is also described as a cooperative motion. In this study we estimate the structural correlation length for various glass forming systems from the collective vibration. The obtained values are in good agreement with the dynamic heterogeneity length estimated by 4 dimensional NMR for the main structural relaxation. Thus the two different motions appear to have very similar length scale for cooperativity. Direct comparison of cooperativity to fragility reveals no correlation. However, we discover that cooperativity correlates with the pure volume contribution to fragility. This result explains why many earlier attempts to find direct relationship between fragility and cooperativity fail. A possible origin for the observed correlation is discussed.

¹National Science Foundation

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Date submitted: 10 Dec 2008

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