

Neutron and Light Scattering Study of the Glass-to-Liquid-to-Glass and Glass-to-Glass Transitions in Dense L64 Copolymer Micellar System

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Abstract

Recent mode coupling theory (MCT) calculations show that if a short-range attractive interaction is added to a pure hard sphere system, one may observe a new type of glass originating from clustering effect (the attractive glass), as a result of the attractive interaction. This is in addition to the known glass-forming mechanism due to the cage effect in the hard sphere system (the repulsive glass). The calculations also indicate that, if the range of attraction is sufficiently short compared to the diameter of the hard sphere, within a certain interval of volume fractions where the two glass-forming mechanisms nearly balance each other, varying the external control parameter, the effective temperature, makes the glass-to-liquid-to-glass re-entrance and the glass-to-glass transitions possible. I shall present experimental evidence of both transitions, obtained from small-angle neutron scattering (SANS) and photon correlation measurements (PCS) taken from dense L64 copolymer micellar solutions in heavy water. Varying the temperature in certain predicted volume fraction range triggers a sharp transition between these two different types of glass. In particular, according to MCT, there is an end point (called A_3 singularity) of this glass-to-glass transition line, beyond which the long-time dynamics of the two glasses become identical. Our PCS results confirm this theoretical prediction. Surprisingly, although the Debye-Waller factor, the long-time limit of the coherent intermediate scattering function, of these two glasses obtained from PCS measurements indeed become identical at the predicted volume fraction, they exhibit distinctly different intermediate time relaxation. Furthermore, our experimental results obtained from volume fractions beyond the end point are characterized by the same features as the repulsive glass obtained before the end point. A complete phase diagram giving the boundaries of the structural arrest transitions for L64 micellar system is given for the first time.

Coworkers: Wei-Ren Chen (MIT), Prof. Francesco Mallamace (Messina, Italy). Dr. Charles Glinka (NIST)

References

- [1] *Small-angle neutron scattering study of the temperature-dependent attractive interaction in dense L64 copolymer micellar solutions and its relation to kinetic glass transition*, Wei-Ren Chen, Francesco Mallamace and Sow-Hsin Chen, Phys. Rev. E **66**, 021403 (2002).
- [2] *The glass-glass transition and its end point in a copolymer micellar system*, Sow-Hsin Chen, Wei-Ren Chen and Francesco Mallamace, Science **300**, 619 (2003).
- [3] *Small-angle neutron and dynamic light scattering studies of the liquid-to-glass and glass-to-glass transitions in dense L64 copolymer micellar solutions*, Wei-Ren Chen, Francesco Mallamace, Charles J. Glinka and Sow-Hsin Chen (Submitted to PRE, May, 2003).