Structural analysis of transient networks formed by star-shaped polymers

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Polymers crosslinked by temporary interactions form transient networks such as slimes, which show fascinating viscoelasticity due to a finite lifetime of crosslinks. Recently, there have been reports that temporary crosslinks play an essential role in polymeric materials' toughness and self-healing ability, making it necessary to understand the correlation between micro-kinetics and macro physical properties, such as viscoelasticity. However, the molecular understanding of such viscoelasticity has remained incomplete because of the difficulty of experimentally evaluating bond lifetimes and heterogeneous structures in conventional transient networks. Primarily, heterogeneous structures in transient networks are comprised of static and dynamic ones. Static heterogeneity includes dangling chains, heterogeneous distribution of network strand length, and branching number, while dynamic heterogeneity is due to a mixture of slow dynamics from network components and fast dynamics from micellar or under structures in one system. These heterogeneities are impossible to evaluate quantitatively and prevent molecular understanding of the physical properties of transient networks.

Recently, we developed a model system of transient networks with well-controlled structures (Tetra-PEG slime) to overcome these problems. The dynamically crosslinked Tetra-PEG gel comprises mutually reactive four-armed polyethylene glycol (tetra-PEG) polymer units. These narrow distribution star-shaped precursors allow the formation of a network composed of regular structures with uniform functionality and strand length. In addition, using symmetric precursors with the same diffusibility allows suppression of the dynamic heterogeneity concerning that of conventional temporary networks. However, the detailed relationships between the structural change and the network topology, such as connectivity and density of crosslinks, still need to be clarified, preventing understanding of the viscoelasticity's molecular origin. In this SANS study, we plan to measure the structural change of the dynamically crosslinked Tetra-PEG slime.

The SANS profiles of PEG derivative solution with different temperatures and concentrations in D2O are shown in Fig. 1. The profiles of the solution could be fitted with Orenstein Zernike (OZ) functions very well. The tendency is consistent with the previous reports, suggesting the validity of the experiments. We will compare the data with that in the crosslinked state, to elucidate the structural change by network formation, which is reported near future.

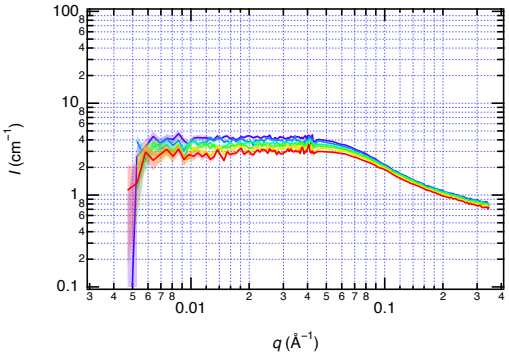

Fig. 1. SANS profiles of PEG derivative solutions with different temperatures and concentrations using D2O as solvent.