

Relationship between structures and mechanical properties of transient networks with uniform network strands

T. Katashima^A, T. Sakai^A, M. Ohira^A, and X. Li^B

^AUniv. of Tokyo, ^BHokkaido Univ.

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 important 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. A 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 unimer structures in one system. These heterogeneities are impossible to evaluate quantitatively and prevent molecular understanding the physical properties of transient networks.

Recently, we developed a model system of transient networks with well-controlled structures (dynamically crosslinked Tetra-PEG gel) 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 comprising regular structures, with uniform functionality and strand length. In addition, the use of symmetric precursors with the same diffusibility allows suppression of the dynamic heterogeneity with respect to that of conventional temporary networks. However, the relationships between the structural change and

the deformation remain unclear, which prevent understanding the molecular origin of the viscoelasticity. In this SANS study, we plan to measure the structural change of the dynamically crosslinked Tetra-PEG gels accompanied with shear deformation.

Due to the limitation of the measurement condition, we only tested non-deformation state. The SANS profiles of tetra-PEG with phenyl boronic acid end groups solutions in D2O are shown in Fig. 1. The profiles of tetra-PEG with phenyl boronic acid end groups could be fitted with Orenstein Zernike (OZ) functions very well. However, the level of scattering intensity is not proportional to the concentration, suggesting the drying of the solvent or sample preparation error. We will optimize the sample preparation and further perform the structure analysis on the dynamically crosslinked Tetra-PEG gels.

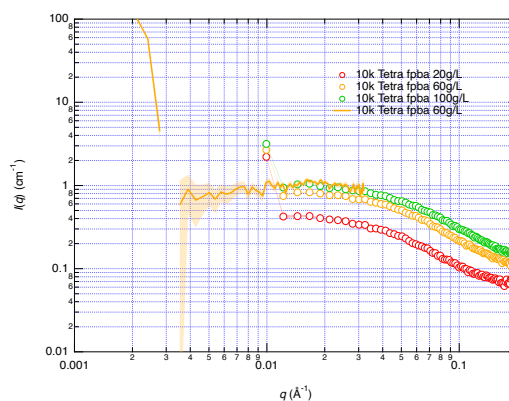


Fig. 1. SANS profiles of tetra-PEG solutions with phenylboronic acid end groups using D2O as solvent, at sample-to-detector (SDD) distances of 2 m and 12 m.