Structural Study on Uniaxial Extension of Nanocomposite Hydrogel with High Clay Concentration by Means of Contrast Variation Neutron Scattering

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In these days, mechanically excellent gels are developed such as topological gels, double network gels, and nanocomposite gels (NC gels). NC gels are hybrid materials consisting of organic polymers (poly(N-isopropyl acrylamide)) and inorganic clay nanoparticles (hectorite) , which have excellent physical properties, such as toughness, high modulus, deformability, etc. It is interesting and important to account for these mechanical properties on the molecular level for material science as well as industrial applications. In order to elucidate the microscopic structure of NC gels under elongation, we carried out contrast variation SANS (CV-SANS) experiments by varying the D_2O fraction in the aqueous solvent. Since NC gels are ternary systems consisting of clays, polymers, and water, observation of partial scattering functions reflecting each component is straightforward for quantitative structural analyses of the multicomponent system. So the CV-SANS techniques are essential for our system. NC gels are synthesized by polymerization with a mixture of N-isopropyl acrylamide monomers and Laponite clay nanoparticles in aqueous solutions. To apply CV-SANS techniques, we prepared five different NC gels consisting of 0, 22.6, 70, 80 and 100vol% D_2O fraction. SANS experiments were performed at SANS-U diffractometer of Institute for Solid State Physics, the University of Tokyo. This experiment has been the first attempt to apply the CV-SANS techniques to uniaxially-stretched hydrogels. With CV-SANS anisotropic 2-dimentional SANS patterns were clearly observed (see Fig. 1). The stretching ratio is $\lambda = 1, 3, 7$. The upper 6 figures indicate scattering from clays, i.e., the scattering contrast between polymer and solvent was matched, and bottom 6 ones are from polymers, i.e., the scattering contrast between clay and solvent was matched. The high-Q (detector length=2m) 2-dimensional patterns show elliptic pattern reflecting the similar orientations of clay and polymer for the uniaxial stretch. The low-Q (detector length=8m) patterns show similar two-lobe patterns for both clay and polymer scattering at $\lambda = 3$, which differ at $\lambda = 7$. This tendency clearly indicates that polymers are adhered to clay surfaces, which start to be torn off at $\lambda = 7$. **Reference**

S. Miyazaki et al., *Macromolecules* **39**, 8112, (2006).

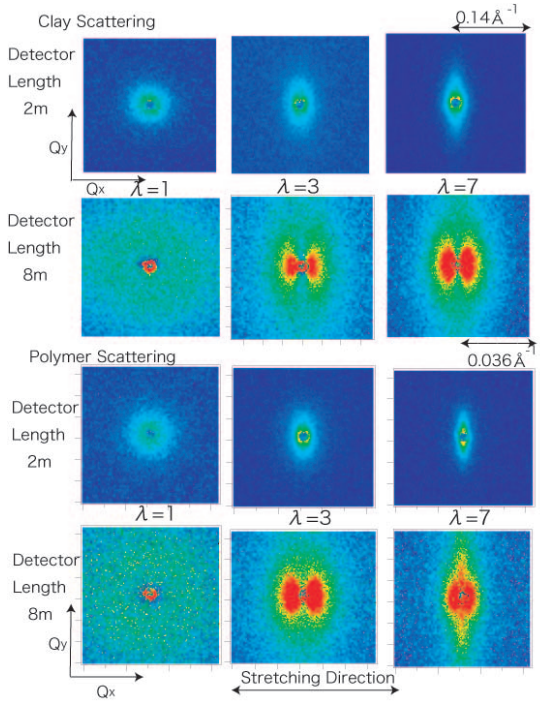

Fig. 1. 2-dimentional SANS patterns for uniaxially stretched NC gels at high-Q and low-Q.