Structural Analysis of N-isopropylacrylamide / Rod-like silica Hybrid Gels by Contrast-variation Small-angle Neutron Scattering

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Polymer gels, which are soft materials that retain large amounts of solvent, can be functionalized or strengthened by incorporation of inorganic compounds. The organic/inorganic hybrid gels thus obtained can be of various morphologies, such as semi-interpenetrating polymer networks with inorganic domains as well as polymer networks that are organically cross-linked and then strengthened by the incorporation of inorganic compounds. In addition, there exist organic hybrid gels free of organic cross-links, but which are cross-linked by the inorganic compounds via chemical or physical cross-linking. Detailed studies on hybrid polymer morphology, gelation dynamics, and interfacial structures between polymers and inorganic compounds have so far been limited by the inherent complexity of the systems. This complexity originates from the molecular interactions of the multi-component systems (i.e., the polymers, the inorganic compounds, and the solvent).

In this study, we investigated the microscopic structure of N-isopropylacrylamide (NIPAm)/rod-like silica hybrid gels. NIPAm gel is one of the widely studied thermo-responsive gels and a model system for studying hydrophobic interactions. It is prepared by using redox-polymerization and shows an attractive interaction with the silica surface via hydrogen bonding. It can be possible to discuss the influence of the distribution of inorganic compounds in the gel by flexibly changing not only the concentration but also the size or the shape (sphere or rod) of silica particles. The systematic study on the mechanical property and microstructure of hybrid gel, which can be tuned by these quantitative or geometrical arrangements of inorganic compounds, should be of great importance in order to design the high-performance gel. One of NIPAm/rod-like silica gel and three types of NIPAm/spherical silica gels, each having different silica sizes, were prepared while maintaining an equal total volume fraction. Contrast-variation small-angle neutron scattering measurements were performed to clarify the silica shape dependence of the hybrid gel microstructure. The presence of an adsorbed layer of NIPAm chains on the silica surface and its dependence on silica particle size are addressed.

Fig.1 shows the intensity profiles for (left) rod-like silica and (right) NIPAm/Rod-like silica hybrid gels. In the case of rod-like silica, the distinct peak appeared in the low Q region indicating the repulsive interaction of each particle. On the other hand, for the hybrid gels, the power law behavior was observed due to the fractal-like aggregation of each particle. Moreover, it should be noted that the shoulder was observed for each sample indicating the form factor of each particle. By using these experimental data, the surface structure around silica particles was discussed.

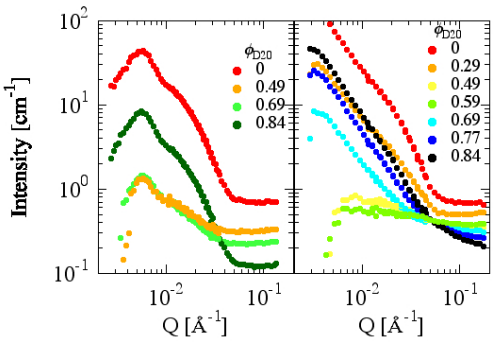

Fig. 1. Intensity profiles for Rod-like silica (left) and NIPAm/Rod-like silica hybrid gels (right).