Strain-induced structural change of slide-ring network in ion gel

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Ion gels, containing an ionic liquid in their polymer network, are a candidate material for flexible electronics. However, the brittleness of the ion gel was an obstacle to playing a role in wearable devices. To solve this problem, we fabricated slide-ring (SR) ion gels (Fig. 1(a)). Slidable cyclodextrin (hpCD) rings crosslinked PEO chains. The movable crosslink prevents stress concentration, resulting in high stretchability. Liu *et al.* [1] found a breakthrough in SR hydrogels with low ring coverage and high polymer concentration, where strain-induced crystallization (SIC, Fig. 1(b)) of oriented poly(ethylene glycol) (PEG) chains provides toughness. Applying this strategy, we successfully fabricated tough SR ion gel. SIC requires the phase separation of pure PEO from ionic liquid-rich phases, and thus, the phase separation structure is related to the mechanism of the formation of SIC.

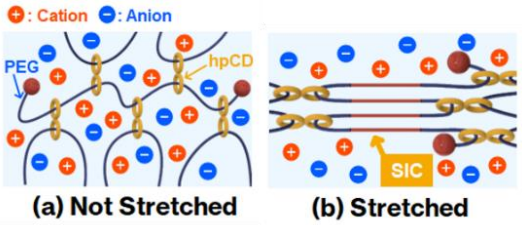

Fig. 1. Schematics of (a) slide-ring ion gel and (b) SIC of oriented PEG chains.

In this study, we obtained a small-angle neutron scattering (SANS) profile of the SR ion gel during stretching using JRR-3, SANS-U. Deutrated ionic liquid, d8-1-ethyl-3-methylimidazolium bis(trifluoromethane-sulfonyl)imide (d8-[C_2mim][NTf_2]) was synthesized and used. The prepared ion gel sample contains 60 wt% of d8-[C_2mim][NTf_2]. The ion gel sample with rectangular shapes (5 cm x 6 cm x 1 mm) was set to a homemade stretching machine. The neutron beam (diameter: 1 cm) is irradiated to the stretched sample. A small-angle X-ray scattering (SAXS) was conducted using BL-10C, Photon Factory.

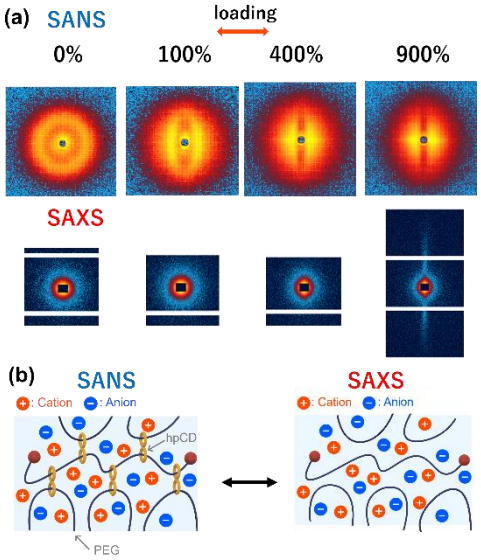

Fig. 2. (a) SANS and SAXS 2D images for stretched SR ion gels. (b) The effect of scattering contrast different between SANS and SAXS.

Fig. 2(a) shows the obtained SANS and SAXS images. In SAXS measurement, the scattering length density of hpCD (crosslinker) is matched to the ionic liquid (Fig. 2(b)). Therefore, SAXS measurement only reflects the contrast between PEG and ionic liquid, while SANS measurement reflects the contrast between hpCD, PEG and ionic liquid. In the SANS image, the orientation change of the structure with the characteristics length of ca. 20 nm was observed during 0-400% stretching, while such a peak was not observed in SAXS. Thus, this change can be ascribed to the change in the arrangement of the crosslinks induced by stretching. At higher loading (900%), a strong streak perpendicular to the stretching axis appears in the SAXS image, indicating the formation of bundles of the PEG parallel to the stretching axis. This study revealed the microscopic structural change in the structure of SR ion gel by stretching, and further investigation could provide insight to the design of a network structure efficiently forming SIC.

[1] C. Liu *et al.*, Science **372**, 1078 (2021).