## Anisotropy in strain-induced structural change of slide-ring network in ion gel

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We conducted structural analysis during stretching on slide-ring (SR) ion gels (Fig. 1).



Fig.1 Schematics for SR ion gels.

SR ion gels comprise a polymer network with a slidable-crosslinking point ((2 hydroxypropyl)-*α*-cyclodextrin, hpCD) and ionic liquid. The slide of the hpCD disperses stress concentration during stretching, and thus, the ion gel shows excellent mechanical toughness. Recently, we performed wide-angle X-ray scattering (WAXS) and found that straininduced crystallization (SIC) of oriented poly(ethylene glycol) (PEG) chains provides extraordinal toughness to the ion gel. The anisotropic network deformation leads to the highly oriented PEG, and the PEG chain crystalizes to prevent the propagation of microcracks. However, the network structure during stretching has not yet been elucidated.

In this experiment, we focused on the reversibility of the network after stretching. Using contract matching, we separated the two factors, i.e., the structures of hpCD and PEG. We used deuterated ionic liquid, d8-1-ethyl-3 methylimidazolium bis(trifluoromethanesulfonyl)imide  $(d8-[C_2mim][NTf_2])$  as the solvent. In X-ray, the scattering length of hpCD (~ 13.4 x 10<sup>-6</sup> Å<sup>-2</sup>) is matched to that of d8-[C<sub>2</sub>mim][NTf<sub>2</sub>] (~ 13.1 x 10<sup>-6</sup> Å<sup>-2</sup>). Therefore, only the PEG structure can be discussed in X-

ray, whereas hpCD structure is also observed in small-angle neutron scattering (SANS). No obvious scattering peak was observed in SAXS, indicating PEG homogeneously disperses in the ion gel. On the other hand, SANS profiles (Fig.2) exhibited a peak, which can be ascribed to the hpCD aggregation. The poor solubility of the hpCD in  $d8$ -[C<sub>2</sub>mim][NTf<sub>2</sub>] might result in the aggregation.



Fig.2 SANS profiles of SR ion gel at pristine, 100% loaded and unloaded states (a) horizontal and (b) vertical to the stretching axis. Intensity is shifted by 10 and 100 times for loaded and unloaded profiles, respectively.

In the vertical to the stretching axis, strain induces small changes in the profiles. On the other hand, in the horizontal to the stretching axis, the peak shift to lower  $q$  is induced by stretching, indicating increased interaggregation distance. However, in the unloaded state, the peak position almost completely recovers to the pristine state. Therefore, we concluded that the network structure is reversible even after the sample experiences 100% of strain. We confirmed that mechanical properties are also reversible, and the ion gel stands thousands of cyclic loading and unloading. We concluded that the structural reversibility of the network contributed to the mechanical reversibility of the SR ion gel.