Structural analysis of thermoresponsive Tetra gel

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Hydrogels are used as scaffolds for tissue engineering, temporary supports for cells, and vehicles for drug delivery systems. Although specially engineered hydrogels are known to exhibit excellent physical properties, they may have limited applicability because conventional hydrogels " swell " in water. Swelling drastically weakens the mechanical toughness.

Our strategy to achieve a robust hydrogel is based on the control of swelling by means of the introduction of thermoresponsive segments into the polymer network. We fabricated hydrogels composed of tetra-armed hydrophilic and thermoresponsive polymer units (poly(ethylglycidyl ether-co-methyl glycidyl ether); PEGE). The cross-linking reaction is based on the mutually reactive functional end-groups (active ester and amino end-groups). Here, the molar amount of active ester end-groups always equals to that of amino end-groups.

According to the previous study, PEG-PEGE hydrogels can endure a compressive stress up to 60 megapascals and can be stretched more than sevenfold without hysteresis. Our results demonstrate that the suppression of swelling helps retain the mechanical properties of hydrogels under physiological conditions.

In spite of such an advancement of research, there remains some mysteries. One of the mysteries is a swelling behavior as shown in Fig. 1. The equilibrium swelling ratio is not proportional to PEGE segment ratio. To clarify this point, we conducted SANS experiment on PEG-PEGE gels with varying PEGE segment ratio. Fig. 2 shows SANS profiles for PEG-PEGE gels. As shown in Fig.2, PEG-PDMS gels show some peaks, which correspond to the characteristic length of tens of nanometer. Domain distances evaluated from the first peak in SANS profiles are shown in Fig.3. When PEGE segment ratio is below 0.35, domain distances decrease as increasing PEGE ratio. However, when PEGE ratio is over 0.35, domain distances increases as increasing PEGE ratio. This inversion can be also observed in the swelling test. These results strongly indicates that conformational transition can be occurred at 0.35. We expect that a variety of morphological transitions can be observed by further investigations.

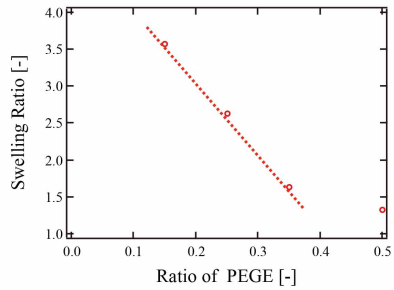

Figure 1. The dependence of equilibrium swelling ratio on PEGE ratio.

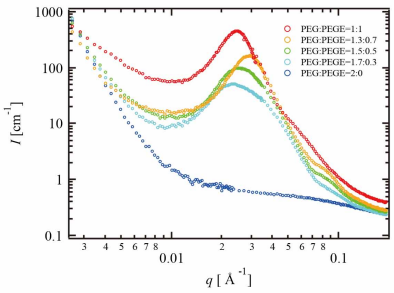

Figure 2. SANS profiles for PEG-PEGE gels.

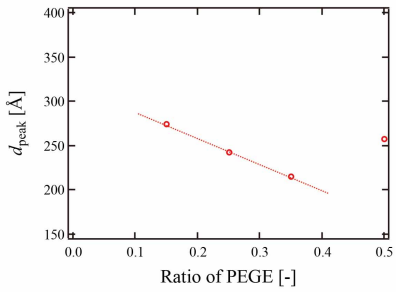

Figure 3. Interdomain distances obtained from SANS profiles.

Fig. 1.