Structural analysis of conversion-limited critical cluster gel by small angle neutron scattering.

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A polymer gel is a single polymer with complex three dimensional network, in which a large amount of solvent can be trapped. Polymer gels can be synthesized by various chemical methods: radical co-polymerization of monomers and cross-linkers, cross-linking the linear polymer chains by gamma-ray irradiation, coupling end-groups of star polymers, etc. Although there are many different chemical systems for various physical quantities (e.g. size-distribution, weight-averaged molecular weight, correlation length).

Recently, we established a gelation system by mixing two type of tetra-functional prepolymers, which have complementary reactive end-groups with the other type of prepolymers. In our study in ANSTO, we measured a series of critical polymer clusters by mixing these two types of prepolymers at off-stoichiometric ratio (Figure 1). With this systematic study, we have confirmed the strong universality of critical polymer clusters in in terms of fractal dimension, and for the first time found that size- distribution is actually a tunable parameter, which has been misunderstood as a universal property (Hayashi, Li et al, Nature Biomedical Engineering, 2017).

In this study, we carried out SANS study to investigate the critical clusters on way become a gel by quenching the gelation reaction near the gel point. First of all, we prepared Amine-terminated tetra-functional polyethylene glycol and N-hydroxysuccinimide-terminated tetra-functional PEG are mixed by stoichiometric molar ratio in D2O buffer(50 mM sodium phosphate, pH 3.0, 3.5, 4.0) at the various final concentration of 15, 30, 60 g/L. The reaction bath quenched near the gel point by adding small amount of HCl solution into the reaction bath to make pH close to 0, in which the reaction of amine and NHS stops. According to our previous study in ANSTO, each resultant critical solution should be diluted into different level (60, 30, 15, 7.5, 3.75, 1.88, 0.94 g/L) to fully characterize the structure and the size-distribution of the critical clusters. As the result is shown in figure 2, there are several interesting features. In the next time, we will be executed detailed analysis.

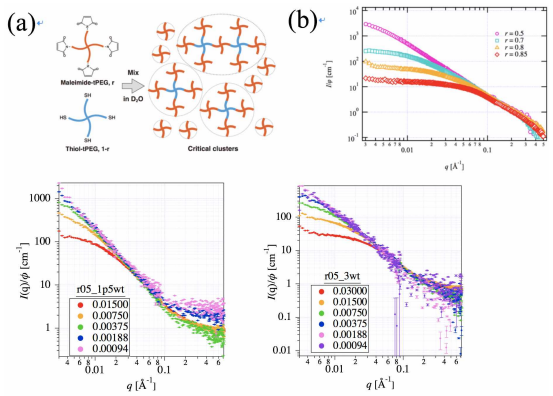

Fig. 1. Figure 1. (a) The critical clusters synthesized by imbalancing the prepolymers (b) The scattered intensities of different critical polymer clusters in previous study. Figure 2. The SANS profiles of critical cluster.