Structure characterization of Tetra-PEG gel having homogeneous network structure

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The structure of Tetra-PEG gel, a new class of biocompatible, easy-made, and high-strength hydrogels consisting of a four-arm polyethylene glycol (PEG) network, has been investigated by means of small-angle neutron scattering (SANS). Since the Tetra-PEG gel is prepared by cross-end-coupling two kinds of four-arm PEG macromers having different functional groups at the ends, i.e., amine group and succinimidyl ester group respectively, coupling reaction occurs exclusively between PEG chains carrying different functional groups. Structure analyses of Tetra-PEG gels were carried out by means of swelling experiments and SANS, and the results were discussed by taking into account the mechanical properties of the same systems. The following facts were disclosed.

(1) Tetra-PEG gels are stoichiometrically prepared irrespective of the initial polymer concentration, and their swelling behaviors are well predicted by the Flory-Rehner theory. (2) The mechanical moduli of Tetra-PEG gels, E and G, are proportional to the initial polymer concentration and is one order of magnitude larger than the corresponding gels made with similar tetra-arm PEG gels prepared with a low-molecular-weight coupling reagent. This indicates that cross-end-coupling of A- and B-type tetra-PEG is essential for gel preparation with extremely low defects. (3) The scattering functions of the macromers can be well reproduced by the scattering function for star polymers. (4) SANS functions of Tetra-PEG gels can be described by simple Ornstein-Zernike function without excess scattering component originating from cross-linking inhomogeneities. This means that Tetra-PEG gels are extremely homogeneous, and an " ideal " network free from defects is formed. (5) Preparation in non-stoichiometric composition leads to formation of defects in the polymer chain network and results in a significant depression of the mechanical properties. Structural models of macromer solutions and of Tetra-PEG gels, which account for the advanced mechanical properties of Tetra-PEG gels, are proposed.

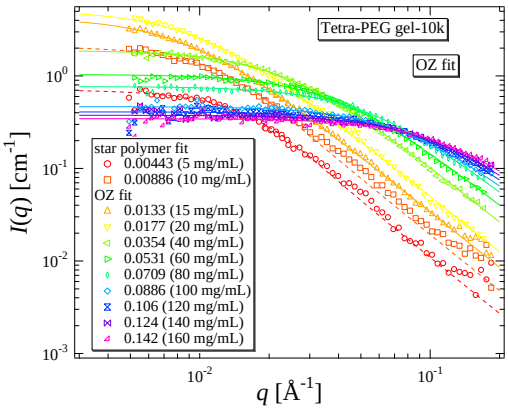

Fig. 1. SANS intensity functions of Tetra-PEG gel. The solid and dashed lines are fitting results of Ornstein-Zernike function for gels at high macromer concentration and 4-arm star polymer for imperfect gels prepared at low macromer concentrations, respectively.