

description of the molecular structure of the amyloid fibril reconciles the classical description of it as a cross- $\beta$  structure with the twisted  $\beta$ -sheet now known to be the lowest energy state, and in doing so generates a new fibrous protein structure in which  $\beta$ -strands can be hydrogen-bonded together over large distances in a stable structure.

#### Time-Resolved SAXS Measurements on Gel Setting and Gel Melting of Triblock Copolymer Gels

K. Reynders, H. Reynaers

Laboratory of Macromolecular Chemistry, Catholic University of Leuven, Celestijnenlaan 200 F, B-3001 Heverlee, Belgium

The unique properties and practical applications of triblock copolymer gels are related to the specific morphology. The endblocks of a triblock copolymer, polystyrene-block-(hydrogenated butadiene)-block-polystyrene (SEBS) or polystyrene-block-(hydrogenated isoprene)-block-polystyrene (SEPS), will associate in a solvent selective for the midblock, and result in a three-dimensional organisation. This implies an electron density difference, detectable in a small-angle scattering experiment, if the superstructure has dimensions in the range of 10 - 1000 Å.

The gels were investigated by small-angle scattering of X-rays and neutrons (SAXS and SANS) and exhibit a pronounced superstructure [1] which appears to be a function of the block copolymer concentration, molar mass, end/mid block ratio, deformation [2] and temperature. From the variations in the two interdomain interference maxima and one or more intradomain interferences with varying sample parameters, it follows [3] that the polymer concentration and molar mass influence the degree of organisation in the gel. A certain molar mass corresponds, for a specific polymer concentration, to the most disordered arrangement. Each deviation from these values introduces more order. The scattering patterns of the isotropic samples are fitted with two different models [3]; the Percus-Yevick hard-sphere interacting liquid model and the local coordination model of distorted hexagonal lattice.

Recent time-resolved SAXS experiments [4] reveal changes in the superstructure. Upon heating, the morphological order disappears, in agreement with DSC and rheological experiments. Gels with high molar mass, show a initial increase in order. These gels reach a thermodynamically more favourable morphology. After heating these gels with high block

molar mass above the temperature where disordering occurs, followed by cooling, they again exhibit increased order. Thermo-reversibility appears for these gels after a second heating scan.

Another effect is the temporarily change in structure for gels with relatively low block molar mass from hexagonal to a cubic-like order at a temperature around 120°C. Isothermal SAXS experiments in a temperature region around 100°C, reveal the same changes in superstructure. These morphological changes involve chain mobility which is relatively low in the case of long chain lengths. This explains the temperature difference, related to a time-effect, between dynamic and isothermal SAXS measurements. These morphological changes correspond to a critical gel formation in rheology experiments [5], performed in the same temperature region.

In the case where no improvement of order occurs during heating, cooling the gel from the temperature where the gel is in the disordered state yields the same scattering curve as before heating, indicating their thermo-reversibility.

#### References

1. N.Mischenko, K.Reynders, K.Mortensen, R.Scherrenberg, F.Fontaine, R.Graulus, H.Reynaers. *Macromolecules*, **27**, 2345-2347 (1994)
2. K.Reynders, N.Mischenko, K.Mortensen, N.Overbergh, H.Reynaers. accepted for *Macromolecules*
3. N.Mischenko, K.Reynders, M.H.J.Koch, K.Mortensen, J.S.Pedersen, F.Fontaine, R.Graulus, H.Reynaers. *Macromolecules*, **28**, 205-2062 (1995)
4. K.Reynders, M.H.J.Koch, N.Mischenko, H.Soenen, H.Reynaers. To be published in *Macromolecules*.
5. H.Soenen, A.Liskova, K.Reynders, H.H.Winter, H.Berghmans submitted to *Macromolecules*.