Structure and Dynamics of Supramolecules Investigated by Contrast Variation Neutron Scattering

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Polyrataxane (PR) is a new category of supermolecular assemblies in which a linear polymer penetrates a few hundreds ring molecules and the axial polymer is terminally-modified with sterically-bulky compounds in order not to allow the ring molecules to escape from the string. The first PR has been synthesized by Harada et al. by using polyethylene glycol (PEG) as the axial polymers and α-cyclodextrin (αCD) as the ring molecules [1]. Figure 1 exhibits the structure of PR schematically.

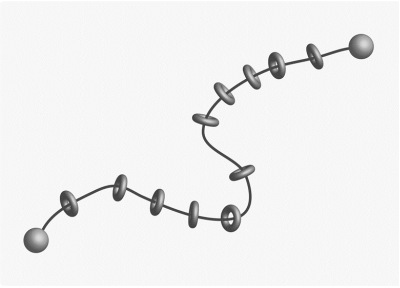

Figure 1: Schematic diagram of polyrotaxane.

The *mechanically interlocked* cyclic molecules in PR can slide and rotate on the axial chain. This additional kinetic freedom has been utilized to develop highly functionalized nanomaterials, such as three-dimensional cross-linked PR networks with movable cross-linkers [2].

We have investigated the structure and dynamics of PR by means of contrast variation small-angle neutron scattering (SANS) and contrast variation neutron spin echo (NSE) experiments [3]. With neutrons as probe, contrast variation techniques based on hydrogen/deuterium replacement can be used to modify the visibility of different component in the system. In order to study the properties of αCD threaded into PEG, partially deuterated αCD and PEG were synthesized for the preparation of PR with protonated αCD and deuterated PEG (αCD visible) and PR with deuterated αCD and protonated PEG (PEG visible). SANS and NSE experiments were performed for these two materials and additional fully protonated PR, then the obtained scattering intensities were decomposed into the partial scattering functions for each components, i.e., two self terms for αCD and PEG as well as one cross term reflecting the cross correlation between αCD and PEG. The detailed analyses of each partial scattering function led to reveal the structure and dynamics of αCD and PEG in PR with nanoscopic temporal-spatial resolution.

References

[1] A. Harada, J. Li, and M. Kamachi, Nature **356**, 325 (1992)
[2] Y. Okumura and K. Ito, Adv. Mater. **13**, 485 (2001).
[3] H. Endo et al., *Polymer J.* in press.