Division of Multidisciplinary Chemistry
– Polymer Materials Science –

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Scope of Research

The structure and molecular motion of polymer substances are studied, mainly using scattering methods such as X-ray, neutron, and light with intent to solve fundamentally important problems in polymer science. The main projects are studied on 1) the morphologies and the dynamics of self-assembling processes in block copolymers, 2) the hierarchical structures in crystalline polymer and rubber-filler systems, 3) the viscoelastic effects in glassy materials, 4) formation processes and ordering structures in polymer thin films.

KEYWORDS

Polymer Physics
Self Assembly
Hierarchical Structure
Polymer Properties
Softmatter

Selected Publications

Nishitsuji, S.; Takenaka, M., Viscoelastic Effects on Dynamics of Concentration Fluctuations in Semi-dilute Polymer Solution in the Good Solvent Regime, *Polymer*, **179,** 121622 (2019).

Hashimoto, T.; Amino, N.; Nishitsuji, S.; Takenaka, M., Hierarchically Self-organized Filler Particles in Polymers: Cascade Evolution of Dissipative Structures to Ordered Structures, *Polym. J.*, **51(2),** 109-130 (2019).

Ono, K.; Masuda, S.; Ogawa, H., Polymer-brush-decorated Colloidal Platelets: Precision Synthesis and Self-assembly, *Polym. Chem.*, **10,** 2686-2696 (2019).

Kinose, Y.; Sakakibara, K.; Ogawa, H.; Tsujii, Y., Main-Chain Stiffness of Cellulosic Bottlebrushes with Polystyrene Side Chains Introduced Regioselectively at the *O*-6 Position (in press).

In-situ Measurement of Self-assembling Block Copolymer Thin Film with GISAXS

Directed self-assembly (DSA) of block copolymers (BCPs) has been one of the most promising next generation lithography candidates. In order to use the DSA lithography for advanced semiconductor device manufacturing, we need to mitigate defects in the DSA materials and processes and to clarify the defect generation mechanism by using in-situ measurement of the self-assembling processes of BCPs. In this study, we, thus, observed the process of the self-assembly of a diblock copolymer by grazing incident angle small angler X-ray scattering (GISAXS) during heating process. Figure 1 shows a two-dimensional images of GISAXS for the self-assembling of Polystyrene-b-Poly methyl-methacrylate (PS-b- PMMA) (Mn = 29800-28700, Mn: number-averaged molecular weight) during heating processes at incident angles with 0.05° and 0.12°. An isotropic structure was induced near the glass transition temperature, and transformation to lamella structure was observed at 135 °C. The lamellar structure has a larger period than isotropic structure. The structure gradually grew according to the temperature, and it turned out that the whole film had a vertical lamellar structure at 145 °C. At higher temperature the long-range order of vertical lamellar structure was developed with temperature.

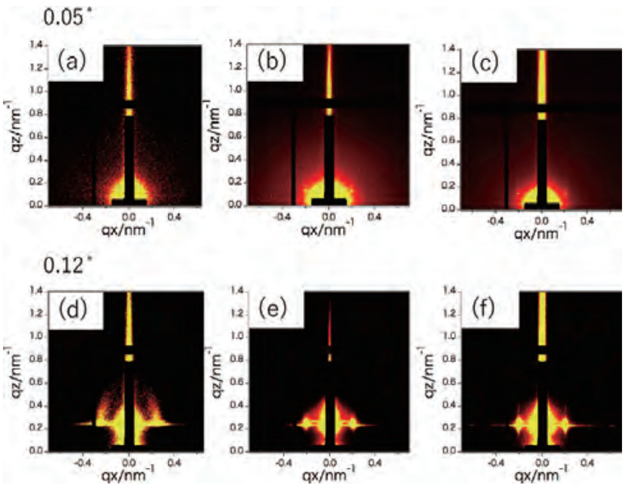

Figure 1. Two-dimensional GISAXS images of PS-b- PMMA in stepwise annealing process. these are at (a)135, (b)145, (c)180 °C with the incident angle of 0.12° and (d)135, (e)145, (f)180 °C with the incident angle of 0.05°.

Improving GISAXS-CT Images by Total Variation Minimization

Grazing incidence small-angle X-ray scattering (GISAXS) coupled with computed tomography (CT) has enabled visualizing the spatial distribution of nanostructures in thin films. The two-dimensional GISAXS images are obtained by scanning along the perpendicular direction to the X-ray beam at each rotation angle. Because the intensities at q positions contain nanostructural information, reconstructed CT images individually represent the spatial distributions of this information (*e.g.* size, shape, surface, characteristic length). These images are reconstructed from the intensities which are acquired at the angular intervals over 180°, though, it prolongs total measurement time. Moreover, this is often to increase with the radiation dosage and can cause damage to the sample. One way to reduce the overall measurement time is simply that scanning GISAXS measurement is performed along the perpendicular direction to the X-ray beam with a limited interval angle. Using filtered back-projection, CT images are reconstructed from the images of sinogram with the limited interval angles from Δ3° to Δ48° (FBP-CT images). However, these images are blurred and result in lower image quality. In this study, to optimize CT image quality, the total variation (TV) regularization is introduced to minimize sinogram image denoising and artifacts. In comparison with the FBP-CT images, we propose that the TV method can apply to down-sampling sinograms for improving CT images (TV-CT images).

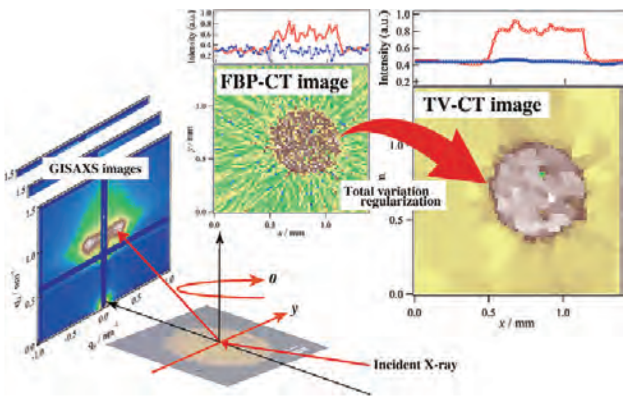

Figure 2. An overview of the FBP-CT and TV-CT images.