# Depicting Phase Separation in Polymer Blends by X-ray Phase Tomography

X-ray phase tomography using a crystal X-ray interferometer has been developed at beamline **BL20XU** [1]. This technique is attractive in the observation of weakly absorbing materials, and biological imaging results have been reported [2]. X-ray phase tomography reveals the distribution of the refractive index difference, which is approximately proportional to the mass density. The detection limit of the density deviation was evaluated to be a few mg/cm$^3$. This sensitivity is also attractive for the observation of polymer blend systems.

Blending polymers is commonly tried because the properties of plastic materials can be tailored to meet various demands, which is not possible with a single polymer. Many combinations of polymers are immiscible, and phase separation occurs in their blends and plays a significant role in determining their properties. Although phase-separated structures have been studied two-dimensionally for technical convenience, important quantities in three-dimensional space, such as the volume fractions of phases, the interfacial areas between coexisting phases, and interfacial curvatures, cannot be evaluated. It is clear that three-dimensional observation and analysis should be performed for the complete understanding of polymer blend morphologies.

X-ray phase tomography meets the demand, and we started research preliminarily with a test blend of polystyrene (PS) and poly(methyl methacrylate) (PMMA). A benzene solution containing 50 vol% PS and 50 vol% PMMA was freeze-dried. The resulting freeze-dried powder was annealed at 180 ºC in cylindrical holes (2 mm in diameter) made on a copper plate sandwiched in a melt-press machine.

Figure 1 shows the experimental setup for X-ray phase tomography. 17.7-keV X-rays were introduced into the X-ray interferometer, which was monolithically cut out of a silicon single-crystal ingot. Three crystal lamellae were formed, and one of those was characteristically thinned down to 40 μm to avoid spatial-resolution degradation [1]. A polymer blend sample was fixed on a rotation rod and immersed in water poured into a sample cell. X-ray interference patterns were recorded with a CCD-based X-ray image detector, whose effective pixel size was 3.14 μm.

Figure 2 shows a tomogram and a three-dimensional rendering view of the reconstructed data of the PS/PMMA blend [3]. Two phases in the blend were clearly depicted, and bicontinuous phase-separated structures were revealed. The volume ratio of the two phases was evaluated to be 52 : 48, in agreement with the blend composition. The bright and dark areas in Fig. 2(a) correspond to PMMA- and PS-rich phases, which were confirmed from the histogram of the reconstructed values shown in Fig. 3. The arrows indicate the values calculated for pure PS and PMMA. The peaks of the histogram appeared slightly inside the arrows, indicating that the PS-rich phase contained a small amount of PMMA and *vice versa*. This result quantitatively suggests that X-ray phase tomography can be used not only to depict structures but also to measure the composition of each phase, thereby allowing the determination of a phase diagram.

X-ray phase tomography is thus demonstrated to be an attractive technique for the three-dimensional observation of a polymer blend as is. Since no

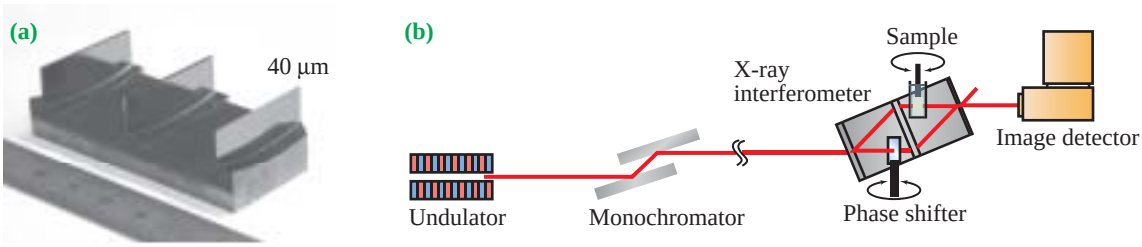


Fig. 1. X-ray interferometer having 40-μm lamella **(a)** and experimental setup for phase-contrast X-ray tomography **(b)**.

treatment is required for contrast enhancement, some intriguing experiments, such as the *in situ* three-dimensional tracing of phase separation and the observation of structural changes under mechanical stress, would be feasible. In addition, X-ray phase tomography can contribute to the observation of not only binary blends but also ternary and more complex blends.

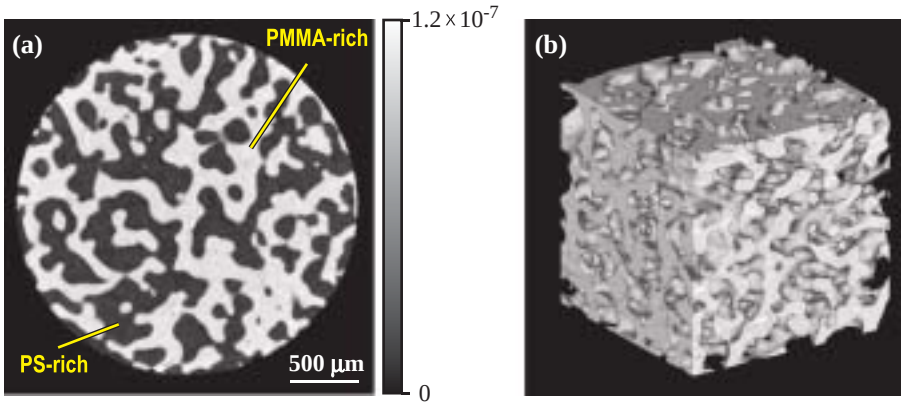


Fig. 2. Image of PS/PMMA blend reconstructed by X-ray phase tomography: **(a)** phase tomogram and **(b)** volume rendering view of reconstructed three-dimensional data, where PS-rich region has been made transparent.

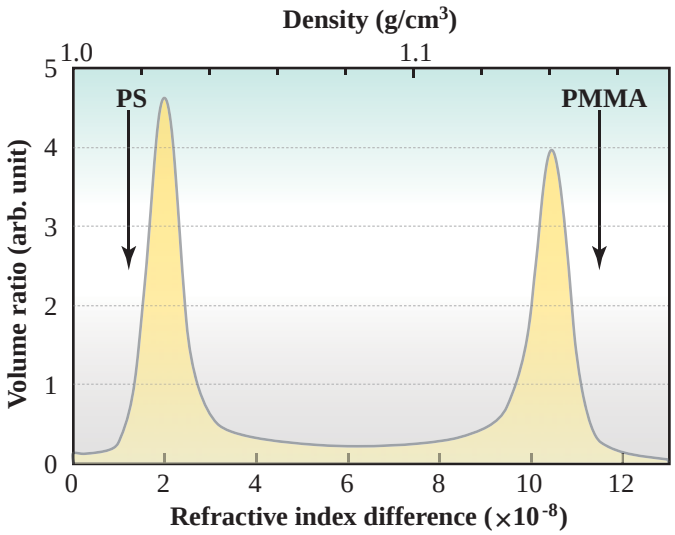


Fig. 3. Histogram of refractive index difference between sample and water. The refractive index differences of pure PS and PMMA are indicated by arrows.

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## References

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