TOPICS

Neutron Reflectometry for Studying Interfaces of Thin Films

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Neutrons are similar to X-rays in terms of materials analysis applications. However, since neutrons have high sensitivity to hydrogen and strong contrast between isotopes, many advances have been made in studies of soft materials by deuterium-labeling techniques, and more developments are expected in the analysis of engineering products such as polymer electrolyte fuel cells. Among the diversity of analysis techniques that are now available (for example, diffraction and small-angle scattering), reflectivity measurements can provide unique information about surfaces and interfaces. When specular reflectivity from a thin film is observed at grazing angles, a cross-section profile of the refractive index can be obtained, and this is representative of the distribution of the component materials. As an example of advances in this topic, we now present a study involving neutron reflectivity analysis of interfaces buried in thin films.[1)]

In order to use a simple model to investigate such interfaces, neutron reflectivity was measured on bilayer films with tunable cross-section profiles. The cross-section profiles were varied in terms of miscibility, and this can be changed with temperature when poly(4-trimethylsilylstyrene)(PTMSS) and polyisoprene (PI) are employed for the individual layers. It should be noted that the PTMSS was deuterium-labeled to enhance its contrast with respect to PI. From the phase behavior of the PTMSS/PI blends, it is predicted that the interface should diffuse as the temperature decreases. The bilayer films were prepared by picking up a top layer consisting of a PTMSS film floated on water onto a bottom layer of PI film spin-coated onto a Si substrate. Two of these bilayer films were subsequently annealed at 150°C and 190°C. Neutron reflectivity measurements were performed on the CRISP reflectometer in the ISIS Facility at the Rutherford Appleton Laboratory in the United Kingdom.

In **Fig. 1**, the neutron reflectivities of PTMSS/PI bilayer films annealed at 190°C (red) and 150°C (blue) are plotted against the scattering wave vector, where the solid lines are the best results of model fittings. The cross-section profiles employed for the fittings are shown in **Fig. 2**, and schematic pictures of the films are presented in **Fig. 3**. From the neutron reflectivity analysis by model fitting, it has been revealed that a broad interface with a thickness of about 30 nm occurs at 190°C, while this interface disappears due to interdiffusion at 150°C. The potential of neutron reflectometry to quantify the cross-section profiles will help the improvement of adhesion, the control of diffusion and so on. Finally, there is promise that industrial applications of neutrons will be triggered by the completion of J-PARC in the year 2007.[2)]

References

1) Harada, M., et al. : J. Polym. Sci. Part B : Polym. Phys., **43**(2005), 1486

2) Available from <http://j-parc.jp/index-e.html>

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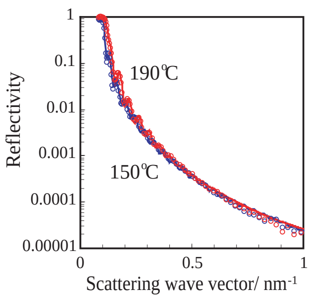

Fig. 1 Neutron reflectivity of PTMSS/PI bilayer thin films. Solid lines are results of the model fitting.

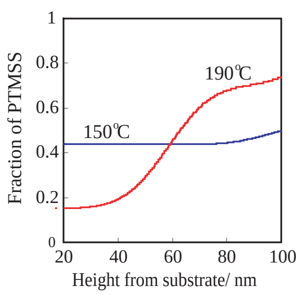

Fig. 2 Cross-section profiles of the fraction of the component polymer. The change in the 190 °C plot indicates the existence of an interface of about 30 nm in thickness. The constant fraction of 150 °C represents the mixing of the polymers.

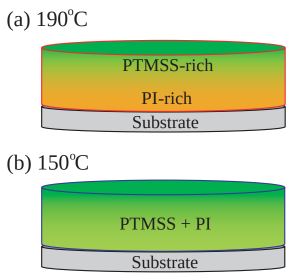

Fig. 3 Structures of PTMSS/PI bilayer thin films.
(a) Annealed at 190 °C. An interface is left between the PTMSS-rich layer and the PI-rich layer. (b) Annealed at 150 °C. PTMSS and PI inter-diffuses into a mixed layer.