In recent years, nanofibers are recognized as an exciting new class of material for various applications. One of the well-known methods for the production of nanofibers is electrospinning, however it can be said that the utilization of the preparation of islands-in-the-sea type bicomponent fibers (S/I fibers) for the production of nanofibers has an advantage of higher controllability of the diameter and mechanical properties of nanofibers [1]. In this technique, the sea component is dissolved into solvent after the formation of S/I fibers.

Regarding the preparation of bicomponent fibers, it can be considered that fiber formation behavior in the melt spinning process can be affected by the cross-sectional configuration even with the same composition of two constituent polymers. Possible factors for the origin of different spinning behavior in the vicinity of spinning nozzle are: (1) effect of flow history in the spinning die on the swelling behavior, and (2) effect of interfacial tension between the two components on the swelling behavior. Different degree of swelling alters the thinning behavior of the spin-line. Thinning behavior also can be affected by the cross-sectional configuration especially if the effect of the development of temperature distribution in the fiber cross-section on cooling is effective. Eventually, structure development and solidification behavior of the spin-line would be affected by the
above mentioned various factors, which also leads to the production of fibers of different characteristics.

In this study, bicomponent melt spinning of polystyrene (PS) and polypropylene (PP) with the PS : PP composition of 1 : 1 was performed to investigate the fundamental melt spinning behavior of the S/I fibers. For comparison, the sheath-core bicomponent fibers and the blend fibers of the same composition were also prepared. Considering the above mentioned factors which would affect the characteristics of resultant fibers, particular attention was paid for the fiber formation behavior in the vicinity of spinning nozzle in this research.

When a molten polymer is extruded through a capillary, the wall pressure at the exit is not zero but somewhat above the atmospheric pressure, which causes the phenomenon of post-extrusion swell (die-swell) or the Barus effect [2]. Origin of the Barus effect can be discussed from two perspectives. One is the elastic fluid flow, and the other is the first normal stress difference [3, 4]. The effect of elastic fluid flow is governed by the reduction ratio of the cross-sectional area of polymer flow in the die, while the effect of first normal stress difference originates from the shear flow in the capillary. In addition, effect of the interfacial tension at the interface of the two molten polymers also can be the origin of the swelling effect [5] especially in case of blend spinning. Some reports showed that phase morphology plays a decisive role on the performance of blend fibers. Zhu et al. [6-8] investigated the non-uniform morphologies of PP/PS and LDPE/PA6 blend fibers developed during a stable spinning process, and presented a hypothetical mechanism of non-uniform deformation and migration of dispersed droplets.

**Experimental**

**Spinning pack**

In this experiment, we used the spinning pack of the sheath-core (S/C) type and the islands-in-the-sea (sea-islands, S/I) type for the fiber formation. The S/I type spinning pack contained 1519 islands in its cross-section. Schematics illustrating the configurations of both types of spinning pack are shown in Fig. 1. The molten polymers passed through the distributors (a) and (c), and then flew into a nozzle of the same diameter of 1 mm in the spinnerets (b) and (d). The reduction ratio of the cross-sectional area from the position of the confluence of two polymer flows to the die exit for the S/C type spinning pack was 16 : 1, whereas that for the S/I type spinning pack was 3200 : 1. The reduction ratio of cross-sectional area in the production of S/I fibers was much larger than that in the production of S/C fibers.

**Materials and spinning conditions**

The materials used in this research were polystyrene (PS) of MFR 18 supplied by PS Japan Corporation and polypropylene (PP) of MFR 20 supplied by Prime Polymer Co., Ltd. Extrusion of the S/C and S/I bicomponent fibers as well as the blend fibers were carried out using a melt spinning apparatus schematically illustrated in Fig. 2. The PP and PS pellets were melted and extruded using extruders A and B, and fed into a spinning pack via metering pumps. Throughput rates of both PP and PS components were set at 2.5 g/min for the S/C, S/I and blend fibers. Extrusion temperatures of 230, 250, 270 and 290 °C were adopted. In this experiment, only the free-fall spinning was performed. A high speed camera was used at the image capturing speed of 765 fps for observing the swelling behavior of the S/C, S/I.
and blend spin-lines. Variation of fiber diameter along the spin-line was evaluated from the obtained images.

**Fiber cross-section**

Extruded blend fiber was quenched using a metal bar at a position near the spinneret and collected for the observation of the cross-sectional configuration. The captured fiber samples were put in liquid nitrogen to prepare a cross-section perpendicular to the fiber axis. Then PS in the cross-section was selectively stained using Ruthenium Tetroxide (RuO4). Observation of the fiber sample was carried out for the positions near the center of the cross-section using a transmission electron microscope (TEM, JEM-2010F, JEOL).

**Results and Discussion**

**Effects of extrusion temperature and type of spinning pack on die swell behavior**

The photographs of free-fall S/I and S/C spin-lines of PP/PS at four different extrusion temperatures are shown in Fig. 3. In this figure, photographs were enlarged along the direction perpendicular to the fiber axis for the better visibility of diameter variation along the spin-line. In these images, the distance of 0‒5 cm from the spinneret is covered, and the magnification in the perpendicular direction to the fiber axis is 3 times higher than that in the direction along the fiber axis.

Diameter of the spin-line increased after the extrusion because of the die-swell effect, which became more distinct with the decrease of extrusion temperature. If the S/I spinning is compared with the S/C spinning, diameter increase for the S/I spin-line was much larger than that for the S/C spin-line. This is considered to be due to the difference in the reduction ratio of the cross-sectional area from the position of the confluence of two polymer flows to the die exit.

Swelling index, the ratio of maximum spin-line diameter to nozzle diameter, was evaluated from Fig. 3 and plotted against extrusion temperature in Fig. 4. The results obtained for the single component spinning, in which the same polymer was fed into both extruders A and B, are also included in the figure. Spinning packs for the S/I and S/C spinning were applied.

![Fig. 3 Photographs of free-fall S/I, S/C spin-lines of PP/PS near the spinneret at different extrusion temperatures. Photographs were expanded three times only in horizontal direction for better visibility of diameter change. Original photograph for the S/I spin-line of 230 °C is also shown for comparison.](image)

![Fig. 4 Swelling indices of free-fall spin-lines at various extrusion temperatures. a) Single component spinning of PP and PS with S/C and S/I spinning packs, b) Bicomponent spinning of S/C and S/I fibers of PP/PS and PS/PP.](image)

In the single component spinning of both PP and PS, swelling indices with the use of S/I spinning pack were larger than those for the use of S/C spinning pack as in the case of PP/PS bicomponent spinning shown in Fig. 3. It can be said that these results supported the previous speculation regarding the effect of the reduction ratio of cross-sectional area on...
the swelling index.

The swelling indices for PS were larger than those for PP especially at low spinning temperatures. It is known that the increase of viscosity with the reduction of temperature is more significant for PS because of its higher activation energy than PP [9, 10]. Difference in the variations of the swelling indices with temperature for PP and PS can be explained simply considering the difference in the variation of viscosity with temperature.

In case of bicomponent spinning, when the position of polymer in the fiber cross-section was exchanged from S/C or S/I = PS/PP to PP/PS, swelling indices for the S/C spin-line became significantly smaller, whereas no substantial change was observed in the S/I spin-line. In the extrusion of S/C fibers through the spinning nozzle, the sheath component experiences higher shear strain rate in comparison with the core component. Therefore, it can be considered that higher swelling index for the S/C = PS/PP especially at lower extrusion temperatures is due to the higher viscosity of PS in comparison with that of PP. At a high temperature of 290 °C, swelling indices for PP and PS single component extrusion were similar both in the cases of S/I and S/C type spinning packs, whereas the swelling index for the S/C = PS/PP extrusion was significantly higher than that for the S/C = PP/PS extrusion. This result cannot be predicted from the result of single component extrusion. Thus, it was suggested that the swelling behavior originated from the PS component is more dependent on the shear rate.

Comparison of blend spinning and bicomponent spinnings

Cross-sectional images of the blend fibers prepared at three different extrusion temperatures using the S/C type spinning pack are compared in Fig. 5. At the extrusion temperature of 230 °C, PS phase was dispersed in the PP matrix, whereas with the increase of extrusion temperature to 250 °C, finer dispersed phase of PS as well as so-called salami structure with inclusion of PP dispersed phase in relatively large PS domain were observed. At 270 °C, phase morphology was almost reversed, and the morphology with dispersed phase of PP in PS matrix dominated the structure. These results suggested that the viscosity of PS was higher than that of PP at low temperature, and because of the difference in the activation energy for the temperature dependence of viscosity, viscosity ratio of PS to PP reduced and even became lower than unity at higher temperatures. These results coincided with the change of swelling index with temperature discussed in the previous section.

Observation of the spinning behavior of blend fibers revealed that the spin-line tended to be unstable especially at high extrusion temperatures. Stable extrusion of the blend fibers was observed only for the extrusion temperature of 230 °C. Photographs of the free-fall S/I, S/C and blend spin-lines of PP/PS

![Fig. 5](image1)

![Fig. 6](image2)
in the vicinity of the spinneret at the extrusion temperature of 230 °C are shown in Fig. 6. As in the cases of single-component and bicomponent spinning, die swell was more significant in the spinning with the S/I type spinning pack. In addition, it was found that in the case of blend extrusion, the swelling behavior continues to downstream and the maximum diameter of swelling appeared at a position further away from the spinneret. Diameter profiles of the spin-line analyzed from these photographs are shown in Fig. 7. The peak position of swelling appeared at positions close to the spinneret for the S/C and S/I bicomponent spinnings, whereas the spin-line diameter continuously increased to the downstream and the maximum diameters appeared at positions far away from the spinneret in the case of blend spinning.

At the temperatures of 250, 270 and 290 °C, extruded spin-line was unstable with a periodic change of diameter. The instability became more significant and the period of diameter change became shorter with the increase of extrusion temperature. A series of photographs for one period of diameter change was prepared for each extrusion temperature. The results for the extrusion temperatures of 250 and 290 °C are shown in Fig. 8. For the extrusion temperature of 250 °C, periodic diameter variation with the period of around 16.2 s can be seen from the variation of diameter at around 5 cm. The minimum and maximum swelling indices judged at this position were 1.46–2.27, which were significantly larger than the result for 230 °C, around 1.45. In other words, swelling effect tended to be enhanced with the increase of temperature. At the extrusion temperature of 290 °C, spin-line became significantly unstable, in that there was a formation of a thick part of the spin-line near the spinneret, the thick part continuously flowed down to the downstream, and afterward a new thick part appeared near the spinneret. The period of such diameter variation was around 12.2 s. By combining the results in Fig. 8-b) and c), diameter profiles of the spin-line of around 0–10 cm was estimated as shown in Fig. 9. It is
interesting to note that along with the shift of the peak of swelling to downstream, there was a continuous increase of the maximum diameter. This result indicates that the spin-line near the peak of swelling locally retracted even though there should be a thinning of the overall spin-line.

These results suggested that the interfacial tension between two components is the key factor for the occurrence of the swelling of the spin-line in the spinning of blend fibers. During the flow in the capillary, there can be an elongation of dispersed phase, which causes the increase in interfacial area between the two polymers. For the reduction of generated excess energy at the interface, the elongated phase needed to be retracted to reduce the interfacial area and caused the swelling of the flow. It seems that the swelling caused by the interfacial free energy tends to appear in downstream in comparison with the swelling caused by the viscoelastic effect. On the other hand, enhanced swelling effect at high temperatures can be explained based on the difference in the temperature dependence of viscosity and interfacial tension. It is known that the decrease of viscosity with the increase of temperature is more significant than the reduction of interfacial tension. For example, activation energies for temperature dependence of viscosity for PS and PP at the temperature of 150-290 °C are reported to be around 77.9 and 15.7 kJ/mol, respectively [9, 10], whereas that for temperature dependence of PP/PS interfacial free energy at the temperature of 200-260 °C are around 3.6 kJ/mol [11]. This is the reason for the enhancement of swelling at higher temperatures.

Conclusions

Effect of flow history in the sheath-core type spinning pack and sea-islands type spinning pack was investigated paying particular attention to the behavior of polymer flow in the vicinity of spinneret. Magnitude of die-swell decreased with the increase of extrusion temperature. The swelling was more significant for the S/I spin-line in comparison with that for the S/C spin-line. When the S/C and S/I components were exchanged from PS/PP to PP/PS, the magnitude of swelling of S/C spin-line decreased whereas that of S/I spin-line did not show any significant change. On the other hand, magnitude of die-swell for the blend spinning was larger than that for the S/I and S/C spinnings and increased with the increase of extrusion temperature. The distance from the spinneret surface to the position of maximum spin-line diameter increased in the order of S/C<S/I<blend. These results suggested that the swelling effect in the S/C and S/I spinning is governed by the viscoelastic effect whereas that in the blend spinning is caused mainly by the interfacial tension between the two components.

Acknowledgements

This work was financially supported in part by Program for Leading Graduate Schools “Academy for Co-creative Education of Environment and Energy Science”, MEXT, Japan.

References