The Study on Die Swell, Fiber Length Distribution, and Crystallinity of PP Composite Through Extrusion

WEN-YEN CHIU and GOANG-DING SHYU, *Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan, Republic of China*

Synopsis

Die swell of PP and glass fiber reinforced PP (GFRPP) through an extruder was investigated. It was observed that the die swell decreased as (1) the temperature increased, (2) the fiber content increased, and (3) the screw speed was lowered. Fiber length distribution in GFRPP was measured. The average fiber length would increase as (1) the fiber content decreased, (2) the temperature increased, and (3) the screw speed was around 25 rpm. The crystallinity of extrudate was measured by DSC and found to be independent of the screw speed and the fiber content.

INTRODUCTION

The die swell of extrudate for viscoelastic fluids would vary with several factors as: geometry of die, flow rate, temperature, molecular weight (MW) and distribution (MWD) of fluids, and additives in fluids. Han and Kim[1] found that the die swell of extrudate decreased with increasing the aspect ratio of die. Graessley et al.[2] observed the die swell of PS and found it increased with increasing the MW and MWD of PS. Huang and White[3] found the die swell through slit die larger than that through round-hole die for the same polymer material. As for the die swell of fiber-reinforced composites, Chan et al.[4] concluded that the primary normal stress coefficient and viscosity of fiber reinforced PS and PE would increase as the fiber content increased, but the die swell of composite was largely reduced. In this work, first we investigated the die swell of PP through an extruder with a round-hole die. The variations of die swell of PP with the extrusion temperature, screw speed, pressure drop over die and cooling way on extrudate were measured and discussed. Second, we investigated the die swell of GFRPP through an extruder with a slit die. The variations of die swell of GFRPP with the fiber content, screw speed, and extrusion temperature were studied.

The mechanical strength of composite was affected by the fiber length and distribution over polymer matrix. Stade[5] observed the variation of fiber length distribution with processing conditions. Kitano and Kataoka[6] studied the effect of fiber length on viscosity and mechanical strength of PP composite through different blending ways. Von Turkovich and Erwin[7] reported that no effect was found on the fiber length distribution by varying the fiber content in composite. In this paper, we investigated the variation of fiber length distribution with extrusion conditions for GFRPP.

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The crystallinity of extrudate, which changed with the processing conditions, affected the mechanical strength of material very much. Pae and Sauer[8] studied the behavior of crystallization of PP upon different annealing ways. In this work, we examined the crystallinity of GFRPP extrudates under different extrusion conditions.

EXPERIMENTAL

Materials

(1) PP: MI = 3.0 g/10 min
(2) Glass fiber (GF): 6 mm chopped strand, d_f = 15–22 μm.

GFRPP Preparation

$$\text{PP} + \text{GR} \xrightarrow{\text{extruder}} \text{GFRPP}$$

PP pellets were dried at 80 °C for 2–3 hs. Glass fibers were dried at 80 °C for 1 h. Both were mixed and blended thoroughly in different proportions through an extruder with the screw speed of 25 rpm and the temperatures of 200 °C in feed zone, 210 °C in compression zone, 230 °C in metering zone, and 230 °C in die. The extrudates from the extruder were then cut into small pellets as the feed material in the die swell experiments. The symbol GFRPP 5 was used to denote the GFRPP material with 5 wt % glass fiber.

Die swell through an Extruder

An extruder equipped with a screw (L/D = 24, D = 35 mm) and either a round-hole die ($5^{\text{o.d.}} \times 35\ \text{mm}^{l}$) or a slit die ($64.5^{l} \times 30^{w} \times 2\ \text{mm}^{t}$) was used to study the die swell of PP and GFRPP. With changing the extrusion temperature and screw speed, PP or GFRPP pellets were fed in the extruder. Part of the extrudates came out in silicone oil at 185 °C for 10 mins, and then cooled in air (say annealed extrudates). Part of them was quenched in water (say quenched extrudates). The dimension of extrudate was measured by a micrometer. For round-hole die, the diameter of extrudate (D) was determined by the ratio of its peripheral length to π. Die swell (χ) was then defined as the ratio of D to the diameter of die (D_0). For slit die, the die swell (χ) was defined as the ratio of the largest thickness over the extrudate to the thickness of die. The variation of die swell of extrudate under various extrusion conditions were observed.

Fiber Content and Fiber Length Distribution in GFRPP Extrudates

The weighed GFRPP extrudate was heated at 650 °C for 30 min to burn out PP, and the remaining fibers were weighed to determine the fiber content. Then the fibers were wetted in glycerine and spread on a glass plate, and the length distribution was observed through a microscope. The lengths of at least 500 fibers were measured carefully in each experiment.

Crystallinity Determination

DSC was used to determine the crystallinity of extrudates. The crystallinity of GFRPP was calculated as $\Delta H_f/(\Delta H_f^0 \cdot W_m)$. ΔH_f was the fusion heat per unit weight of extrudate, which was obtainable experimentally from the endothermic peak on DSC diagram. ΔH_f^0 was the ideal fusion heat of PP in pure crystalline state, which was read as 50.0 cal/g from reference.[9] W_m was the weight fraction of PP in GFRPP. The variation of crystallinity of extrudate under various extrusion conditions was examined.

RESULTS AND DISCUSSION

Figure 1 showed the die swell of PP from a round-hole die extruder at different screw speeds and different extrusion temperatures. The three zones and die in extruder were all kept at isothermal condition in each case. The die swell increased with increasing the screw speed, but decreased with increasing the extrusion temperature. The higher screw speed induced higher stress and elastic response on the polymer fluid. The relaxation of stress and the elastic memory were of course the main causes for the larger die swell observed in the case of higher screw speed. On the other hand, higher temperature reduced the viscosity, normal stress, and elastic response of the polymer, so an opposite effect on the die swell showed. Figure 2 showed that the die swell of PP in Figure 1 vs. the pressure drop over die could be lumped in a linear line at several temperatures. Figure 3 showed the influence of thermal history on the die swell of PP from round-hole die, where T_1, T_2, T_3, and T_D were the temperatures in the feed zone, in the compression zone, in the metering zone, and in the die of extruder respectively. Larger die swell was found in the upper line, where PP experienced lower temperatures in T_1 and T_2 before it came out through the die. The effect of thermal history on the die swell through extruder was very significant. When PP flew through the feed and compression zones with lower temperatures, the induced normal stress and the left elastic memory were higher on the extrudate, so that a higher die swell

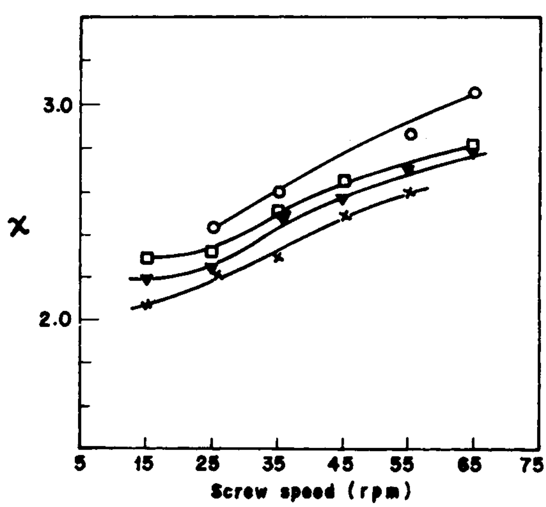

Fig. 1. Die swell of PP from a round-hole die extruder at different screw speeds: (○) 210 °C; (□) 220 °C; (▽) 230 °C; (×) 240 °C.

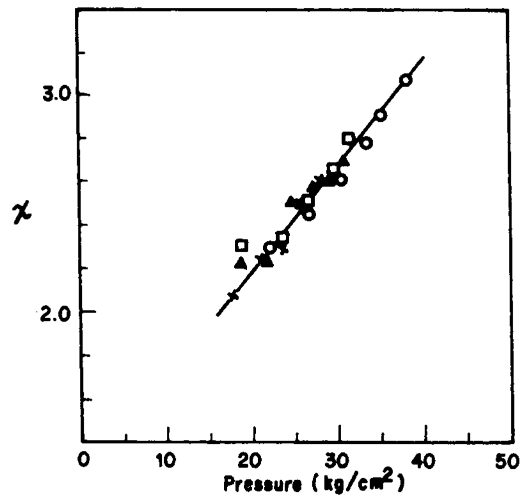

Fig. 2. Die swell of PP from a round-hole die extruder vs. pressure drop over die: (○) 210° C; (□) 220° C; (△) 230° C; (×) 240° C.

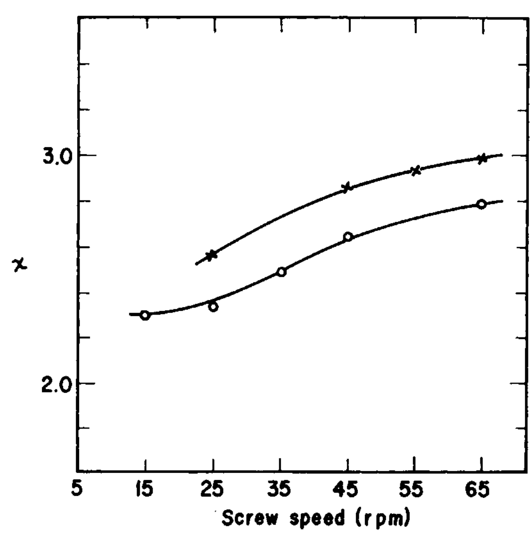

Fig. 3. Influence of thermal history on the die swell of PP from round-hole die. Extrusion temperature (T_1–T_2–T_3–T_D): (×) 180–210–220–220° C; (○) 220–220–220–220° C.

was observed. Figure 4 showed the effect of cooling way for the extrudate on its die swell through the round-hole die extruder. The annealed extrudates showed much larger die swell than the quenched extrudates. Because a large amount of stress in quenched extrudates was frozen in, the relaxation of stress, hence the die swell, was very limited. Figure 5 showed the die swell of GFRPP with different fiber contents vs. screw speed from a slit-die extruder, where the temperature of extruder was set as: 200° C in the feed zone (T_1), 220° C in the compression zone (T_2), 230° C in the metering zone (T_3), and 230° C in the die (T_D). It was reported that[10] the elastic recovery of the

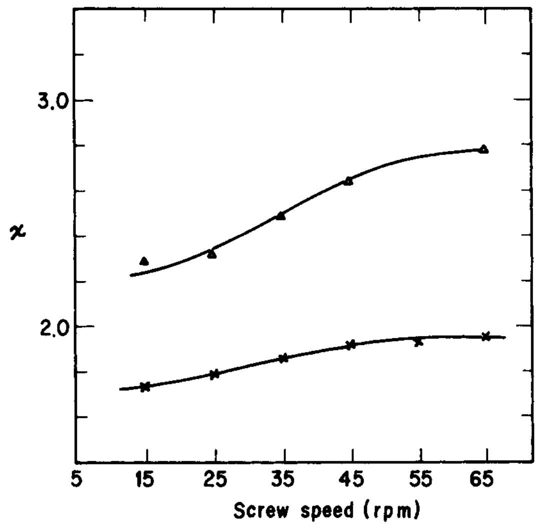

Fig. 4. Die swell of PP through round-hole die for annealed extrudates (△) and quenched extrudates (×). Extrusion temperature (T_1–T_2–T_3–T_D): 220–220–220–220° C.

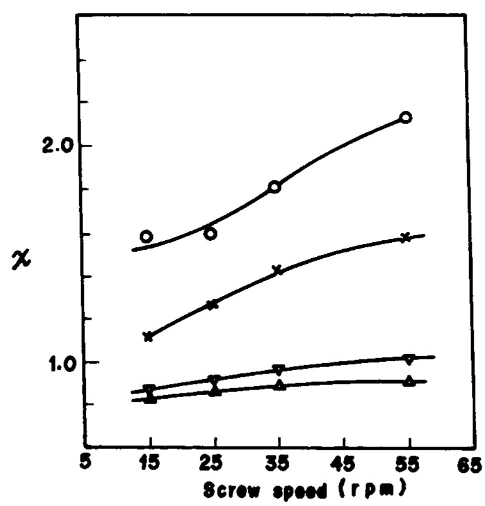

Fig. 5. Die swell of GFRPP vs. screw speed from a slit-die extruder: (○) PP; (×) GFRPP 5; (▽) GFRPP 15; (△) GFRPP 25. Extrusion temperature (T_1–T_2–T_3–T_D): 200–220–230–230° C.

polymer mainly came from the recovery of entanglements of polymer molecules. In GFRPP extrudates, the fiber's distribution in the PP matrix would retard the recovery of entanglements of PP molecules while flowing out of the die. So the die swell of GFRPP decreased largely as the fiber content increased to 15 wt %.

Fibers would never distribute uniformly over GFRPP extrudates. It was found that the fiber content in the central part of extrudate from a slit-die was a little less than that at the edge as seen in Figure 6. The fiber length in GFRPP processed through an extruder twice was much shorter than that processed once. Figures 7 and 8 showed the fiber length distribution of

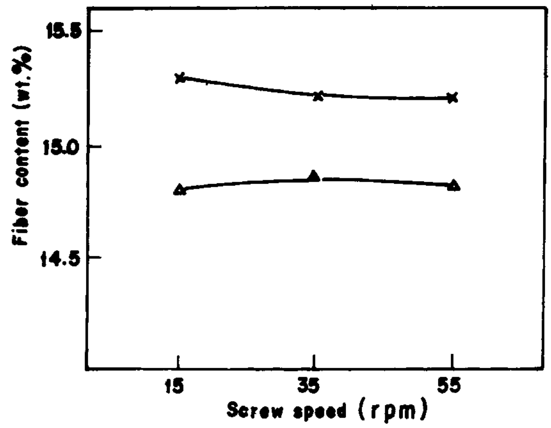

Fig. 6. Fiber contents in GFRPP 15 slit extrudate at different positions: (△) center; (×) edge. Extrusion temperature (T_1-T_2-T_3-T_D): 200-220-230-230° C.

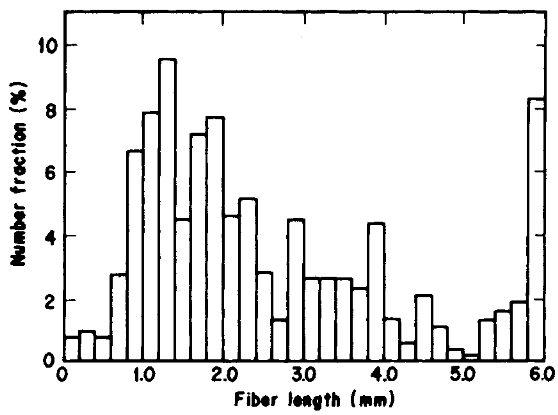

Fig. 7. Fiber length distribution of GFRPP 5 extruded once. Extrusion temperature [T_1-T_2-T_3-T_D (round-hole die)]: 200-210-230-230° C. Screw speed: 25 rpm. $\bar{l}_n$ = 2.66 mm, $\bar{l}_m$ = 3.66 mm.

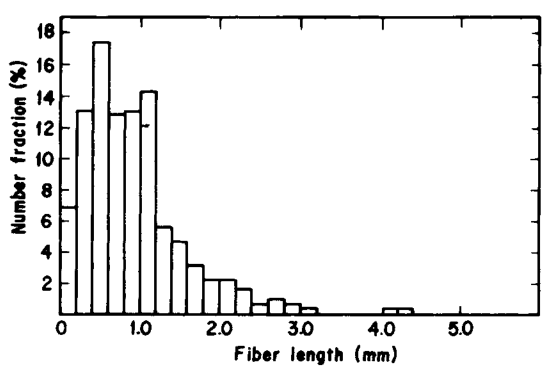

Fig. 8. Fiber length distribution of GFRPP 5 extruded twice. Extrusion temperature (T_1-T_2-T_3-T_D): (I) 200-210-230-230° C (round-hole die); (II) 200-220-230-230° C (slit die). Screw speed: (I) 25 rpm; (II) 15 rpm. $\bar{l}_n$ = 0.96 mm; $\bar{l}_w$ = 1.33 mm.

GFRPP 5 extruded once and twice, respectively. The number average fiber length ($\overline{l_n}$) in Figure 7 was about 2.66 mm, while in Figure 8 it was about 0.96 mm. In the extruder, the high shear and friction forces upon fibers caused the breakage of fibers. There were several other factors to influence the fiber length in extrudate, such as screw speed, temperature, and fiber content. Table I showed the influence of screw speeds and fiber contents on the fiber length, where $\overline{l_n}$ and $\overline{l_w}$ were the number and weight average fiber length. The average fiber length decreased with increasing the fiber content, and had larger values around 25 rpm screw speed. When the fiber content increased, fibers contacted more, which produced more friction force and made the fibers broken more seriously. As for the reason for why fibers were longer in the case of 25 rpm screw speed, it probably came from two factors: (1) the residence time of GFRPP in the extruder and (2) the shear stress in the extruder. When screw speed increased, the residence time decreased, but the shear stress increased. Their effects on the fiber breakage were in the opposite trend. So we found the fiber lengths maximum around 25 rpm screw speed. Table II showed the influence of extrusion temperature on the fiber length in GFRPP 15. The higher extrusion temperature would reduce the visocisty and shear stress of the polymer fluid, which made the fiber lengths longer in the extrudates.

By examining the DSC endothermic peaks of GFRPP extrudates, it was found that the annealed extrudate showed an additional small peak around 150–155 °C, slightly lower than the main melting temperature around 170 °C which indicated that new crystallites formed during the annealing process. Figure 9 showed the crystallinity of GFRPP vs. fiber content. The annealed

TABLE I
Influence of Screw Speeds and Fiber Contents on Fiber Length[a]

	rpm	$\overline{l_n}$ (mm)	$\overline{l_w}$ (mm)
GFRPP 25	15	0.70	0.98
	25	0.83	1.07
	55	0.69	0.86
GFRPP 15	15	0.70	1.05
	25	0.88	1.26
	55	0.64	1.02

[a] 1 mm = 10^{-3} m, 1 rpm = $1/60\ s^{-1}$; extrusion temperature (T_1–T_2–T_3–T_D): 200–220–230–230 °C; sample position: central part of the slit die extrudate.

TABLE II
Influence of Temperature on Fiber Length in GFRPP 15[a]

Extrusion temperature (°C)					
T_1	T_2	T_3	T_D	$\overline{l_n}$ (mm)	$\overline{l_w}$ (mm)
200	220	230	230	0.73	0.96
200	220	240	240	0.83	1.13

[a] Screw speed: 15 rpm; sample position: edge of the slit die extrudate.

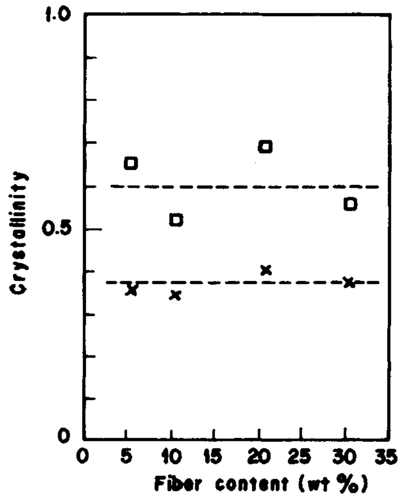

Fig. 9. Crystallinity of GFRPP extrudate vs. fiber content from a slit-die extruder: (□) annealed extrudate; (×) quenched extrudate. Extrusion temperature (T_1–T_2–T_3–T_D): 200–220–230–230° C. Screw speed: 35 rpm.

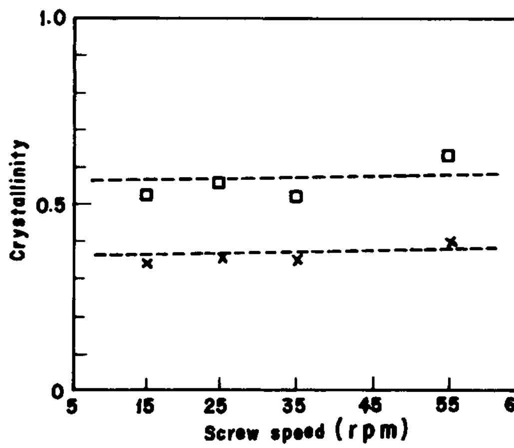

Fig. 10. Crystallinity of GFRPP5 extrudate vs. screw speed from a slit-die extruder: (□) annealed extrudate; (×) quenched extrudate. Extrusion temperature (T_1–T_2–T_3–T_D): 220–220–230–230° C.

extrudate had higher crystallinity as predicted. Figure 10 showed the crystallinity of GFRPP 5 extrudate vs. screw speed. There were no effects found on crystallinity with varying either the screw speed or the fiber content.

CONCLUSION

Die swell of PP and GFRPP from an extruder was investigated. The extrudate was annealed slightly above its melting point to allow complete relaxation of stresses. It is observed that the die swell ratio decreased as (1) the temperature increased, (2) the fiber content increased, and (3) the screw speed was lowered. Under different temperatures, the plot of die swell vs. pressure is approximately a straight line.

Fiber content and fiber length distribution of GFRPP after extrusion were determined. Burning the extrudate in an oven at 600°C for about 30 min, the weight percentage of glass fiber was obtained gravimetrically. It is found that fiber content in the central part of the extrudate is a little less than that at the edge. Fiber length distribution was measured by a microscope. The average fiber length in GFRPP processed through an extruder twice is much shorter than that processed once. Increasing the extruder temperature will increase the average fiber length. The fiber length will decrease as the fiber content increases. It is seen that when the speed of the screw is 25 rpm, the average fiber length is the longest.

The crystallinities of quenched and annealed extrudates were measured by DSC. It is found that crystallinity is independent of screw speed and fiber content. The annealed extrudates showed an additional endothermic peak or shoulder at 150–155°C on the DSC diagram.

References

1. C. D. Han and K. U. Kim, *Polym. Eng. Sci.*, **11**, 395 (1971).
2. W. W. Graessley, S. D. Glasscock, and R. L. Grawley, *Trans. Soc. Rheol.*, **14**, 519 (1970).
3. C. D. Huang and J. L. White, *Polym. Eng. Sci.*, **9**, 609 (1979).
4. Y. Chan, J. L. White, and Y. Oyanagi, *J. Rheol.*, **22**, 507 (1978).
5. K. Strade, *Polym. Eng. Sci.*, **17**, 50 (1977).
6. T. Kitano and T. Kataoka, *Rheol. Acta*, **19**, 753 (1980).
7. Richard Von Turkovich and L. Erwin, *Polym. Eng. Sci.*, **23**, 743 (1983).
8. K. D. Pae and J. A. Sauer, *J. Appl. Polym. Sci.*, **12**, 1901 (1968).
9. R. Greco, H. B. Hopfenberg, E. Martuscelli, G. Ragosta, and G. Demma, *Polym. Eng. Sci.*, **18**(8), 654 (1978).
10. Y. Chan, J. L. White, and Y. Oyanagi, *Polym. Eng. Sci.*, **18**, 268 (1978).

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