Effects of Fiber Characteristics on the Mechanical and Rheological Properties of Poly(butylene terephthalate)/Glass Fiber Composites

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ABSTRACT: Morphological, mechanical, and rheological properties of poly(butylene terephthalate) (PBT)/glass fiber (GF) composites and their fiber lengths after extrusion were studied, along with the molecular weight of PBT and the sizing effect of the fiber. From the molecular weight effect of PBT on mechanical properties, the lower molecular weight PBT composites prepared showed higher flexural and tensile strengths compared to the PBT/GF composites prepared with a higher molecular weight PBT when the fiber content was over 20 wt %. This behavior was mainly due to the increased fiber length of the PBT/GF composite prepared with lower molecular weight PBT. For the PBT composites with GF coated with organic materials having polarity similar to that of the PBT matrix, better mechanical properties were observed due to higher bonding forces between the matrix and fibers. Due to the effect of fiber length on the complex viscosity of the PBT/GF composite and its resulting high elasticity, the shear thinning behavior was more significant for the composites containing long fibers than in composites containing short fibers. From the above results, it is suggested that the optimum mechanical properties of the fiber-reinforced polymer composite can be achieved by varying the processing conditions such as molecular weight of the matrix polymer, surface treatment of the fiber, and feeder location of the fiber. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 116: 3005–3012, 2010

Key words: polymer composite; morphology; mechanical property; rheology; poly(butylene terephthalate); glass fiber

INTRODUCTION

In engineering polymer industries, poly(butylene terephthalate) (PBT) has been widely used as a matrix polymer in the production of fiber-reinforced composites to improve physical properties of end products, due to its desirable properties such as good mechanical properties, chemical resistance, moldability, and high crystallization rate.[1–10] Glass fiber (GF) reinforced PBT composites have been investigated by many researchers, who have reported their mechanical properties,[11,12] crystallization kinetics, and thermal properties[13–16] as well as those of rubber toughened PBT/GF composites.[16–18] Metal parts have been replaced by polymer composites reinforced by GF due to their moderate cost and light weight.[8] The presence of GF in the matrix is associated with an enhancement in stiffness and strength.[9,10]

Recently, the market for high filler loaded engineering plastics containing about 50% filler has shown a tendency to increase; accordingly, increased attention has recently been paid to the development of high filler loaded polymer composites. For example, the PBT/GF (50 wt %) composite is mainly used for the outside mirror of automobiles, and can be utilized in a nonhalogen flame retardant PBT system. Other polymer systems of poly(ethylene terephthalate),[19–23] nylon,[24,25] poly(phenylene sulfide),[26] and polyolefins[26–29] with fillers such as GF, graphite, and carbon nanotubes can be coordinated. However, problems associated with surface troubles during the extrusion process can occur when the content of GF reaches about 50% by weight.

In this study, PBT/GF composites with a GF content of up to 50 wt % were prepared by melt extrusion. The effects of fiber length after extrusion on the mechanical and rheological properties of the PBT/GF composites were studied. The effects of molecular weight of the PBT and surface treatment of the fiber on the mechanical properties of the PBT/GF composites were also studied.

TABLE I
Characteristics of Poly(butylene terephthalate) Used in this Study

Samples	M_w	T_g (°C)	T_m (°C)	Melt index (235°C)
PBT (25)	25,000	56.2	225	45 ± 4.5 (g/10 min)
PBT (33)	33,000	62.1	225	27 ± 4.5 (g/10 min)

EXPERIMENTAL

Materials

The polymers used in this study were supplied from a commercial source, and two different types of PBT were provided by LG Chemical. (Seoul, Korea). The characteristics of the PBT are summarized in Table I. The molecular weight of the PBT was provided by the supplier. PBT (25) and PBT (33) denote PBTs of weight-average molecular weighst of 25,000 and 33,000, respectively. For the measurement of T_g, a dynamic mechanical thermal analyzer was used at the frequency of 6.28 rad/sec and 0.05% strain. The sample was heated from 30 to 250°C. For the T_m, differential scanning calorimetry was used. The heating rate and cooling rate of the sample were 10 and 20 K min^{-1}, respectively, and the sample was heated from 50 to 300°C. Temperature calibration was performed using indium ($T_m = 156.6$°C). Two different types of GFs were used as reinforcing agents, and the fiber was E-glass type. The glass fiber (RH) denoted the GF which was treated with aromatic hydrocarbon compounds representing polarity, and the glass fiber (LH) denoted the GF which was treated with aliphatic hydrocarbon compounds representing nonpolarity. GFs were supplied by Owens Corning Korea (Seoul, Korea). Specific gravity, length and diameter of the GFs were provided by supplier. The characteristics of GFs used in this study are summarized in Table II.

Composite preparation

PBT/GF composites were prepared by a twin screw extruder. The GF contents used in this study varied from 0 to 50 wt %. The diameter of the twin screw was 40 mm and the screw ratio of length to diameter was 40 : 1 (Hankook E. M., Seoul, Korea). The temperatures of the feeding zone and die zone of the extruder were 255 and 260°C, respectively. The screw speed of the extruder was set at 250 rpm. GF feeding was performed at the first side feeder (1), second

side feeder (2), and main feeder (3), and the distances from the tip of the die were 87, 127, and 176 cm, respectively. Table III lists the operating conditions for the production of PBT/GF composites (samples A to E) with molecular weight of PBT, surface treatment of GF and feed location of the GF.

Mechanical properties

The tensile and flexural strength of PBT/GF composites were investigated under ambient conditions using a universal testing machine (Instron 4467, Norwood, MA). Tensile test was conducted according to the ASTM D-638, and wedge action grip was used for the test. The crosshead speed was 5.0 mm/min. The flexural test was performed according to the ASTM D-790. The span length and the speed of cross-head movement were 40 mm and 2.8 mm/min, respectively. For tensile and flexural strength test, five specimens were measured and averaged.

Morphology

The morphologies of the PBT/GF composites were determined by field emission scanning electron microscope (Hitachi S-4300, Tokyo, Japan). The samples were fractured at the cryogenic condition and coated by Pt before scanning. The accelerating voltage was 15 kV. To measure the fiber length of the composites, we have used o-chlorophenol as a solvent to dissolve the polymers, then the fiber length was measured using a microscope. In this study, a method similar to the ISO 22,314 specifications for measuring the fiber length was applied, except that a furnace was used to burn out the polymers in the composites.

Rheology

Rheological properties of the PBT/GF composites were measured using an advanced rheometric expansion system (ARES, Rheometrics, New Jersey, NY) in oscillatory shear at 8% strain in the parallel-plate arrangement with a 25 mm plate. The frequency sweep from 0.1 to 100 rad/s was performed at 220°C under dry nitrogen conditions. For the ARES measurements, the samples were tested within the linear viscoelastic strain range.[30]

TABLE II
Characteristics of Glass Fibers Used in this Study

Samples	Specific gravity	Length/diameter	Fiber surface treatment
Glass fiber (RH)	2.60	4 mm/14 μm	Treated with aromatic hydrocarbon
Glass fiber (LH)	2.60	4 mm/14 μm	Treated with aliphatic hydrocarbon

TABLE III
Sample Preparation Conditions of Poly(butylene terephthalate) (PBT)/Glass Fiber (GF) Composites

Sample code	PBT	GF	Feeder location of glass fiber
A	PBT (33)	GF (RH)	Feeder 1
B	PBT (33)	GF (RH)	Feeder 2
C	PBT (33)	GF (RH)	Feeder 3
D	PBT (25)	GF (RH)	Feeder 1
E	PBT (33)	GF (LH)	Feeder 1
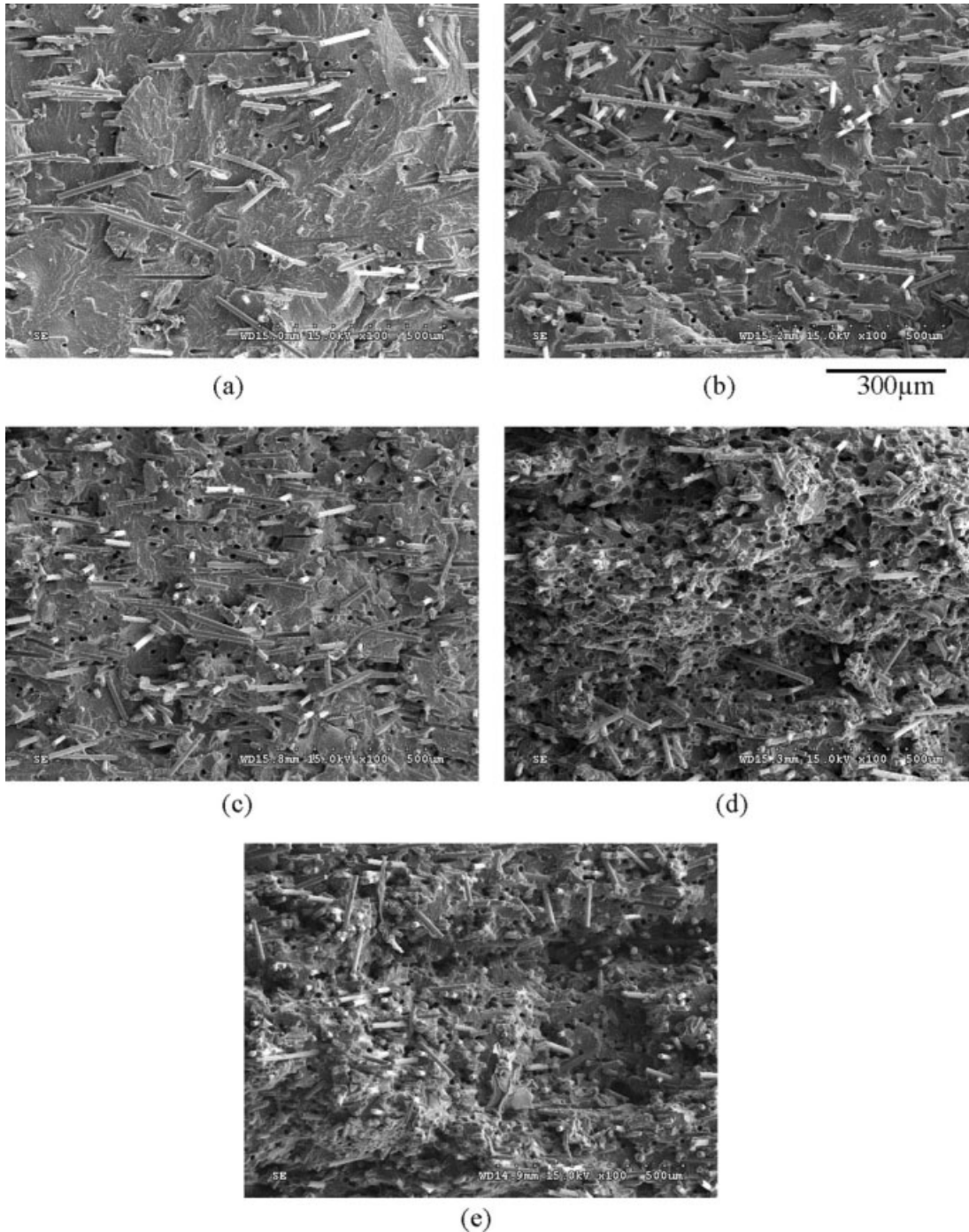
Figure 1 Scanning electron micrographs of cryogenically fractured cross-sectional surfaces of poly(butylene terephthalate) (33)/glass fiber (RH) composite (sample A) with glass fiber content (wt %): (a) 10, (b) 20, (c) 30, (d) 40, (e) 50.
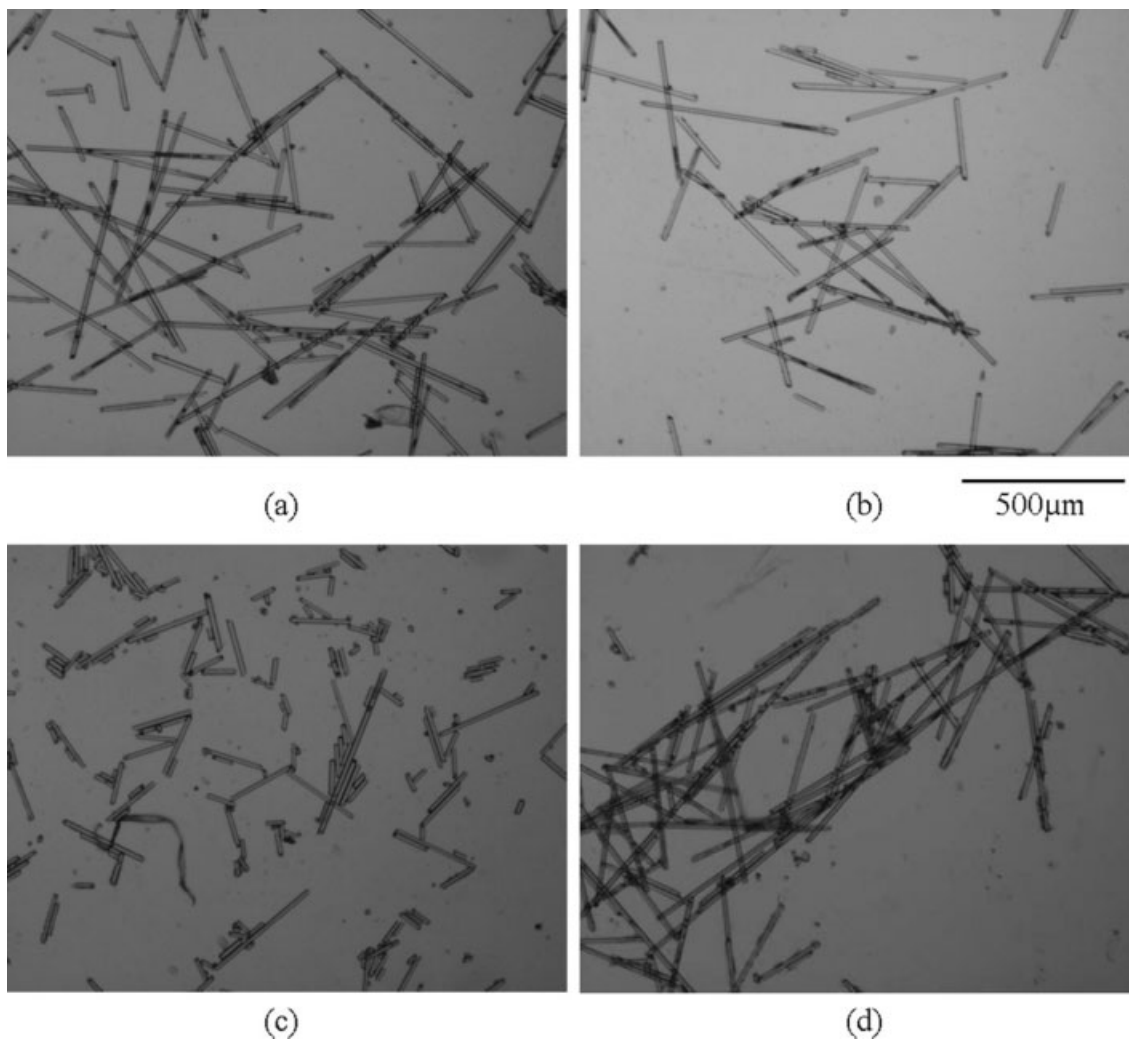
Figure 2 Polarized micrographs of glass fiber after extrusion of poly(butylene terephthalate)/glass fiber (60/40 wt %) composite: (a) sample A, (b) sample B, (c) sample C, (d) sample D.

RESULTS AND DISCUSSION

SEM of PBT/GF composites

Figure 1(a–e) shows scanning electron micrographs of cross-sectional surfaces of the PBT (33)/GF(RH) composites (sample A) with GF contents of 10, 20, 30, 40, and 50 wt %, respectively. Figure 1 shows that the GFs are somewhat oriented in the composites containing up to 30 wt % of GF. For the composites with 40 and 50 wt % GFs, however, random orientation and aggregation of the fibers are observed in the PBT/GF composites because of the high loading of the fibers. The degree of fiber dispersion in the polymer matrix is one of the most important factors affecting the mechanical properties of the composites. The mechanical strength of the PBT/GF composites with the fiber content and feed location of the fiber will be discussed in the next section.

Effect of feeder location of GF on mechanical properties

Figure 2(a–d) shows the optical microscopy images of the GFs after extrusion of the PBT/GF (60/40 wt %) composites, which were prepared by the processing conditions of samples A to D shown in Table III. From Figure 2, we can see that the fiber lengths of samples A to D for PBT/GF (60/40 wt %) composites were found to be 640, 600, 314, and 745 μm, respectively. Standard deviations of the samples A to D were found to be 143, 94, 104, and 280 μm, respectively. Detailed explanations of the relationships between the feeder location of the fiber and the fiber length after extrusion of the composites represented in Figure 3 will be discussed.

Figure 3 shows the average fiber length after extrusion of the PBT/GF composites (samples A–D). From Figure 3, it shows that the fiber length
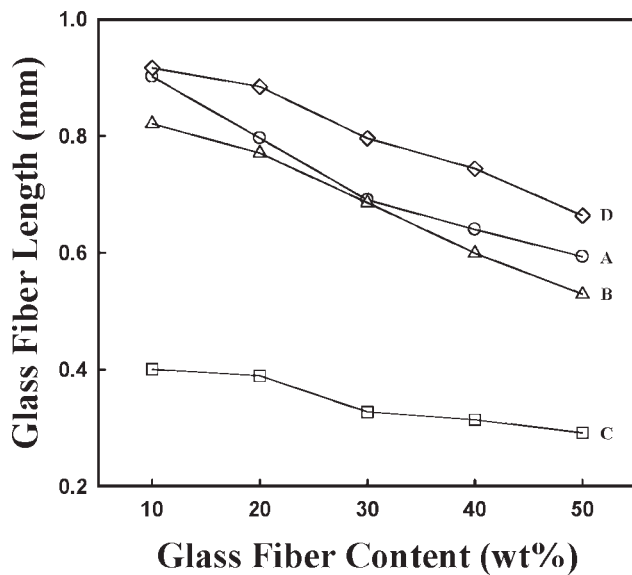
Figure 3 Effect of feeder location of glass fiber on the average fiber length of poly(butylene terephthalate)/glass fiber composite with fiber content for samples A, B, C, and D.

decreases with the increase of the fiber content. This behavior is possibly due to the fact that the collision among the fibers increases with the increase of the fiber content. When the fiber length is compared among the samples A, B, and C shown in Figure 3, the fiber length of sample C was shortest. The fiber length of sample C after extrusion was found to decrease from 400 μm to 294 μm with an increase of fiber content from 10 to 50 wt %. This is because the feeder location of the GF for sample C was the main feeder (3), and the distance from the tip of the die to the main feeder was 176 cm, the longest distance among the samples.

When the fiber length of the samples A and D are compared, the fiber length of sample A, which was prepared with the higher molecular weight PBT (M_w = 33,000), appears to be shorter than that of sample D, which was prepared with the lower molecular weight PBT (M_w = 25,000). The samples of A and D were prepared such that the feeder location of the fiber and type of the GF were same except molecular weight of the PBT. From this result, it is suggested that the molecular weight of PBT matrix is closely related with the fiber length of the PBT/GF composite after extrusion. The relation between the fiber length after extrusion and the mechanical properties of the PBT/GF composites will be discussed in the next section.
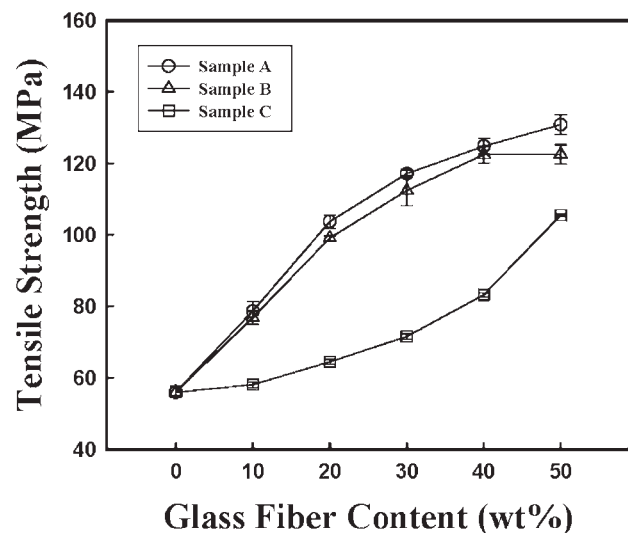
Figure 4(a,b) shows the flexural and tensile strengths of the PBT/GF composites, respectively, for samples A, B and C with GF content. The GF was added from different feeder locations for samples A, B, and C, which were prepared by the proc-

essing conditions shown in Table III. Regardless of the feeder locations of the GF, flexural and tensile strengths of the composites increased with increasing GF content.

Figure 4 also shows the effect of feeder location of GF on the flexural and tensile strengths of the PBT/GF composites. The distance between the fiber feeder and the end of the die was the shortest for sample A (87 cm), and the longest for sample C (176 cm). The flexural and tensile strengths of sample C showed the lowest values among the samples A to C. For samples A and B, the flexural and tensile strengths of the composites showed similar values, but slightly increased for sample A. These results are mainly due to the fact
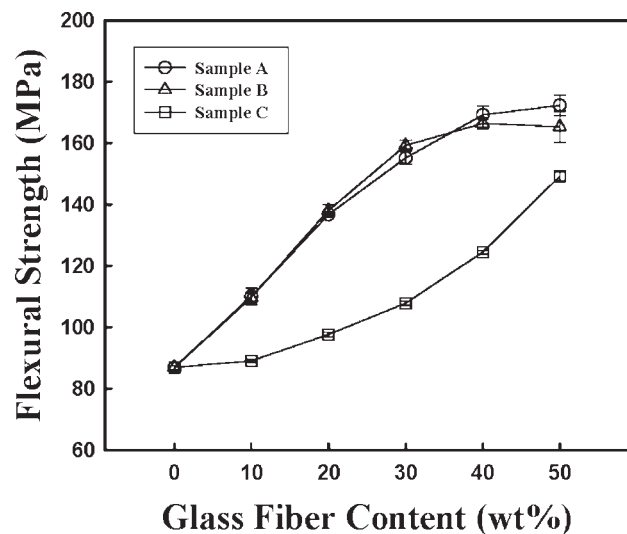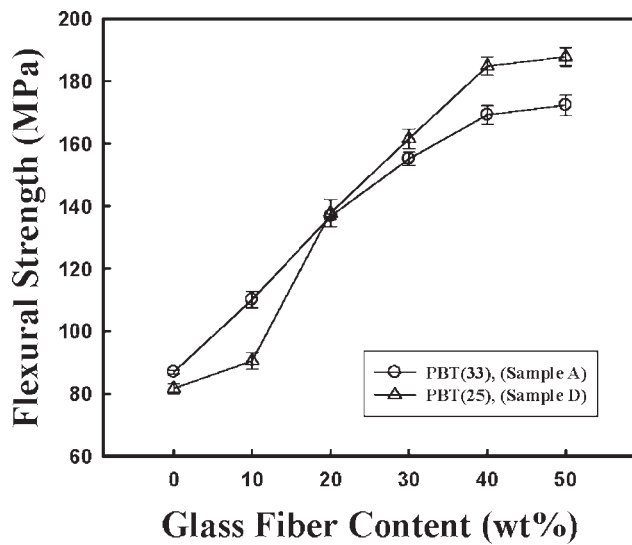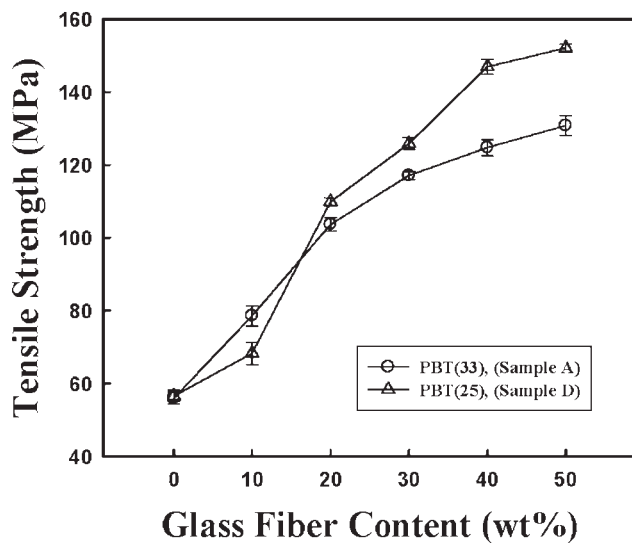

(a)

(b)

Figure 4 Effect of feeder location of glass fiber on the flexural and tensile strengths of poly(butylene terephthalate)/glass fiber composite with fiber content for samples A, B, and C.
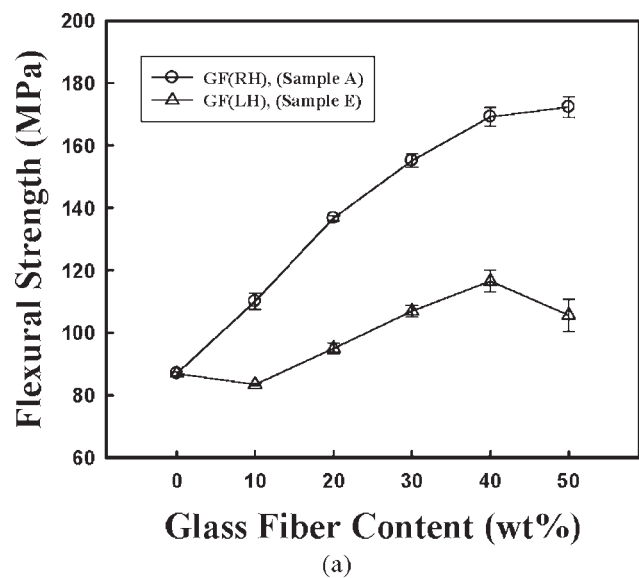
(a)

(b)

Figure 5 Effect of molecular weight of PBT on the flexural and tensile strengths of poly(butylene terephthalate)/glass fiber composite with fiber content for samples A and D.

that there is enough time for fibers to be broken severely while they are in the extruder when the distance between fiber feeder and the end of the die becomes longer. In Figure 3, the average fiber length of samples A, B, and C for the PBT/GF (60/40 wt %) composites has been found to be 640, 600, and 314 µm, respectively, which showed that the fiber length for sample C was the shortest among the samples. Therefore, the results of the fiber length after extrusion shown in Figure 3 are consistent with the results of the mechanical strengths of the PBT/GF composites shown in Figure 4. From these results, it is suggested that the fiber length after extrusion is closely

related to the mechanical strength of the fiber reinforced polymer composites.

Effect of molecular weight of PBT

Figure 5 shows the flexural and tensile strengths of PBT/GF(RH) composites with GF content and different molecular weights of PBT such that the weight average molecular weight of PBT (25) and PBT (33) are 25,000 and 33,000, respectively. From Figure 5, when the GF content is 10 wt %, the PBT/GF(RH) composite made with PBT (33) shows higher flexural and tensile strengths compared with the PBT/GF composite made with PBT (25). For GF content higher than 20 wt %, however, the composites

(a)
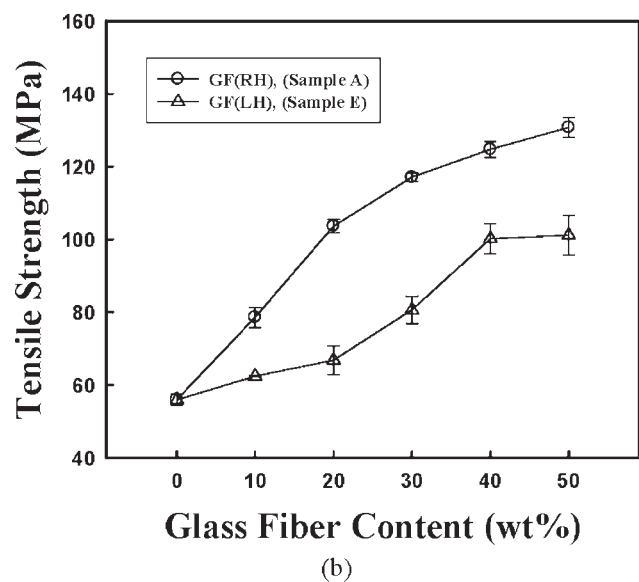
(b)

Figure 6 Effect of surface treatment of glass fiber on the flexural and tensile strengths of poly(butylene terephthalate)/glass fiber composite with fiber content for samples A and E.
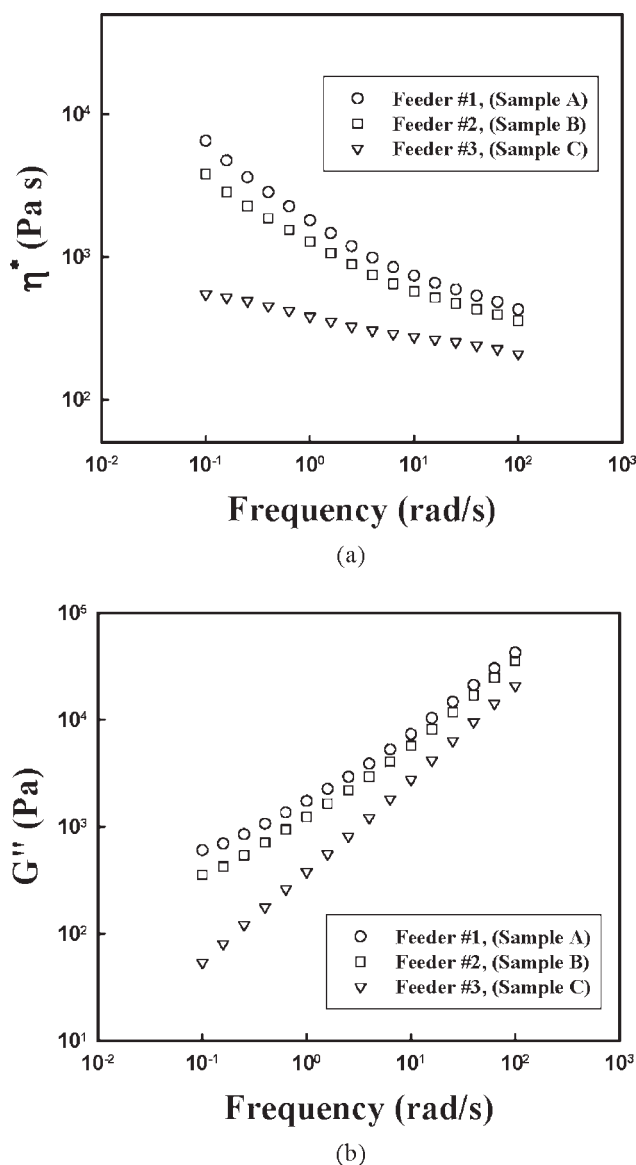
(a)

(b)

Figure 7 Effect of feeder location of glass fiber on the complex viscosity (h^*) and loss modulus (G') of poly(butylene terephthalate)/glass fiber composite with fiber content of 50 wt % for samples A, B, and C.

prepared with PBT (25) showed higher flexural and tensile strengths compared with those of the PBT/GF(RH) composites prepared with PBT (33). From this result, it is suggested that the molecular weight of the PBT matrix has an effect greater than that of the fiber length of the composites on the mechanical strength when the fiber content is low (10 wt %).

At fiber contents of greater than 20 wt %, the fiber length of the composite is a predominant factor on the mechanical strengths of the PBT/GF composites. From the results of Figure 3, we have seen that the length of GF of the composite made with lower molecular weight PBT (25) was longer than the composite made with higher molecular weight PBT (33). This behavior is probably due to more severe break-

age of GF in the more viscous matrix during mixing of the PBT and GF in the extruder.

Effect of surface treatment of GF

Figure 6 shows the flexural and tensile strengths of PBT(33)/GF composites with different types of GFs coated with different organic compounds. GF (RH) was coated with polar polyester, and GF (LH) was coated with nonpolar aliphatic hydrocarbon. The results in Figure 6 show that the flexural and tensile strengths of the PBT(33)/GF (RH) composites are higher than the PBT(33)/GF (LH) composites. From the results of Figure 6, it is suggested that the bonding force between the PBT matrix and GF (RH) is higher than that of the PBT matrix and GF (LH). When the GF is coated with organic materials having a polarity similar to the PBT matrix, mechanical properties can be improved due to a higher bonding force between the matrix and fiber.

Effect of fiber length on rheological properties

Figure 7 shows the effects of fiber length on the complex viscosity (η^*) and loss modulus (G'') of the PBT(33)/GF(RH) (50/50 wt %) composites. The results of Figure 7 show that the complex viscosity of the PBT(33)/GF(RH) composites (samples A–C) decreases with increasing frequency representing shear thinning behavior. The shear thinning behavior of sample C was less significant than that of samples A and B because the fiber length after extrusion of sample C was found to be the shortest, as we have seen in Figure 3. The fiber reinforced polymer composite containing long fiber shows non-Newtonian behavior more significantly because of the high elasticity of the composite compared to long fibers.

Figure 7 also shows that the complex viscosity and loss modulus of the PBT(33)/GF(RH) composites (samples A, B, and C) increases with the increase of fiber length of the PBT(33)/GF(RH) composites after extrusion. As we have seen in Figure 3, the fiber length of the PBT(33)/GF(RH) composites (samples A, B, and C) after extrusion was found to be 600, 532, and 294 µm, respectively. From the previous results of Figures 4 and 7, it is suggested that the fiber length of the composite after extrusion is closely related to the mechanical strength and rheological properties of the PBT/GF composites.

CONCLUSIONS

Morphological, mechanical, and rheological properties of the PBT/GF composites were investigated by melt extrusion. From the results of molecular weight

effects of PBT on the mechanical properties of the PBT/GF composites, the composites prepared with lower molecular weight PBT showed higher flexural and tensile strengths than the PBT/GF composites prepared with higher molecular weight PBT when the fiber content was over 20 wt %. The fiber length after extrusion of the composite prepared with lower molecular weight PBT was longer than that of the composite prepared with the higher molecular weight PBT. This behavior was probably due to more severe breakage of the GF in more-viscous matrix during mixing of the PBT and GF in the extruder.

From the effect of fiber length on the complex viscosity of the PBT/GF composite, shear thinning behavior was more significant for the composites containing long fibers than for composites containing short fibers, because of its high elasticity. From the previous results, it is suggested that optimum mechanical properties of the fiber reinforced polymer composite can be achieved by varying processing conditions such as feeder location of the fiber, molecular weight of the matrix, and surface treatment of the fiber.

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