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An experimental study of the water-assisted injection molding of glass fiber filled poly-butylene-terephthalate (PBT) composites

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Abstract

The purpose of this report was to experimentally study the water-assisted injection molding process of poly-butylene-terephthalate (PBT) composites. Experiments were carried out on an 80-ton injection-molding machine equipped with a lab scale water injection system, which included a water pump, a pressure accumulator, a water injection pin, a water tank equipped with a temperature regulator, and a control circuit. The materials included virgin PBT and a 15% glass fiber filled PBT composite, and a plate cavity with a rib across center was used. Various processing variables were examined in terms of their influence on the length of water penetration in molded parts, and mechanical property tests were performed on these parts. X-ray diffraction (XRD) was also used to identify the material and structural parameters. Finally, a comparison was made between water-assisted and gas-assisted injection molded parts. It was found that the melt fill pressure, melt temperature, and short shot size were the dominant parameters affecting water penetration behavior. Material at the mold-side exhibited a higher degree of crystallinity than that at the water-side. Parts molded by gas also showed a higher degree of crystallinity than those molded by water. Furthermore, the glass fibers near the surface of molded parts were found to be oriented mostly in the flow direction, but oriented substantially more perpendicular to the flow direction with increasing distance from the skin surface.

Keywords: Water assisted injection molding; Glass fiber reinforced poly-butylene-terephthalate (PBT) composites; Processing parameters; B. Mechanical properties; Crystallinity; A. Polymer matrix composites; Processing

1. Introduction

Water-assisted injection molding technology [1] has proved itself a breakthrough in the manufacture of plastic parts due to its light weight, faster cycle time, and relatively lower resin cost per part. In the water-assisted injection molding process, the mold cavity is partially filled with the polymer melt followed by the injection of water into the core of the polymer melt. A schematic diagram of the water-assisted injection molding process is illustrated in Fig. 1. Water-assisted injection molding can produce parts incorporating both thick and thin sections with less shrinkage and warpage and with a better surface finish, but with a shorter cycle time. The water-assisted injection molding process can also enable greater freedom of design, material savings, weight reduction, and cost savings in terms of tooling and press capacity requirements [2–4]. Typical applications include rods and tubes, and large sheet-like structural parts with a built-in water channel network. On the other hand, despite the advantages associated with the process, the molding window and process control are more critical and difficult since additional processing parameters are involved. Water may also corrode the steel mold, and some materials including thermoplastic composites are difficult to mold successfully. The removal of water after molding is also a challenge for this novel technology. Table 1 lists the advantages and limitations of water-assisted injection molding technology.

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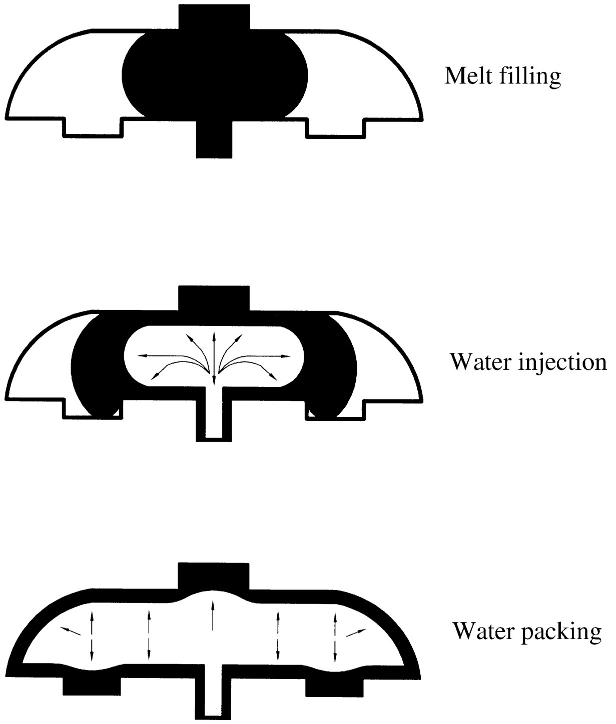

Fig. 1. Schematic diagram of water-assisted injection molding process.

Table 2
A comparison of water and gas-assisted injection molding

	Water	Gas
1. Cycle time	Short	Long
2. Medium cost	Low	High
3. Internal foaming	No	Yes
4. Residual wall thickness	Small	Large
5. Outside surface roughness	Low	High
6. Outside surface gloss	High	Low
7. Fingering	Greater	Less
8. Asymmetrical penetration	More stable	Unstable
9. Material crystallinity	Low	High
10. Part transparency	High	Low
11. Internal surface (semi-crystalline materials)	Smooth	Less smooth
12. Internal surface (amorphous materials)	Rough	Smooth

Water assisted injection molding has advantages over its better known competitor process, gas assisted injection molding [5], because it incorporates a shorter cycle time to successfully mold a part due to the higher cooling capacity of water during the molding process. The incompressibility, low cost, and ease of recycling the water makes it an ideal medium for the process. Since water does not dissolve and diffuse into the polymer melts during the molding process, the internal foaming phenomenon [6] that usually occurs in gas-assisted injection molded parts can be eliminated. In addition, water assisted injection molding provides a better capability of molding larger parts with a small residual wall thickness. Table 2 lists a comparison of water and gas assisted injection molding.

With increasing demands for materials with improved performance, which may be characterized by the criteria of lower weight, higher strength, and a faster and cheaper production cycle time, the engineering of plastics is a process that cannot be ignored. These plastics include thermoplastic and thermoset polymers. In general, thermoplastic polymers have an advantage over thermoset polymers in terms of higher impact strength, fracture resistance and strains-to-failure. This makes thermoplastic polymers very popular materials in structural applications.

Poly-butylene-terephthalate (PBT) is one of the most frequently used engineering thermoplastic materials, which is formed by polymerizing 1.4 butylene glycol and DMT together. Fiber-reinforced composite materials have been adapted to improve the mechanical properties of neat plastic materials. Today, short glass fiber reinforced PBT is widely used in electronic, communication and automobile applications. Therefore, the investigation of the processing of fiber-reinforced PBT is becoming increasingly important [7–10].

This report was made to experimentally study the water-assisted injection molding process of poly-butylene-terephthalate (PBT) materials. Experiments were carried out on an 80-ton injection-molding machine equipped with a lab scale water injection system, which included a water pump, a pressure accumulator, a water injection pin, a water tank equipped with a temperature regulator, and a control circuit. The materials included a virgin PBT and a 15% glass fiber filled PBT composite, and a plate cavity with a rib across center was used. Various processing variables were examined in terms of their influence on the length of water penetration in molded parts, which included melt temperature, mold temperature, melt filling speed, short-shot size, water pressure, water temperature, water hold and water injection delay time. Mechanical property tests were also performed on these molded parts, and XRD was used to identify the material and structural

Table 1
Advantages and disadvantages of water-assisted injection molding

Advantages	Disadvantages
1. Short cycle time	1. Corrosion of the steel mold due to water
2. Low assisting medium cost (water is much cheaper and can be easily recycled)	2. Larger orifices for the injection pin required (easier to get stuck by the polymer melt)
3. No internal foaming phenomenon in molded parts	3. Some materials are more difficult to mold (especially amorphous thermoplastics)
	4. Removal of water after molding is required

parameters. Finally, a comparison was made between water-assisted and gas-assisted injection molded parts.

2. Experimental procedure

2.1. Materials

The materials used included a virgin PBT (Grade 1111FB, Nan-Ya Plastic, Taiwan) and a 15% glass fiber filled PBT composite (Grade 1210G3, Nan-Ya Plastic, Taiwan). Table 3 lists the characteristics of the composite materials.

2.2. Water injection unit

A lab scale water injection unit, which included a water pump, a pressure accumulator, a water injection pin, a water tank equipped with a temperature regulator, and a control circuit, was used for all experiments [3]. An orifice-type water injection pin with two orifices (0.3 mm in diameter) on the sides was used to mold the parts. During the experiments, the control circuit of the water injection unit received a signal from the molding machine and controlled the time and pressure of the injected water. Before injection into the mold cavity, the water was stored in a tank with a temperature regulator for 30 min to sustain an isothermal water temperature.

2.3. Molding machine and molds

Water-assisted injection molding experiments were conducted on an 80-ton conventional injection-molding machine with a highest injection rate of 109 cm^3/s. A plate cavity with a trapezoidal water channel across the center was used in this study. Fig. 2 shows the dimensions of the cavity. The temperature of the mold was regulated by a water-circulating mold temperature control unit. Various processing variables were examined in terms of their influence on the length of water penetration in water channels of molded parts: melt temperature, mold temperature, melt fill pressure, water temperature and pressure, water injection delay time and hold time, and short shot size of the polymer melt. Table 4 lists these processing variables as well as the values used in the experiments.

Table 3
Characteristics of the glass–fiber reinforced PBT composite

Property	ASTM	PBT	15% G.F. PBT
Yield strength (kg/cm^2)	D-638	600	1000
Bending stress (kg/cm^2)	D-570	900	1500
Hardness (R-scale)	D-785	119	120
Heat distortion temperature (°C) (18.6 kg/cm^2)	D-648	60	200
Melt flow index (MFI)	D-1238	40	25
Impact strength (Kg-cm/cm)	D-256	5	5
Melting temperature (°C)	DSC	224	224

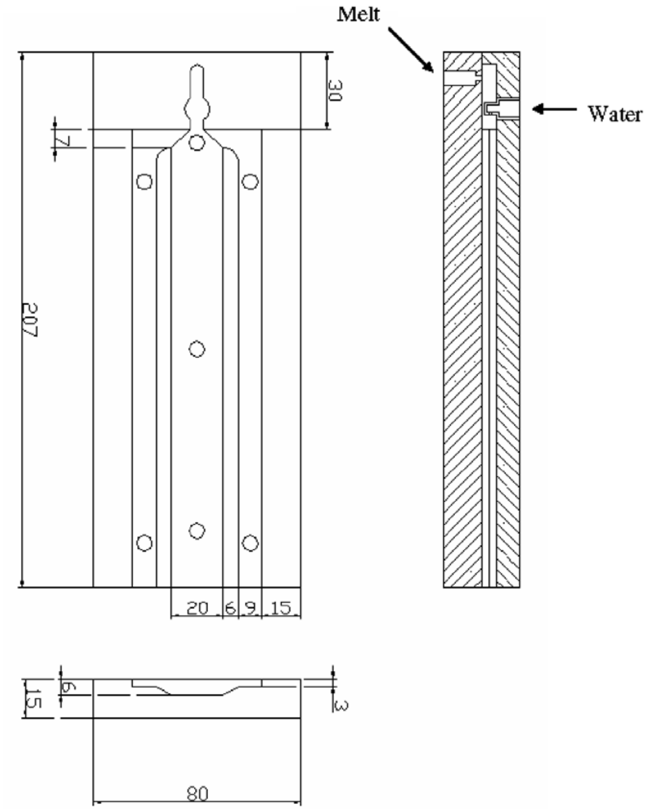

Fig. 2. Layout and dimensions of mold cavity (unit: mm).

2.4. Gas injection unit

In order to make a comparison of water and gas-assisted injection molded parts, a commercially available gas injection unit (Gas Injection PPC-1000) was used for the gas-assisted injection molding experiments. Details of the gas injection unit setup can be found in the Refs. [11–15]. The processing conditions used for gas-assisted injection molding were the same as that of water-assisted injection molding (terms in bold in Table 4), with the exception of gas temperature which was set at 25 °C.

2.5. XRD

In order to analyze the crystal structure within the water-assisted injection-molded parts, wide-angle X-ray diffraction (XRD) with 2D detector analyses in transmission mode were performed with Cu Kα radiation at 40 kV and 40 mA. More specifically, the measurements were performed on the mold-side and water-side layers of the water-assisted injection-molded parts, with the 2θ angle ranging from 7° to 40°. The samples required for these analyses were taken from the center portion of these molded parts. To obtain the desired thickness for the XRD samples, the excess was removed by polishing the

Table 4
The processing variables as well as the values used in the experiments

A	B	C	D	E	F
Melt pressure (Mpa)	Melt temperature (°C)	Short shot size (%)	Water pressure (Mpa)	Water temperature (°C)	Mold temperature (°C)
140	280 (270)	76	8	80	80
126	275 (265)	77	9	75	75
114	**270 (260)**	**78**	**10**	**70**	**70**
98	265 (255)	80	11	65	65
84	260 (250)	81	12	60	60

*The values in the parentheses are the melt temperatures used for virgin PBT materials.

samples on a rotating wheel on a rotating wheel, first with wet silicon carbide papers, then with 300-grade silicon carbide paper, followed by 600- and 1200-grade paper for a better surface smoothness.

2.6. Mechanical properties

Tensile strength and bending strength were measured on a tensile tester. Tensile tests were performed on specimens obtained from the water-assisted injection molded parts (see Fig. 3) to evaluate the effect of water temperature on the tensile properties. The dimensions of specimens for the experiments were 30 mm × 10 mm × 1 mm. Tensile tests were performed in a LLOYD tensiometer according to the ASTM D638M test. A 2.5 kN load cell was used and the crosshead speed was 50 mm/min.

Bending tests were also performed at room temperature on water-assisted injection molded parts. The bending specimens were obtained with a die cutter from parts (Fig. 3) subjected to various water temperatures. The dimensions of the specimens were 20 mm × 10 mm × 1 mm. Bending tests were performed in a micro tensile tester according to the ASTM D256 test. A 200 N load cell was used and the crosshead speed was 50 mm/min.

2.7. Microscopic observation

The fiber orientation in molded specimens was observed under a scanning electron microscope (Jeol Model 5410). Specimens for observation were cut from parts molded by water-assisted injection molding across the thickness (Fig. 3). They were observed on the cross-section perpendicular to the flow direction. All specimen surfaces were gold sputtered before observation.

3. Results and discussion

All experiments were conducted on an 80-ton conventional injection-molding machine, with a highest injection rate of 109 cm^3/s. A plate cavity with a trapezoidal water channel across the center was used for all experiments.

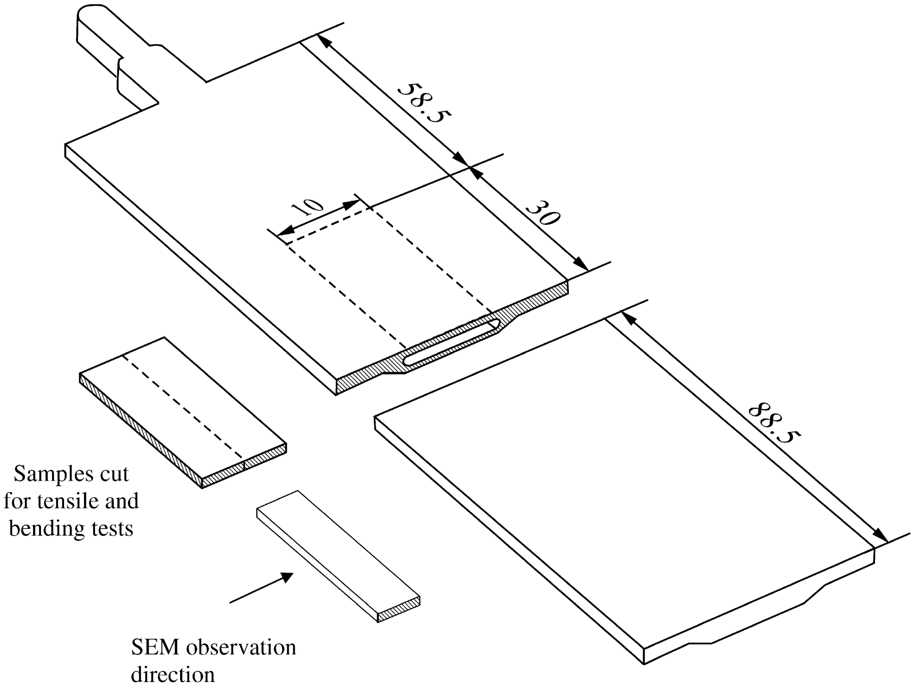

Fig. 3. Schematically, the positioning of the samples cut from the molded parts for tensile and bending tests and microscopic observations.

3.1. Fingerings in molded parts

All molded parts exhibited the water fingering phenomenon at the channel to plate transition areas. In addition, molded glass fiber filled composites showed more severe water fingerings than those of non-filled materials, as shown photographically in Fig. 4. Fingerings usually form when a less dense, less viscous fluid penetrates a denser, more viscous fluid immiscible with it. Consider a sharp two phase interface or zone where density and viscosity change rapidly. The pressure force (P_2-P_1) on the displaced fluid as a result of a virtual displacement δx of the interface can be described by [16],

$$\delta P = (P_2 - P_1) = [(\mu_1 - \mu_2)U/K]\delta x \tag{1}$$

where U is the characteristic velocity and K is the permeability. If the net pressure force is positive, then any small displacement will be amplified and lead to an instability and part fingerings. For the displacement of a dense, viscous fluid (the polymer melt) by a lighter, less viscous one (water), we can have $\Delta\mu = \mu_1 - \mu_2 > 0$, and $U > 0$ [16]. In this case, instability and the relevant fingering result when a more viscous fluid is displaced by a less viscous one, since the less viscous fluid has the greater mobility. The results in this study suggest that glass fiber filled composites exhibit a higher tendency for part fingerings. This might be due to the fact that the viscosity difference $\Delta\mu$ between water and the filled composites is larger than the difference between water and the non-filled materials. Water-assisted injection molded composites thus exhibit more severe part fingerings.

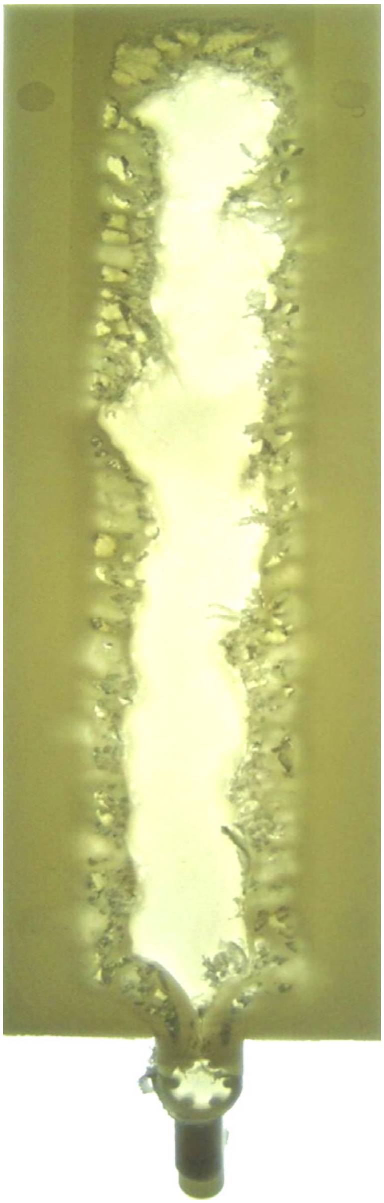

Fig. 4. Photograph of water-assisted injection molded PBT composite part.

3.2. Effects of processing parameters on water penetration

Various processing variables were studied in terms of their influence on the water penetration behavior. Table 4 lists these processing variables as well as the values used in the experiments. To mold the parts, one central processing condition was chosen as a reference (bold term in Table 4). By changing one of the parameters in each test, we were able to better understand the effect of each parameter on the water penetration behavior of water assisted injection molded composites. After molding, the length of water penetration was measured. Figs. 5–10 show the effects of these processing parameters on the length of water penetration in molded parts, including melt fill pressure, melt temperature, mold temperature, short shot size, water temperature, and water pressure.

The experimental results in this study suggest that water penetrates further in virgin PBT than in glass fiber filled PBT composites. This is due to the fact that with the reinforcing glass fibers the composite materials have less volumetric shrinkage during the cooling process. Therefore, they mold parts with a shorter water penetration length.

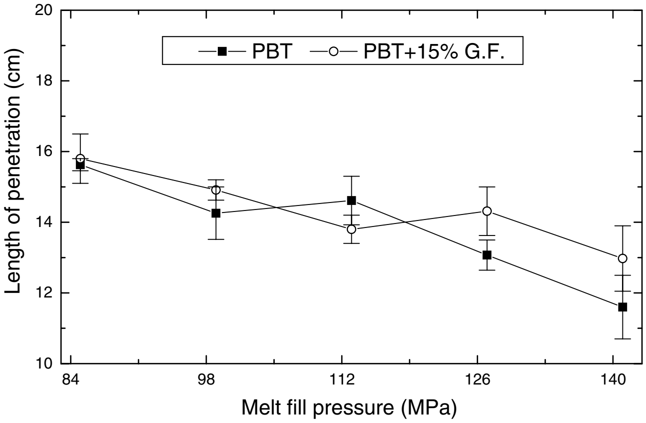

Fig. 5. Effects of melt fill pressure on the length of water penetration in molded parts.

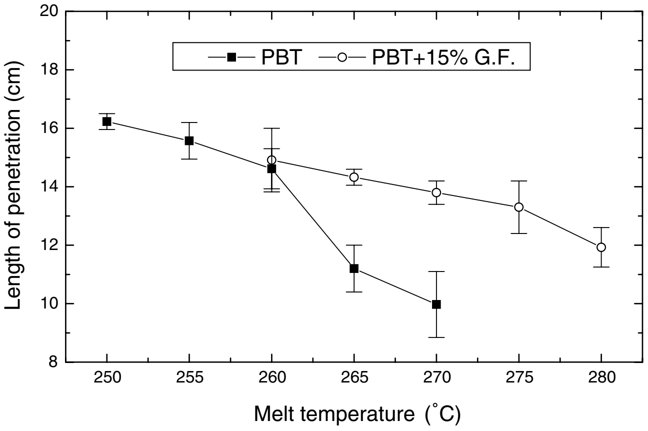

Fig. 6. Effects of melt temperature on the length of water penetration in molded parts.

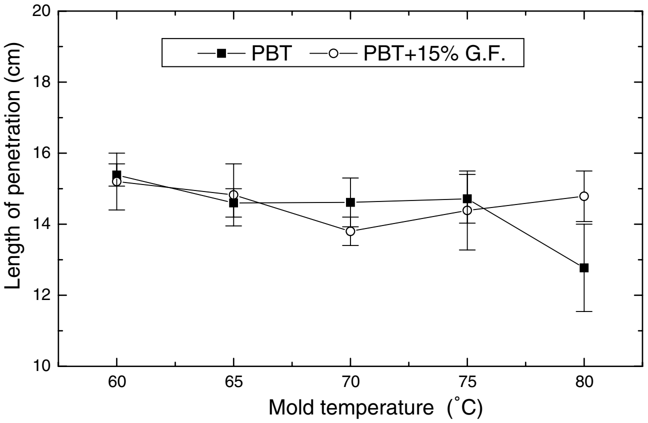

Fig. 7. Effects of mold temperature on the length of water penetration in molded parts.

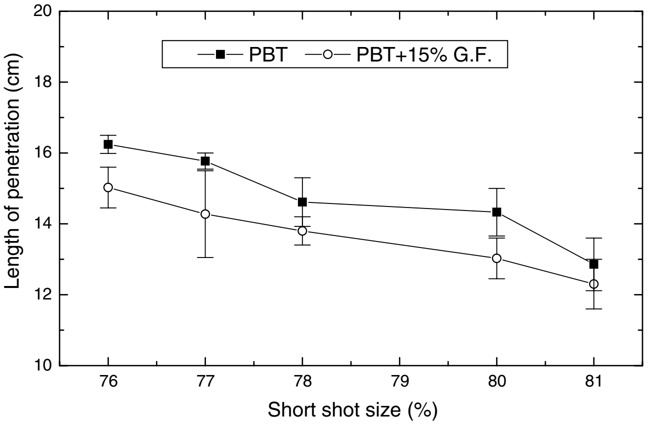

Fig. 8. Effects of short shot size on the length of water penetration in molded parts.

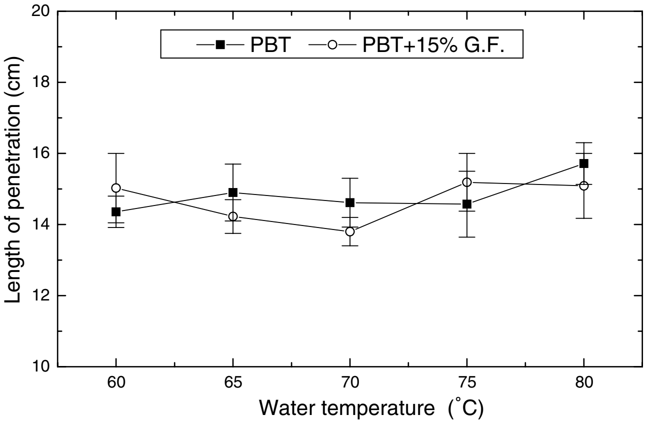

Fig. 9. Effects of water temperature on the length of water penetration in molded parts.

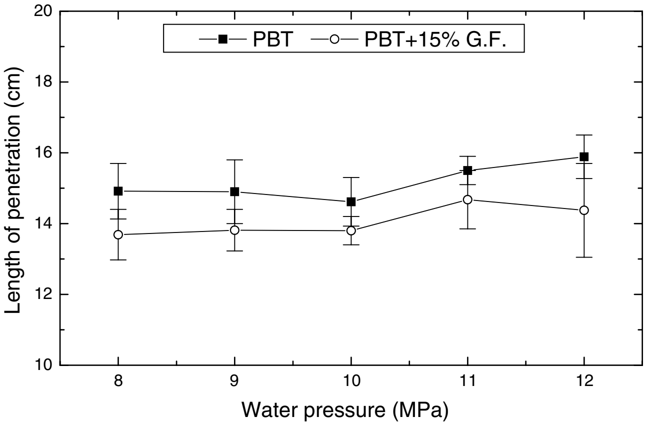

Fig. 10. Effects of water pressure on the length of water penetration in molded parts.

The length of water penetration decreases with the melt fill pressure (Fig. 5). This can be explained by the fact that increasing the melt fill pressure increases the flow resistance inside the mold cavity. It is then more difficult for the water to penetrate into the core of the materials. The length of water penetration decreases accordingly [3].

The melt temperature was also found to reduce the water penetration in molded PBT composite parts (Fig. 6). This might be due to the fact that increasing the melt temperature decreases viscosity of the polymer melt. A lower viscosity of the materials helps the water to pack the water channel and increase its void area, instead of penetrating further into the parts [4]. The hollow core ratio at the beginning of the water channel increases and the length of water penetration may thus decrease.

Increasing the mold temperature decreases somewhat the length of water penetration in molded parts (Fig. 7). This is due to the fact that increasing the mold temperature decreases the cooling rate as well as the viscosity of the materials. The water then packs the channel and increases its void area near the beginning of the water channel, instead of penetrating further into the parts [3]. Molded parts thus have a shorter water penetration length.

Increasing the short shot size decreases the length of water penetration (Fig. 8). In water-assisted injection molding, the mold cavity is partially filled with the polymer melt followed by the injection of water into the core of the polymer melt [4]. Increasing the short shot size of the polymer melt will therefore decrease the length of water penetration in molded parts.

For the processing parameters used in the experiments, increasing the water temperature (Fig. 9) or the water pressure (Fig. 10) increases the length of water penetration in molded parts. Increasing the water temperature decreases the cooling rate of the materials and keeps the polymer melt hot for a longer time; the viscosity of the materials decreases accordingly. This will help the water penetrate further into the core of the parts [3]. Increasing the water pressure also helps the water penetrate into the materials. The length of water penetration thus increases.

Finally, the deflection of molded parts, subjected to various processing parameters, was also measured by a profilemeter. The maximum measured deflection is considered as the part warpage. The result in Fig. 11 suggests that the part warpage decreases with the length of water penetration. This is due to the fact that the longer the water penetration, the more the water pressure can pack the polymeric materials against the mold wall. The shrinkage as well as the relevant part warpage decreases accordingly.

3.3. Crystallinity of molded parts

PBT is a semi-crystalline thermoplastic polyester with a high crystallization rate. In the water-assisted injection molding process, crystallization occurs under non-isothermal conditions in which the cooling rate varies with cooling time. Here the effects of various processing parameters (including melt temperature, mold temperature, and water temperature) on the level of crystallinity in molded parts were studied. Measurements were conducted on a wide-angle X-ray diffraction (XRD) with 2D detector analyses (as described in Section 2). The measured results in Fig. 12 showed that all materials at the mold-side layer exhibited a higher degree of crystallinity than those at the water-side layer. The result indicates that the water has a better cooling capacity than the mold during the cooling

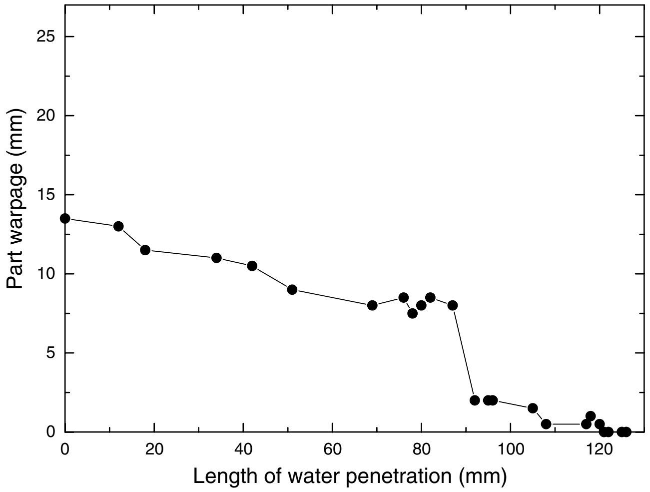

Fig. 11. Measured warpage of molded parts decreases with the length of water penetration.

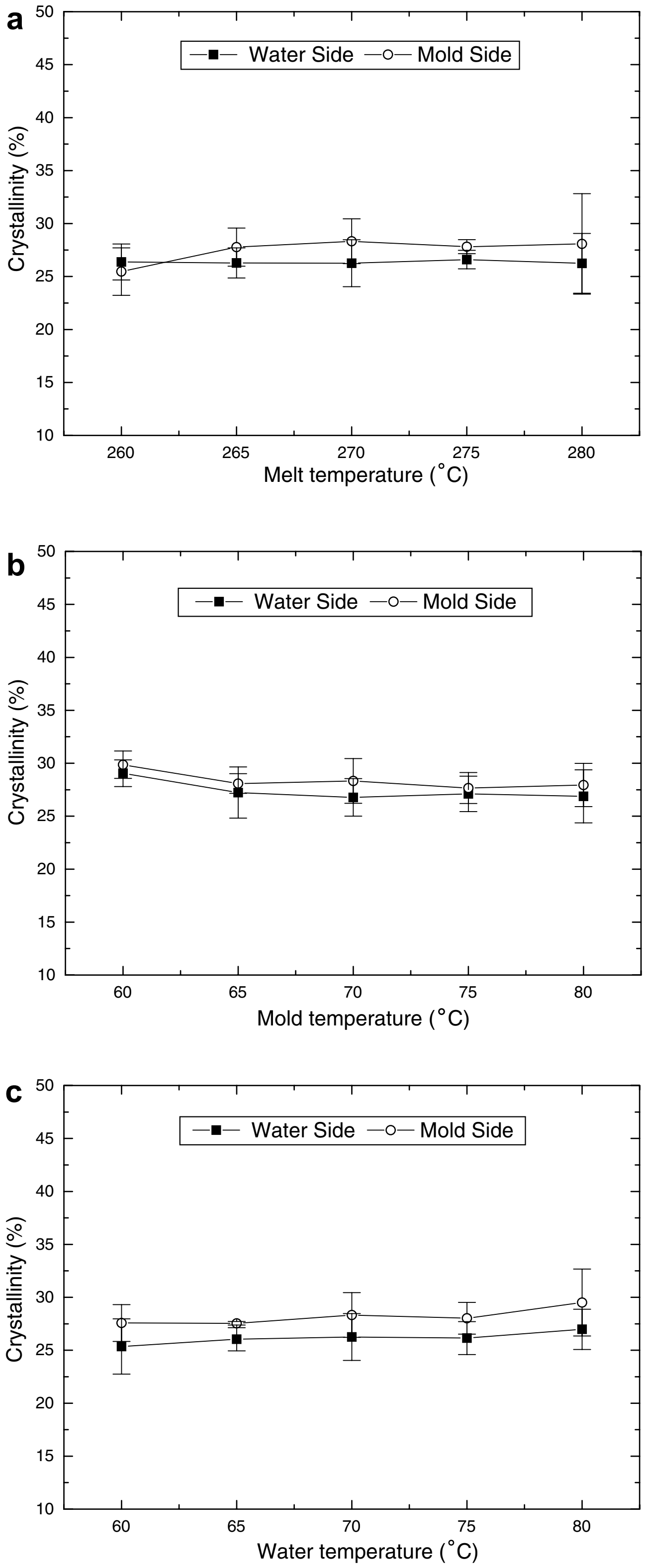

Fig. 12. Effect of melt temperature, mold temperature, and water temperature on the level of crystallinity of water-assisted injection molded parts.

process. This matches our earlier finding [17] by measuring the in-mold temperature distribution. In addition, the experimental result in Fig. 12c also suggests that the crystallinity of the molded materials generally increases with the water temperature. This is due to the fact that increasing the water temperature decreases the cooling rate of the materials during the cooling process. Molded parts thus exhibited a higher level of crystallinity.

On the other hand, to make a comparison of the crysallinity of parts molded by gas and water, gas-assisted injection molding experiments were carried out on the same injection molding machine as that used with water, but equipped with a high-pressure nitrogen gas injection unit [11–15]. The measured results in Fig. 13 suggests that gas-assisted injection molded parts have a higher degree of crystallinity than water-assisted injection mold parts. This is due to the fact that water has a higher cooling capacity and cools down the parts faster than gas. Parts molded by water thus exhibited a lower level of crystallinity than those molded by gas.

3.4. Mechanical properties

Tensile tests were performed on specimens obtained from the water-assisted injection molded parts to examine the effect of water temperature on the tensile properties. Fig. 14 showed the measured decrease subjected to various water temperatures. As can be observed, both yield strength and the elongational strain at break of water assisted molded PBT materials decrease with the water temperature. On the other hand, bending tests were also performed at room temperature on water-assisted injection molded parts. The measured result in Fig. 15 suggests that the bending strength of molded parts decreases with the water temperature.

Increasing the water temperature generally decreases the cooling rate and molds parts with higher level of crystallin-

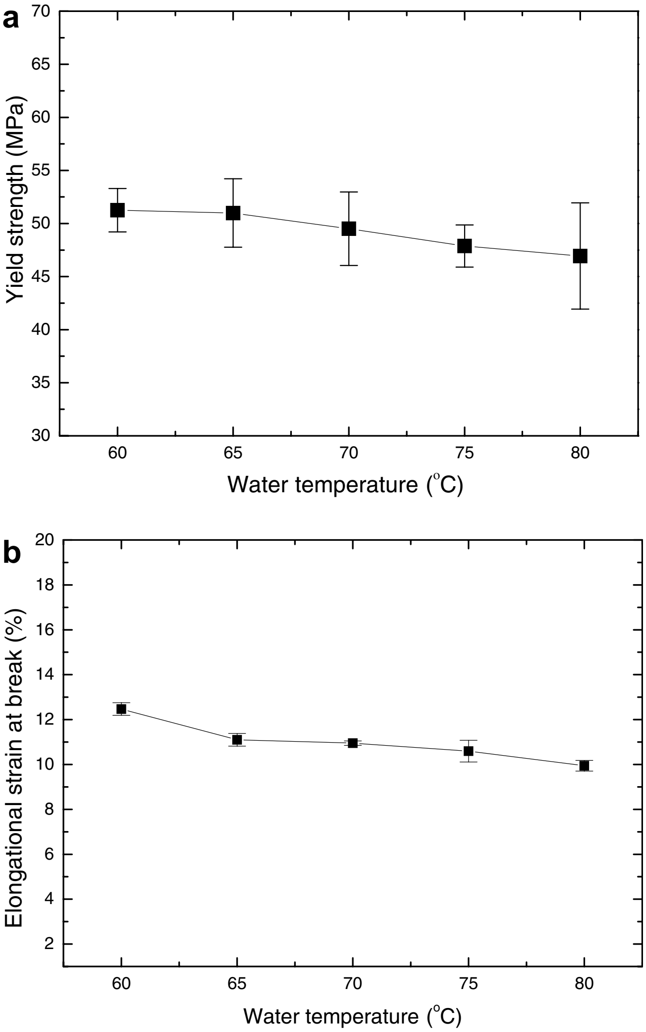

Fig. 14. Effect of water temperature on the tensile properties of molded PBT parts.

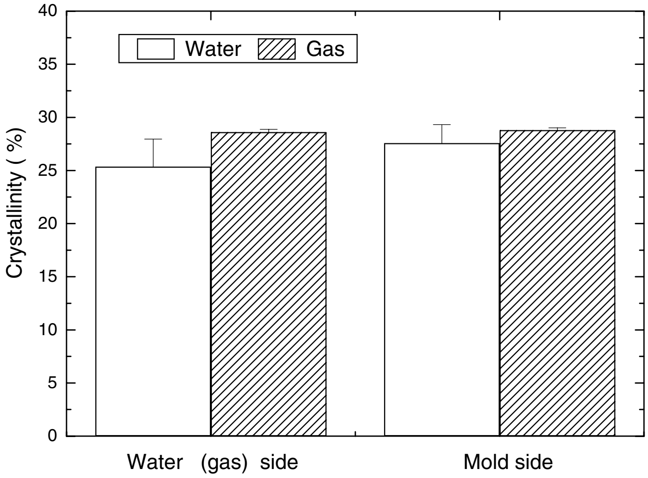

Fig. 13. Level of crystallinity of parts molded by gas and water injection molding.

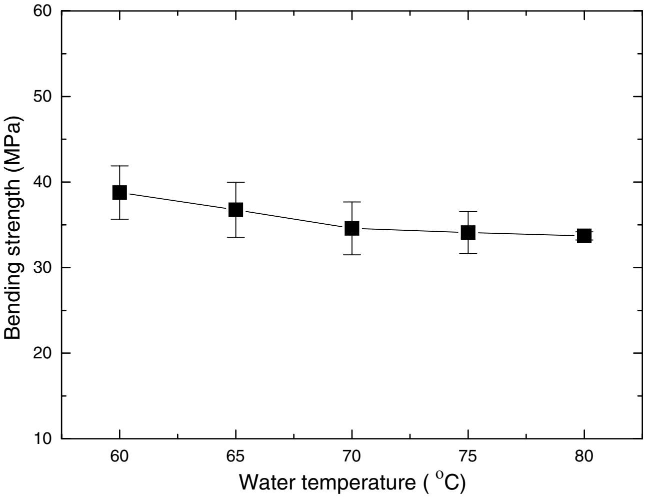

Fig. 15. Effect of water temperature on the bending strength of molded PBT parts.

ity. As is usually encountered in semi-crystalline thermoplastics, a higher degree of crystallization means a lower content of free volume and therefore an increasing level of stiffness. However, the experimental results here suggest that the quantitative contribution of crystallinity to PBT's mechanical properties is negligible, while there is a more important quantitative increase of tensile and bending strength for the PBT materials. The mechanical properties of molded materials are dependent on both the amount and the type of crystalline regions developed during processing. The fact that the ductility of PBT decreases with the degree of crystallinity may indicate that a more crystalline and stiffer PBT developed at a lower cooling rate during processing and did not exhibit higher stress values in tensile tests because of a lack of ductility, and therefore did not behave as strong as expected from their stiffness [18]. Nevertheless, more detailed experiments will be needed for the future works to investigate the morphological parameters of water-assisted injection molded parts and their correlation with the parts' mechanical properties.

3.5. Fiber orientation in molded parts

Small specimens were cut out from the middle of molded parts in order to observe their fiber orientation. The position of the specimen for the fiber orientation observation is as shown in Fig. 3. All specimen surfaces were polished and gold sputtered before observation. Fig. 16 shows the microstructure of the water-assisted injection molded composite parts. The measured result suggests that the fiber orientation distribution in water-assisted injection molded parts is quite different from that of conventional injection molded parts.

In conventional injection molded parts, two regions are usually observed: the thin skin and the core. In the skin region near the wall, all fibers are oriented parallel to the flow direction, while at the core, the fibers are oriented randomly in the flow plane [19,20]. Compared to conventional injection molding, water-assisted injection molding technology is different in the way the mold is filled. With a conventional injection molding machine, one cycle is characterized by the phases of filling, packing and cooling. In the water-assisted injection molding process, the mold cavity is partially filled with the polymer melt followed by the injection of water into the core of the polymer melt. The novel filling process influences the orientation of fibers and matrix in a part significantly.

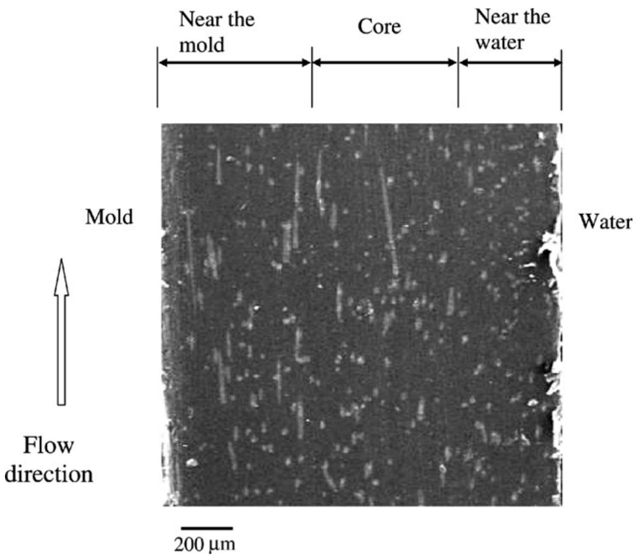

Fig. 16. Fiber orientation across the thickness of water-assisted injection molded PBT composites.

From Fig. 16, the fiber orientation in water-assisted injection molded parts can be approximately divided into three zones. In the zone near the mold-side surface where the shear is more severe during the mold filling, fibers are principally parallel. For the zone near the water-side surface, the shear is smaller and the velocity vector greater. In this case, the fiber tends to be positioned more transversely in the direction of injection. At the core, the fibers tend to be oriented more randomly. Generally speaking, the glass fibers near the mold-side surface of molded parts were found to be oriented mostly in the flow direction, and oriented substantially perpendicular to the flow direction with increasing distance from the mold-side surface.

Finally, it should be noted that a quantitative comparison of morphology and fiber orientation [21] in water-assisted molded and conventional injection molded parts will be made by our lab in future works.

4. Conclusions

This report was made to experimentally study the water-assisted injection molding process of poly-butylene-terephthalate (PBT) composites. The following conclusions can be drawn based on the current study.

1. Water-assisted injection molded PBT parts exhibit the fingering phenomenon at the channel to plate transition areas. In addition, glass fiber filled composites exhibit more severe water fingerings than those of non-filled materials.
2. The experimental results in this study suggest that the length of water penetration in PBT composite materials increases with water pressure and temperature, and decreases with melt fill pressure, melt temperature, and short shot size.
3. Part warpage of molded materials decreases with the length of water penetration.
4. The level of crystallinity of molded parts increases with the water temperature. Parts molded by water show a lower level of crystallinity than those molded by gas.
5. The glass fibers near the surface of molded PBT composite parts were found to be oriented mostly in the flow direction, and oriented substantially perpendicular to the flow direction with increasing distance from the skin surface.

Acknowledgement

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