INVESTIGATION ON PROPERTIES OF RECYCLED GLASS FIBER REINFORCED FLAME RETARDANT PBT

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Abstract: Due to its excellent comprehensive properties, glass fiber reinforced flame retardant PBT was widely applied in electrical accessory. In this paper, the glass fiber reinforced flame retardant PBT with five times mold-injection effect on its carboxyl content, characteristic viscosity of PBT and microscopic appearance were studied**.** The properties of recycled glass fiber reinforced flame retardant PBT including mechanical, electrical and frame retardant properties were well investigated. The results demonstrated that when the recycled glass fiber reinforced flame retardant PBT were multiple processed, its carboxyl content increased while the characteristic viscosity of PBT and the length of glass fiber decreased. After five times mold-injection, all its physical properties decreased, especially the notched impact strength, tensile strength, flexural strength drastically decreased from 75 J/m to 33 J/m, 115.6 MPa to 54.9 MPa, 161.1 MPa to 95.2 MPa, respectively.

Keywords: glass fiber reinforced flame retardant PBT; mold-injection; physical properties; recycle

1. Introduction

Polyesters, especially poly(1,4-butylene terephthalate) (PBT), is one of the most important engineering polymers with a wide range of applications because it possesses good dimensional stability, low moisture absorption, high heat distortion temperature, good chemical resistance, excellent electrical properties, creep resistance and high strength and rigidity. Due to its numerous virtues, PBT is one of the most frequently used engineering thermoplastic materials. Halogen-containing additives were found to be very efficient fire retardants in PBT, with the addition of brominated compounds, a great number of systems are available to make PBT flame retardant. Glass-fiber reinforced poly(butylene terephthalate) (PBT/GF), flame retarded with brominated styrene, brominated phenylethane, brominated epoxy resin and brominated polycarbonate, are the rare halogen materials to achieve a V-0 classification in UL 94 test. PBT fiber-reinforced composite materials have been adapted to improve the mechanical properties of neat plastic materials. Today, glass fiber reinforced PBT is used in various insulating parts for electrical engineering and electronics, such as electronic relay, communication and automobile applications. Therefore, the investigation of the processing of fiber-reinforced PBT is becoming increasingly important [1-4]. When the glass fiber reinforced flame retardant PBT (GFFRPBT) was mold-injected by five times, how the mold-injection process effect on its carboxyl content, characteristic viscosity of PBT and microscopic appearance were unclear**.** The properties of recycled glass fiber reinforced flame retardant PBT (RGFFRPBT) including, mechanical, electrical and frame retardant properties were still unknown. In this paper, the RGFFRPBT with five times mold-injection effect on its carboxyl content, characteristic viscosity of PBT and microscopic appearance were studied**.** The properties of RGFFRPBT including mechanical, electrical and frame retardant properties were well investigated to demonstrate the properties of RGFFRPBT affected by mold-injection times.

2. Experiment

2.1 Materials

PBT (the virgin PBT samples, having intrinsic viscosity of 1.25 dl/g, were kindly supplied by Du Pont de Nemours (Luxembourg)) was reinforced with 30 wt% glass fibers (Vetrotex EC 10um) (PBT/GF). Portions of 14 wt% flame retardants were added, consisting of brominated phenylethane (USA Chemtura FM2100) and Sb_2O_3 (China Yueyang Chemist Factory), the content of each flame retardant (around 10.3 wt% for brominated phenylethane and 3.7 wt% for Sb_2O_3) corresponds to the content in the combination.

2.2 Preparation of flame retardant PBT with 30 wt% glass fibers (GFFRPBT)

Before compounding, the PBT and brominated phenylethane were dried at 120 °C for 5 h in an air-circulating oven, then PBT, brominated phenylethane and glass fibers were compounded in a twin-screw extruder(L/D=25/1) by two process at the temperatures and screw speed was 250 °C, 25 rpm, respectively. The extruded strands were passed through a water bath and pelletized. The blends were further dried at 120 °C, for 4 h before injection molding the test specimens for properties determination, all bars were injection molded (Germany, ARBUR 420M)at 275 °C , and the mold temperature was kept at 50 °C. The recycled glass fiber reinforced flame retardant PBT collected by injection molding was processed with five times.

2.3 Characterization

The glass fiber reinforced flame retardant PBT recycled (RGFFRPBT) from different times mold-injection were crashed by high-speed knapper and samples with total 0.2 g with the 56 wt% PBT resin was maintained in 25 ml mixture of phenol/1,1,2,2-tetrachloroethane (50/50, w/w) at 120°C for 24 h to achieve a complete solution. The solution then cooled to room temperature and filtered through a disposable membrane filter made from Teflon, then the solution was diluted to 100.0 ml at 20 °C. Carboxyl end-group content (CC) of the PBT was determined according to Pohl's method [5] by titrating a solution of the resin in benzyl alcohol/chloroform with standard NaOH in benzyl alcohol in the presence of phenol red as indicator. Intrinsic viscosity measurements were performed using an Ubbelohde viscometer at 30 °C in a mixture of phenol/1,1,2,2-tetrachloroethane (50/50, w/w). The burnised surfaces of the mold-injection specimen were studied using optical microscope (Japan, OLYMPUS GX51) and the length of GF were averaged. The mechanical properties of such as tensile (ASTM D638-03) and flexural ((ASTM D790-07) properties evaluation were done on a Universal test machine (Reger, RGT-20A). The notched impact strength (ASTM D 256) was performed using a impact tester (Sans Tested, ZBC1400-2) at room temperature. The flammability of samples was studied by UL 94 tests according to IEC 60695-11-10 [6].Glow-wire ignitability test on materials was processed by IEC 60695-2-1/3(1994-03) and the test temperature was 750 °C. To measure the tracking, 50 drops of 0.1% ammonium chloride solution are dropped on the material, and the voltage measured for a 3mm thickness is considered representative of the material performance.

1.Results and Discussion

3.1 Effect of mold-injection times on carboxyl content, characteristic viscosity and length of glass fiber of RGFFRPBT

Tab. 1 Effect of mold-injection times on carboxyl content, characteristic viscosity and length of glass fiber of RGFFRPBT

samples	Carboxyl content (mg/kg)	Intrinsic viscosity	Average length of GF (um)
GFFRPBT	72	0.95	895
RGFFRPBT-1st	135	0.68	767
RGFFRPBT-2nd	203	0.56	616
RGFFRPBT-3rd	244	0.52	590
RGFFRPBT-4th	263	0.46	480
RGFFRPBT-5th	294	0.4	430

Tab. 1 was the carboxyl content, characteristic viscosity and length of glass fiber of RGFFRPBT changed by the mold-injection times. It could see that with the increase of mold-injection times, the carboxyl content of RGFFRPBT increased from 72 mg/kg to 294 mg/kg, while the characteristic viscosity and length of glass fiber of RGFFRPBT decreased with the mold-injection ranged from one times to five times, due to the thermal degradation of the polyesters that takes place during heating [7-9]. These changes indicated that during the thermal-process with high pressure, the shear by mold-injection could snip both the molecular chain and GF, and the degree of damage increased with the mold-injection times. As a result, both the characteristic viscosity and length of glass fiber of RGFFRPBT decreased to rather low value: the characteristic viscosity of samples with three times (presented by RGFFRPBT-3rd) and four times (presented by RGFFRPBT-4th) mold-injection was 0.52, 0.46, respectively.

3.2 Effect of mold-injection times on mechanical properties of RGFFRPBT

Tab. 2 Effect of mold-injection times on mechanical properties of RGFFRPBT

samples	Notched impact strength (J/m)	Tensile strength (MPa)	Flexural strength (MPa)
GFFRPBT	75	115.6	161.9
RGFFRPBT-1st	67	109.3	143.4
RGFFRPBT-2nd	54	96.1	128
RGFFRPBT-3rd	42	85.9	115.1
RGFFRPBT-4th	36	63.7	105.2
RGFFRPBT-5th	33	54.9	95.2

The notched impact strength, tensile strength and flexural strength of RGFFRPBT were shown in Tab.2. With increasing in the mold-injection times, the notched impact strength, tensile strength and flexural strength of samples decreased. The first time recycled GFFRPBT (presented by RGFFRPBT-1st) have an acceptable properties though the mechanical properties have a few decrease. There was almost sudden decrease after twice mold-injection (presented by RGFFRPBT-2nd), and when the mold-injection was increased to five times (presented by RGFFRPBT-5th), the mechanical properties of sample show drastically decreased to 33 J/m (44% of original value), 54.9 MPa (47.5% of original value), 105.2 MPa (58% of original value), respectively. The RGFFRPBT exhibited brittleness, from microstructural point of view, there are several possible reasons for this decrease: low molecular weight, high degree of crystallinity and large spherulites can substantially reduce the ductility of polymeric material[10]. According the Tab.1, it could be explained that the thermal degradation of PBT molecular chain and the length of glass fiber, caused the mechanical properties of RGFFRPBT decreased with the increase of mold-injection times.

3.3 Effect of mold-injection times on flammability and comparative tracking index (CTI) of RGFFRPBT

Tab. 3 Effect of mold-injection times on flammability and CTI of RGFFRPBT

samples	UL94 classification	Glow-wire ignitability time (s)	CTI (V)
GFFRPBT	V-0	8.4	225
RGFFRPBT-1st	V-0	15	225
RGFFRPBT-2nd	V-0	18.7	225
RGFFRPBT-3rd	V-0	18.9	225
RGFFRPBT-4th	V-0	17.1	225
RGFFRPBT-5th	V-0	15.3	225

When GFFRPBT is used in various insulating parts for electrical engineering and electronics, both the glow wire ignitability time and CTI should be evaluated. Tab. 3 was the flammability and CTI data of samples with different mold-injection times, it could be seen that all the samples could obtain V-0 classification and CTI with 225 V show excellent flame retardant properties [11]. The results illuminated that when the recycled glass fiber reinforced flame retardant PBT were multiple processed, it had not influenced the flammability and CTI properties significantly. The reason maybe that microtacticity of surface [12] and brominated phenylethane had no changed by the mold-injection. The reduction in flammability of PBT is a major concern, especially in the domains of transportation, construction, civil and electrical engineering, where fire protection is required[13].The glow-wire ignitability time was changed by mold-injection times, GFFRPBT show the shortest ignitability time about 8.4 second, while the RGFFRPBT-1st burns 15second, and the extinction time increased with the increase of mold-injection times.With the degradation of PBT molecular chain and the length of glass fiber, the molecular force and thermal stability of PBT decreased[14]. When the sample was heated at 750 °C glow wire, it prolongs the ignitability time. Though that, it still could be illuminated that the RGFFRPBT showed good flammability and CTI properties and it was suitable to use in the electrical accessory[14].

4. Conclusion

When the glass fiber reinforced flame retardant PBT were five times mold-injection, its carboxyl content increased while the characteristic viscosity of PBT and the length of glass fiber decreased. After five times mold-injection, all its mechanical properties decreased, especially the notched impact strength and tensile strength drastically decreased, while the flammability and CTI properties not exhibited significantly to change. The results demonstrated that the RGFFRPBT-1st had an acceptance physical properties when it was used in the field of electrical accessory, such as relay. Due to the lower mechanical properties, when the mold-injection times was higher than two, RGFFRPBT was unsuitable to be adopted as engineering polymer solely.

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