ELSEVIER

Polymer 40 (1999) 5957–5960

polymer

Polymer Communication

Thermal and physical properties of poly(phenylene oxide) blends with glass fiber reinforced syndiotactic polystyrene

S.-H. Hwang[a,*], Y.-S. Kim[a], H.-C. Cha[a], J.-C. Jung[b]

[a]*Materials Process Technology R and D Center, Korea Institute of Industrial Technology, Chonan 330-820, South Korea*
[b]*Semiconductor Advanced Research Division, Hyundai Electronics Industries Co., Ltd., Ichon 467-701, South Korea*

Received 19 October 1998; received in revised form 2 February 1999; accepted 6 February 1999

Abstract

Melt blends of poly(phenylene oxide)(PPO) with glass fiber reinforced syndiotactic polystyrene(GFRsPS) were prepared using a single screw extruder. DSC and dynamic mechanical measurements indicate that the PPO/GFRsPS blends are miscible in amorphous state as shown by the existence of a single T_g whose values are blend composition-dependent. From the mechanical test results, it is shown that the tensile and flexural modulus increase, but the tensile and flexural strength do not improve as the GFRsPS content increases. An increase in the melt flow index (MFI) as GFRsPS contents increase implies that the processing problem of neat PPO is improved.

Keywords: Blend; Poly(phenylene oxide); Glass fiber reinforced syndiotactic polystyrene

1. Introduction

Poly(phenylene oxide)(PPO) resins, composed of phenolic monomers, have a very high glass transition temperature(T_g). With the T_g above 200°C, neat PPO is very difficult to process; melting does not begin below 255°C. At extrusion or injection molding temperature of 300–350°C, melt flow is very stiff and special precautions must be taken to minimize oxidation. PPO is soluble in all proportions with general purpose polystyrene (GPPS) and other styrenic polymers, which allows blends or alloys to be produced commercially by GE Plastics under the trade name Noryl [1,2]. PPO–GPPS blend resin is cream colored and characterized by excellent melt flow. Added to this, electrical properties are outstanding and moisture absorbance is low. With excellent dimensional stability, the alloys can be made into very large parts. Although hydrolytic stability is very good, organic solvent resistance is poor. This has limited applications such as large automotive parts [3–6]. Therefore, many researches have made attempts to blend this with other component polymers [7–10].

We consider the melt blends of PPO with a glass fiber reinforced syndiotactic polystyrene (GFRsPS). The unmodified syndiotactic polystyrene (sPS) is a crystalline polymer whose benzene rings have a high degree of stereospecificity, and whose properties are completely different from those of GPPS. In addition to having inherent characteristics to the conventional polystyrene—low specific gravity, hydrolytic resistance, good moldability and excellent electrical properties—the fact that sPS has the heat resistance (T_m: 270°C) and chemical resistance of crystalline polymers make it a new engineering plastic.

In this report, miscible PPO/GFRsPS blends were prepared and their thermal and physical properties were investigated.

2. Experimental

2.1. Materials

The unmodified PPO sample used in this study, obtained from General Electric Plastics, had an intrinsic viscosity of 0.42 dl/g measured in chloroform at 25°C. The GFRsPS was supplied from Idemitsu Petrochemical Co., Ltd. The specific gravity is 1.11 and includes 15 wt.% glass fiber.

2.2. Blend preparation

Samples were dry-mixed at the desired composition,

* Corresponding author. Address: Advanced Materials Process Research Team, Materials Process Technology R and D Center, Korea Institute of Industrial Technology (KITECH), 35-3, Hongchon-Ri, Ipjang-Myun, Chonan 330-820, South Korea. Tel.: + 82-417-560-8664; fax: + 82-417-560-8640.

E-mail address: bach@kitech.re.kr (S.-H. Hwang)

0032-3861/99/$ - see front matter
PII: S0032-3861(99)00115-9

Table 1
Thermal characteristics of PPO/GFRsPS blends

PPO/GFRsPS blend ratio	T_g[a] (°C)	T_g[b] (°C)	T_m[c] (°C)	T_c (°C)	HDT[d] (°C)
50:50	155.1	176.3	ND	ND	138.3
67:33	178.7	191.6	ND	ND	153.2
75:25	185.7	201.1	ND	ND	160.5

[a] Measured by DSC.
[b] Measured by DMTA.
[c] Not detected by DSC.
[d] Deflection temperature under 1.82 MPa load(ASTM D-648).

followed by melt-mixing at 280°C in a single screw extruder(Brabender Plasticoder, $L/D = 30$, $D = 2.5$ cm). The test specimens were injection molded using a BOY injection-molding machine. The molding conditions were as follows: barrel temperature 290°C; mold temperature 80°C; injection pressure 700 kg/cm^2; and total cycle time 30 s.

2.3. Characterization

The thermal characterization studies were carried out using a Perkin–Elmer, DSC-7. The heating rate was 20°C/min and calibration of the instrument was carried out using high purity indium and zinc. The crystalline structures of the samples were determined by a wide angle X-ray diffractometer (MAC Science Co., Ltd.) using CuK_α at 100 kV, and 50 mA. The tensile and flexural tests were carried out using Instron UTM (Model 4202). The tensile and flexural properties were determined following the standard procedure described in ASTM D-638 and ASTM D-790, respectively. Notched impact strength was measured using an Izod impact tester (Testing Machines Inc.) (ASTM D-256).

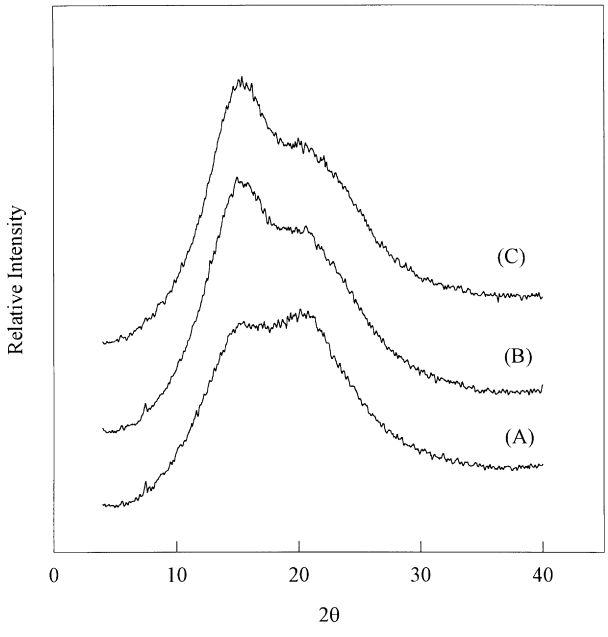

Fig. 1. Wide angle X-ray diffractograms of: (A) PPO:GFRsPS(50:50); (B) 67:33; (C) 75:25.

Dynamic mechanical thermal measurements were performed with a RSA-II (Rheometrics Co., Ltd.), operated in the dual cantilever bending mode, at a frequency of 2 Hz and a heating rate of 5°C/min.

3. Results and discussion

Table 1 reports the thermal properties relative to three PPO/GFRsPS blends. We always detected one T_g whose values are blend composition-dependent. The presence of one T_g indicated that the two polymers had a miscibility in the amorphous state. However the temperatures that correspond to the crystallization and the melting peak are not detected. The X-ray diffraction patterns of the three blends are shown in Fig. 1. When sPS was melt-crystallized, it presents a complex polymorphic pattern, both crystalline forms containing zigzag planar (α and β) in the pure or mixed state, depending on several factors [11]. By adding the PPO in the GFRsPS, the polymorphic behavior of sPS in the blend disappeared. From the thermal and X-ray measurements, it is clear that PPO in the blend strongly interferes with the crystallization of sPS from the amorphous phase. The heat deflection temperature (HDT) of blends show a decreasing trend as GFRsPS contents increase because the T_g of PPO is higher than that of sPS. Also, this phenomenon elucidates that the effect of glass fiber in the PPO/GFRsPS blends does not contribute in HDT property, remarkably.

Summarized physical properties of PPO/GFRsPS blends, including tensile and flexural properties and Izod impact strength and melt flow index, are shown in Table 2. As shown in the Table 2, the tensile modulus increased with the increasing GFRsPS. One can easily see that the flexural modulus also increases as the GFRsPS contents increase. The rising of tensile and flexural moduli should be expected as results of the glass fiber reinforcement effects and inherent nature of sPS. The tensile strength slightly increased and the flexural strength hardly changed with increasing GFRsPS contents. Whereas, elongation at break of PPO/GFRsPS blends decreased as GFRsPS content increased. Usually, an increase of elastic modulus leads to a decrease of elongation at break. The slight increasing in tensile strength can be explained by good miscibility between the

Table 2
Physical properties of PPO/GFRsPS blends

PPO:GFRsPS blend ratio	Elongation at break (%)	Tensile strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural Modulus (MPa)	MFI[a] (g/10 min)	Izod impact strength (kJ/m^2)
50:50	10.0 ± 0.3	73.9 ± 0.5	1317.1 ± 6.9	110.3 ± 0.5	3102.1 ± 25.8	21.4	9.7 ± 1.1
67:33	12.9 ± 0.8	73.0 ± 0.7	1197.4 ± 9.7	111.9 ± 0.6	2796.6 ± 16.4	7.4	8.8 ± 0.5
75:25	13.6 ± 0.8	72.1 ± 0.6	1144.5 ± 6.9	110.2 ± 1.7	2636.0 ± 11.2	4.9	7.9 ± 0.4

[a] Measured at 275°C under 21.6 kg.

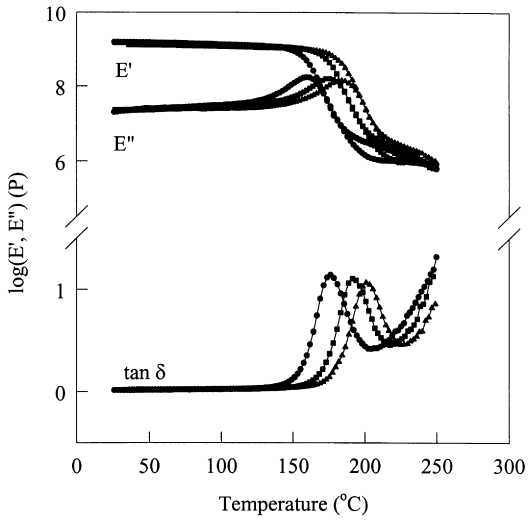

Fig. 2. Dynamic mechanical thermal spectrum of PPO/GFRsPS blends. (●) PPO/GFRsPS (50:50) (■) 67:33 (▲) 75:25.

sPS and PPO. Also, Table 2 shows that the impact strength increases with increasing amount of GFRsPS because the notch sensitivity of a neat sPS is higher than that of PPO and the effect of glass fiber in the blends. As shown Table 2, MFI value rapidly increases with adding GFRsPS content in the blend. This result indicated that adding of GFRsPS in PPO matrix improved processing problem of PPO.

The dynamic mechanical thermal analysis for PPO/GFRsPS blend was performed to investigate the dynamic storage modulus (E') and loss modulus (E'') and tanδ. Fig. 2 shows that the deflection temperature of the storage modulus for PPO/GFRsPS blend was decreased as the GFRsPS was added. This trend shows that the storage modulus of the blend is not related to the glass fiber in the blend, but to the matrix polymer in the blends. The glass transition relaxation of the PPO/GFRsPS blend decreases with increasing GFRsPS content in the blend. This phenomenon agrees quite well with those of DSC measurements and strongly suggests that mixing of PPO and sPS at the segmental level is achieved.

To summarize, DSC and Dynamic mechanical thermal measurements indicated that the PPO/GFRsPS blends appear to be miscible in amorphous state as shown by the existence of a single T_g whose values are blend composition-dependent. In the amount of incorporated PPO in the PPO/GFRsPS blend is more than 50 wt.%, the PPO strongly interferes with a crystallization of sPS as shown in thermal and X-ray measurements. From the mechanical test results, it was shown that the tensile and flexural modulus increases, but tensile and flexural strength is not improved as the GFRsPS content increases. On the other one, adding GFRsPS to PPO leads to decrease in elongation at break, and heat deflection temperature of blends. The melt flow index increasing as GFRsPS contents increase implied that the processing problem of neat PPO is improved.

References

[1] Van der Meer R. US Pat. 4 659 760, April 1987.
[2] Gallucci RR, Avakian RW. US Pat. 4 659 763, April 1987.
[3] Stoeling J, Karasz FE, MacKnight WJ. Polym Engng Sci 1970;10:133.
[4] Prest Jr. WN, Porter RS. J Polym Sci Part A-2 1972;10:1639.
[5] Djordjevic MB, Porter RS. Polym Engng Sci 1983;23:650.
[6] Composto RJ, Kramer E. Macromolecules 1988;21:2580.
[7] Chiang C-R, Chang F-C. Polymer 1997;38:4807.
[8] Gallucci RR. US Pat. 5 260 374, December 1993.
[9] Coumans MA, Heikens D, Sjoerdsma SD. Polymer 1981;21:205.
[10] Chiang C-R, Chang F-C. J Appl Polym Sci 1996;61:2411.
[11] Guerra G, Vitagliano V, De Rosa C, Petraccone V, Corradini P. Macromolecues 1990;31:1539.