# Functional MBS Impact Modifiers for PC/PBT Alloy

#### WILLIAM T. W. TSENG,<sup>1</sup> J.-S. LEE<sup>2</sup>

<sup>1</sup> No. 31, Ta Kung Rd., Tayuan Kou Ta Industrial Park, Tayuan Hsiang, Taoyuan Hsein, Taiwan, Republic of China

<sup>2</sup> Union Chemical Laboratories, Industrial Technology Research Institute, Hsinchu, Taiwan

Received 10 May 1999; accepted 10 October 1999

ABSTRACT: Functional group containing MBS impact modifiers for the poly carbonate/ poly(butylene terephthalate (PC/PBT) alloy were synthesized and characterized in this study. The novel MBS consisted of three layers, in which the inner, middle, and outer layers were styrene butadiene rubber (SBR) latex, polystyrene, and a copolymer of the methacrylic acid (MMA) and the functional group containing vinyl monomer, respectively. Three different kinds of the functional monomers were used in this study, glycidyl methacrylate (GMA), acrylamide (AAM), and MAA. The functional group was used to improve the adhesion between the MBS and the PC/PBT alloy. Our results showed that the layer composition of the MBS exhibited a significant effect on the impact strength. A large variation of the impact strength from 14.1 to 23.6 ft-lb/in. was observed when the SBR content increased from 30 to 70%. An optimum amount (4-6%)of the functional monomer enhanced the adhesion between the MBS and the PC/PBT alloy and thus improved the impact strength. Furthermore, a much smaller amount of the functional group containing MBS in the PC/PBT allow than the conventional MBS could obtain an impact strength as high as 25.2 ft-lb/in. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 76: 1280-1284, 2000

Key words: functional MBS; PC/PBT alloy; core-shell; impact strength

## **INTRODUCTION**

Polycarbonate/poly(butylene terephthalate) (PC/ PBT) alloy has been widely used in shaped articles because of its easy processability, good size stability, heat resistance, and solvent resistance.<sup>1-10</sup> However, the brittle problem of the PC/ PBT alloy results in a low-impact strength at low temperatures and thus limits its applications. Modifiers such as impact modifiers,<sup>3-10</sup> compatibilizers,<sup>1-2</sup> and glass fiber<sup>10</sup> were used to improve the physical properties of the PC/PBT alloy.

Among these modifiers, MBS impact modifiers demonstrated a significant impact-modifying effect at low temperatures.<sup>6–9</sup> However, it required a large amount of the MBS impact modifiers added into the PC/PBT alloy to obtain a significant impact strength. For example, Chacko et al.<sup>6</sup> reported the addition of 20 wt % MBS impact modifier to obtain an acceptable impact strength. The reason for the large amount of the MBS required for the PC/PBT alloy is probably because the conventional MBS has no suitable interaction between the interface of the MBS and the PC/PBT alloy. Therefore, the compatibility of the conventional MBS with the alloy is poor. Consequently, the impact-modifying effect of the conventional MBS is limited. Furthermore, such a large amount of MBS adversely affects some physical properties such as heat distortion temperature.

In this study, a functional-group-containing MBS impact modifier was used to provide a suitable interaction between the MBS and PC/PBT alloy. Therefore, the improved MBS can be evenly

Correspondence to: W. T. W. Tseng.

Journal of Applied Polymer Science, Vol. 76, 1280–1284 (2000) © 2000 John Wiley & Sons, Inc.



**Figure 1** The structure of the functional-group-containing MBS impact modifier, the inner layer (I): rubber: SBR; the middle layer (II): SM polymer; and the outer layer (III): copolymer of MMA and a functional group (F) containing vinyl monomer.

dispersed in the PC/PBT and the impact-modifying effect can be enhanced. The structure of this novel MBS impact modifier was shown in Figure 1, which was composed of three layers. The inner layer was the styrene butadiene rubber (SBR), which mainly absorbs the outer impact energy. The middle layer was polystyrene, which mainly connects the inner and outer layers. The outer layer includes a copolymer of methyl methacrylate (MMA) and a functional-group-containing vinyl monomer, which imparts compatibility of the functional-group-containing MBS with the PC/ PBT alloy. In the study, glycidyl methacrylate (GMA), acrylamide (AAM), and MAA were used as the functional-group-containing vinyl monomers. The effects of the SBR content and the functional monomer GMA composition on the impact strength and ductile brittle transition temperature (DBTT) were also investigated in this study.

#### **EXPERIMENTAL**

#### Materials

PC and PBT were obtained from General Electric Plastics (trade name: Lexan 171) and Taiwan Shinkong Synthetic fibers (trade name: D201), respectively. Styrene butadiene copolymer (SM/BR = 25/75, 0.5% divinyl benzene, ph = 9, mean particle size = 180 nm) was provided by Taiwan Hopax Chems. Mfg. Co. Sodium lauryl sulfate (SLS; > 95%, TCI), styrene monomer (SM; > 99%, TCI), MMA (99%, Jassen Chimica), sodium formaldehyde sulfoxylate (SFS; 98%, Acros), *t*-butyl hydroperoxide (TBHP; 90%, Aldrich), AAM (> 98%, TCI), MAA (> 99%, TCI), GMA (> 95%, TCI), and antioxidants (Irgnox 1076, CIBA, and Weston 619, GE) were used without further purification.

Table I The Composition for the MBS Impact Modifiers for the PC/PBT Alloy

Components (parts)	SBR Content or Functional Group							
	SBR1	SBR2	SBR3	SBR4	SBR5	AAM	MAA	GMA
Inner layer (core)								
SBR latex (I)	30	40	50	60	70	60	60	60
SLS	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Water	100	100	100	100	100	100	100	100
Middle layer								
SM	35	30	25	20	15	20	20	20
SFS	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
TBHP	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Water	70	70	70	70	70	70	70	70
Outer layer (shell)								
MMA	35	30	25	20	15	19.6	19.6	19.6
Functional group	_			_		0.4	0.4	0.4
SFS	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
TBHP	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Water	80	80	80	80	80	80	80	80



**Figure 2** The effect of the SBR content in MBS on the impact strength and DBTT of the PC/PBT alloy.

#### **Synthesis**

# Preparation of the Functional-Group-Containing MBS

The inner layer was prepared with various amounts of SBR latex as shown in Table I, 0.5 parts of sodium lauryl sulfate, and 100 parts of water in a reaction tank. The mixture was heated to 50°C under nitrogen with constant stirring. Then, the middle layer containing a various portion of styrene monomer as shown in Table I, 0.5 parts of SFS, 0.25 parts of TBHP, and 70 parts of water were added dropwise into the reaction tank over a period of 2 h. Subsequently, the outer layer containing MMA, or a functional-group-containing vinyl monomer, 0.5 parts of SFS, 0.25 parts of TBHP, and 80 parts of water were added dropwise over a period of 2 h. The mixture was stirred for an additional hour to obtain an MBS latex. The MBS latex was mix-treated with an antioxidant (Irganox1076 and Weston 619), then isolated by freezing, condensing with an acid, salting out or spray drying, filtered, and dried to obtain the MBS powders. Table I shows the composition for preparing the MBS impact modifiers.

Blended were 50 parts of PC, 50 parts of PBT, 10 parts of the prepared MBS, and 0.1 part of antioxidant with a twin-screw machine and extruded to obtain a test sample for determining the impact strength.

#### Characterization

The notched Izod impact strength of the tested sample was determined by a TMI impact tester at 23°C according to ASTM D256 method. The DBTT was determined by the relationship between the impact strength and the tested temperature from the ASTM D256 method.

### **RESULTS AND DISCUSSION**

#### Effect of the SBR Content

Figure 2 and Table II show the effect of the SBR content in the MBS on the impact strength and DBTT for the PC/PBT alloy. A large variation of the impact strength from 14.1 to 23.6 ft-lb/in. was observed when the SBR content increased from 30 to 70%. The above results can be explained by the impact-modification mechanism proposed by Schmitt and Keskkula.<sup>11</sup> They suggested that the rubbery particle (core material in this case) functions as a stress concentrator and stress arrestor. A too-thick shell would terminate the propagation of the impact stress, thereby making the mechanism incomplete. Hence, only a slight impact improvement would result. Furthermore, our previous publication on the core/shell impact-modifier/ PVC blends also observed similar phenomena as the present study.<sup>12</sup> Hence, the experimental results on Table II can be explained as follows. When the SBR content increased from 30 to 50% in the MBS, the outer layer (shell) was probably too thick to propagate the impact stress into the inner layer (rubber core) and resulted in a poor impact strength. On the other hand, the 70% SBR content resulted in a thin shell and the compatibility between the MBS and the PC/PBT alloy became poor. Hence, the impact strength reduced from the 60% SBR content to the 70% SBR con-

 
 Table II
 Effect of the SBR Content in the MBS Impact Modifiers on the Impact Strength of the PC/ PBT Alloy

	SBR Content						
Property	SBR1	SBR2	SBR3	SBR4	SBR5		
Impact strength (ft-lb/in.)	$14.1~\pm~0.7$	$15.7~\pm~0.8$	$15.1~\pm~0.4$	$23.6~\pm~0.9$	$14.3 \pm 0.6$		



**Figure 3** The effect of the GMA content in MBS on the impact strength of the PC/PBT alloy.

tent. The DBTT in Figure 2 reduced from 0 to  $-17^{\circ}$ C as the SBR content increased from 30 to 70%, which was due to the low glass transition temperature of SBR (-59 to  $-64^{\circ}$ C).<sup>12</sup> From the above study, the 60% SBR content in the MBS was chosen for the investigation of the functional-group-containing MBS impact modifiers.

#### Effect of the GMA Content on the Impact Strength

Figure 3 shows the effect of the functional group GMA content in the MBS on the impact strength and DBTT of the PC/PBT alloy. The impact strength increased from the 0% GMA content with 23.6 ft-lb/in. to 4 and 6% with 24.1 ft-lb/in. and decreased in the 8% with 22.9 ft-lb/in. and 10% with 18.1 ft-lb/in. One possibility of improving the adhesion between the MBS and the PC/ PBT alloy by GMA is the epoxy functional group reacted with the OH end group of PBT. A further investigation is required to elucidate the adhesion mechanism. Therefore, the 4 and 6% of the GMA content observed the improvement of the adhesion effect. An excess amount of the GMA content enhanced the adhesion between the MBS and the PC/PBT alloy but might inhibit the crazing and the shear yielding. It can be used to explain the

poor impact strength of the 8 and 10% of the GMA content in the MBS. Hence, 4% of the functional group was chosen for studying the effect of the functional group.

# Effect of the Functional Group on the Impact Strength

Table III shows the effect of the functional group in MBS on the impact strength and DBTT of the PC/PBT alloy. All three functional groups containing MBS, GMA, AAM, and MMA showed a higher impact strength than the MBS impact modifier without a functional group. Furthermore, the addition of 10% of the functional MBS in the PC/PBT alloy exhibited an impact strength comparable with adding 20% of the conventional MBS.<sup>13,14</sup> Therefore, the proposed functionalgroup-containing MBS impact modifiers could enhance the adhesion between the MBS and the PC/PBT alloy and result in an excellent impact strength.

# CONCLUSIONS

Three different kinds of functional-group-containing MBS impact modifiers for the PC/PBT alloy were synthesized and characterized, GMA, AAM, and MAA. Our results show that the layer composition of the MBS exhibited a significant effect on the impact strength. If the shell of the MBS was too thick, the impact stress could not effectively propagate to the rubber core. On the other hand, the compatibility between the MBS and the PC/PBT alloy became poor if the shell is too thin. An optimum amount of the functional group enhanced the adhesion between the MBS and the PC/PBT alloy and thus improved the impact strength. Furthermore, a much smaller amount of the functional group containing MBS in the PC/ PBT allow than the conventional MBS could obtain an impact strength as high as 25.2 ft-lb/in.

Table III Effect of the Functional Group on the Impact Strength and DBTT of PC/PBT Alloy

Property	Functional Group					
	GMA	AAM	MAA	Blank		
Impact strength (ft-lb/in.) DBTT (°C)	$\begin{array}{rrr} 24.0 \ \pm \ 0.3 \\ -7 \end{array}$	$\begin{array}{rrr} 24.9 \ \pm \ 0.4 \\ -10 \end{array}$	$25.2 \pm 0.4 \ -2.5$	$23.6~\pm~0.9\ -5$		

We thank the technical assistance of our former colleague, Mr. Cheng-Lung Chao.

### REFERENCES

- Vol. 154, Deanin, R. D.; Cruguola, A. M., Eds.; American Chemical Society: Washington, DC, 1976.
- Rubber-Toughened Plastics, Adv. Chem. Ser., Vol. 222, Riew, C. K., Ed., American Chemical Society: Washington, DC, 1989.
- Baron, A. L.; Bailey, J. V. (to Mobay Chemical Co.), U.S. Pat. 4,034,016, 1977.
- Neuray, D.; Nouvertne, W.; Binsack, R.; Rempel, D.; Muller, P. R. (to Bayer AG), U.S. Pat. 4,482,672, 1984.
- Chung, J. Y. J.; Neuray, D.; Witman, M. W. (to Mobay Chemical Co.) U.S. Pat. 4,554,314, 1985.

- Chacko, V. P.; DeSito, P.; Baum, G. A. (to Celanese Engineering Resins, Inc.) U.S. Pat. 4,677,150, 1987.
- Saitoand, A.; Itoi, H. (to GE Plastics, Japan) U.S. Pat. 5,455,302, 1995.
- Wittman, D.; Schoeps, J.; Piejko, K.-E.; Weirauch, K. (to Bayer AG), U.S. Pat. 5,484,846, 1996.
- Hoshino, M.; Ogihara, T.; Kadoto, Y.; Maruyama, K. (to Kureha Kagaku Kogyo Kabushiki Kaisha and Tsutsunaka Plastic Kogyo Kabushika Kaisha) U.S. Pat. 5,516,842, 1996.
- Ishiwa, K.; Itoi, H. (to General Electric Co.) U.S. Pat. 5,565,515, 1996.
- 11. Schmitt, J. A.; Keskkula, H. J Appl Polym Sci 1960, 3, 132.
- Li, K.-H.; Tang, K. C.; Lee, J.-S.; Chao, C.-L.; Chang, R.-K. J Vinyl Additive Technol 1997, 3, 17.
- 13. Jpn Pat. 01141944, 1987.
- 14. Jpn Pat. 01204962, 1988.