# Application of Phase Dispersion–Crosslinking Synergism on Recycling Commingled Plastic Wastes

ZHENGPING FANG, MINFENG ZENG, GUOPING CAI, CHENGWEI XU

Institute of Polymer Composites, Zhejiang University, Hangzhou 310027, China

Received 13 December 2000; accepted 14 March 2001

ABSTRACT: A tetra-component blend, consisting of low-density polyethylene (LDPE), polyvinyl chloride (PVC), polypropylene (PP), and polystyrene (PS), was studied as a model system of commingled plastic wastes (LDPE/PVC/PP/PS, mass ratio: 70/10/10/ 10). Effects of chlorinated polyethylene (CPE), ethylene-propylene-diene monomer (EPDM), styrene-butadiene-styrene (SBS), and their mixture (CPE/EPDM/SBS, mass ratio: 2/2/2) on the mechanical properties and morphology of the system were investigated. With addition of several elastomers and their mixture, the tensile strength of the blends decreased slightly, although both the elongation at break and the impact strength increased. Among these elastomers, EPDM exhibited the most significant impact modification effect for the tetra-component blends. SBS and the mixture have a good phase-dispersion effect for the tetra-component blend. By adding a crosslinking agent [dicumyl peroxide (DCP)], the mechanical properties of the tetra-component blends also increased. When either SBS or the mixture was added to the blend together with DCP, the probability that the crosslinking agent (DCP) would be at the interface improved because of the phase-dispersion effect of SBS. Therefore, more co-crosslinked products will form between LDPE and other components. Accordingly, remarkable improvement of the interfacial adhesion and hence the mechanical properties of the tetra-component blends occurred. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 2947-2952, 2001

**Key words:** phase dispersion-crosslinking; recycling; commingled plastic wastes; polymer blends

# **INTRODUCTION**

Nowadays, an increasing number of people are coming to the realization that plastic wastes are a potential worldwide source of raw materials. As we know, plastic wastes are usually commingled thermoplastics, that is, a mixture of polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polystyrene (PS), and other common plastics. To recycle the commingled plastic waste in the form of blends is very attractive because it avoids the difficult task of separation. Because of their poor compatibility, the mechanical properties of polymer blends from these common thermoplastics are inferior. Many studies have been done on the modification of properties of commingled plastic wastes.<sup>1-10</sup> Vivier et al.,<sup>11</sup> for instance, used peroxide to modify the properties of a model system of commingled plastic wastes with a simulated waste plastic fraction (LDPE/HDPE/ PVC/PS/HIPS/PP/PET, mass ratio: 45/15/15/7.5/ 7.5/5/5). Recently, Ha et al.<sup>12</sup> reported their studies on the recycling of commingled plastics by cellulose reinforcement. They found that the

Correspondence to: Z. Fang (zpfang@mail.hz.zj.cn). Contract grant sponsor: National Natural Science Foundation; contract grant number: 59773024.

Journal of Applied Polymer Science, Vol. 82, 2947–2952 (2001)  $\circledcirc$  2001 John Wiley & Sons, Inc.

Materials	Code	Character	Supplier
Low-density polyethylene (LDPE)	2F2B	MI: 2.0 g/10 min; Density: 0.922 g/cm <sup>3</sup>	Shanghai Petrochemical Co., Ltd., China
Polyvinyl chloride (PVC)	WS-800S	Average polymerization degree: 750–850	Shanghai Chlor-Alkali Chemical Co., Ltd., China
Polypropylene (PP) Polystyrene (PS)	RD701 535	MI: 7.0 g/10 min	Neste Works, Finland Zhanjiang Zhongmei Chemical Industries Co., Ltd., China
Styrene-butadiene-styrene (SBS)	YH791		Baling Petrochemical Co. Ltd., China
Ethylene–propylene–diene rubber (EPDM)	4045	Containing 56% (wt %) ethylene	Jiling Chemical Works, China
Chlorinated polyethylene (CPE)		CI: 35%	Wuhu Chemical Industries Works, China
Dicumyl peroxide (DCP)			Gaoqiao Petrochemical Co., China

Ta	ble	Ι	Main	Μ	lateri	ials	U	Jsed	in	This	St	ud	y
----	-----	---	------	---	--------	------	---	------	----	------	----	----	---

properties of the commingled blends were much improved by the reactive blending of cellulose with the commingled blends by peroxide and maleic anhydride.

In our previous works,<sup>13–16</sup> we found that the mechanical properties of the binary incompatible polymer blends could be upgraded remarkably by the synergism between crosslinking agent and compatibilizer or solid-phase dispersant. The kernel of the phase dispersion-crosslinking synergism is the enhanced probability of the crosslinking agent existing at the interface of incompatible polymer blends, thus forming more co-crosslinked products. Good synergism will occur and the mechanical properties will be modified as a result of the improvement in interfacial adhesion. In principle, such a synergism should also exist in more complicated blends. Our preliminary examination<sup>17</sup> showed that it really works for blends consisting of more than two components.

In the present work, we chose PE/PVC/PP/PS with a simulated waste plastic fraction (70/10/10/10) as a model system of commingled plastic wastes. The purpose was to determine to what extent the mechanical properties and morphology of the model system can be upgraded with addition of several elastomers and dicumyl peroxide (DCP).

## EXPERIMENTAL

### Materials

The main materials are listed in Table I.

#### Processing

A plasticized polyvinyl chloride (PVC) material was compounded according to the constituents in Table II. PP, PS, and PVC (mass ratio: 10/10/10) were premixed for 10 min in a two-roll mill (front roll 150–155°C; rear roll 135–140°C). PE, elastomers, and the PP/PS/PVC premixed sheet were milled for 10 min (front roll 125–130°C; rear roll 105–110°C). Finally, DCP was added and mixed for another 5 min. The well-mixed pieces were then transferred to a preheated compression mold and hot-pressed at 170°C under 14 MPa for 12 min to give a blend sheet for testing.

# Characterization

A Hitachi S-570 scanning electron microscope (Hitachi, Japan) was used to examine the morphology of the impact fracture surfaces of the samples. The tensile test was performed according to GB1040-89 (similar to ISO527-1993) on an XL-250 tensile instrument at a crosshead speed of

Table II	Composition	of Plasticized	Polyvinyl
Chloride			

Material	Content (g)
Polyvinyl chloride	100
Dioctyl phthalate	32
Tribasic lead sulphate	3
Dibasic lead phosphite	3
Stearic acid	1

50 mm/min. The impact test was performed according to GB1043-79 (similar to ISO179-1993) on an XCJ-4 Charpy impact instrument. The temperature of the testing room was  $23 \pm 2$ °C. The results of the tensile test and impact test were obtained by averaging the results of five measurements.

# **RESULTS AND DISCUSSION**

It was previously reported that chlorinated polyethylene (CPE) acts as a compatibilizer for LDPE and PVC pairs<sup>2</sup>; so too, ethylene-propylenediene monomer (EPDM) for LDPE and PP pairs,<sup>18</sup> and styrene-butadiene-styrene (SBS) for LDPE and PS pairs.<sup>19,20</sup> The addition of a compatibilizer into the incompatible polymer blends enhanced the mechanical properties because the additives locate at the interface and increase the interfacial adhesion. Figure 1 shows the morphologies of the impact fracture surfaces of the samples. In this tetra-component blend, PVC, PS, and PP dispersed in the major component (LDPE), that is, LDPE acts as the matrix and the other three components act as the domain. Given their poor compatibility, as seen from Figure 1(a), the domain size is very large (~ 10  $\mu$ m) and appears spherical.

The addition of CPE (6 g) appears to have little effect on the compatibilization of the tetra-component blend [Fig. 1(b)]. The main reason for this behavior is that its compatibility with PP and PS is poor. As Figure 1(c) shows, when EPDM (6 g) was added, although the size of the domain is still large, the toughness of the matrix is significantly modified. When we concentrated on SBS [Fig. 1(d)], we found that the phase dispersion of the tetra-component was well upgraded. It reveals that SBS works as a good solid-phase dispersant, in spite of the fact that SBS does not appear to have the means of physical association with the PVC component of the blend. SBS can perfectly regulate the differing viscosity levels of the components of the blend.<sup>19–22</sup> Therefore, the phase dispersion will be upgraded during the blending process and, because of the limited activity of the polymer chains, this phase morphology will be maintained after the sample is cooled, even though it is not thermomechanically stable.<sup>16</sup> Almost the same phenomenon happened with the addition of the mixture CPE/EPDM/SBS (2/2/2); as Figure 1(e) shows, the size of the domain decreases remarkably. However, the interface is

clear and the improvement of the phase adhesion caused by the mixture is limited. Thus, its effects on modification of the mechanical properties of the blend are poor (Fig. 2). Furthermore, its effect of upgrading of the phase dispersion on the blend must be attributed to the solid-phase dispersant (SBS).

The mechanical properties (Fig. 2) are consistent with the morphology of their impact fracture surface. Without the addition of other additives, the mechanical properties of the tetra-component are reasonably poor. As Figure 2 shows, the modification of EPDM on the impact strength of the blend is the best. It seems that the mechanical properties of a multicomponent blend having spherical domains are mainly dependent on the matrix properties. Thus, the behavior of EPDM of the blend is explicable in view of its good modification of toughness on the matrix. The other two elastomers have little effect on the impact strength of the blend. Furthermore, the effect of these three elastomers on the elongation at break of the blend is limited. It reveals the interfacial adhesion does not improve when the elastomers are added alone. We also found that the tensile strength decreased with the addition of any of these three elastomers or their mixture. As a conclusion, none of these three elastomers and their mixture acts as a good compatibilizer for this tetra-component blend. However, SBS and the mixture have a good phase-dispersion effect for the tetra-component blend. Moreover, EPDM exhibited the most significant impact modification effect for the tetra-component blends.

It has been found<sup>14-17</sup> that the core of the phase dispersion-crosslinking synergism for binary blends is the increasing probability of crosslinking agent existing at the interface when the phase dispersion of incompatible polymer blends is enhanced by a solid-phase dispersant. Therefore, more co-crosslinked products will form. Good synergism will occur and the mechanical properties will be modified as a result of the improvement of the interfacial adhesion. With the addition of the crosslinking agent (DCP), as seen from Figure 1(f), the phase dispersion of the tetracomponent blend is not sufficient. The contact area between LDPE and the other three components is small. Thus, DCP mainly initiates the crosslinking of LDPE itself and the product of co-crosslinking of LDPE and other components is rare. Although the spherical domain becomes a bit flat, its size is still large.



30um

X1.00K

A Partice



**Figure 2** Influences of several elastomers and DCP on the impact strength (a), tensile strength (b), and tensile elongation (c) of PE/PVC/PP/PS (mass ratio: 70/10/10/10) blend. Sample code: (A) without elastomers; (B) with 6 g CPE; (C) with 6 g EPDM; (D) with 6 g SBS; (E) with 2 g each of CPE, EPDM, and SBS.

The properties of the blends were upgraded mainly because of the crosslinking of the matrix (LDPE). In comparison, the blend to which DCP and the mixture were simultaneously added had a smaller domain size and a much more indistinct interface [Fig. 1(g)]. The blend was characterized by improved toughness fracture, and the elongation at break and tensile strength of the blend were also well upgraded. In this case, the mixture can promote the phase dispersion of the blend and cause the phase size to decrease. Then, the contact area between LDPE and the other three components is upgraded, and the probability that the crosslinking agent (DCP) would exist at the interface of incompatible polymer blends is also improved. Meanwhile, more co-crosslinked products will form between LDPE and the other three components. The co-crosslinking action can improve the phase interfacial adhesion. Furthermore, its product proved to be a suitable compatibilizer of the blend; thus the mechanical properties of the blend were remarkably modified.

From Figure 2, we can also find the same behavior when SBS and DCP were simultaneously added. It reveals that the phase dispersioncrosslinking synergism for binary blends is also adaptable for this tetra-component polymer blend. To the other two elastomers, CPE and EPDM, their synergism with DCP are comparably inferior, which the elongation at break of the blends [Fig. 2(c)] can demonstrate. On simultaneously adding several elastomers and DCP to PE/PVC/PP/PS (70/10/10/10) blend, if we take the mechanical properties for comparison, we can find that the better the effects of elastomer on the phase dispersion of the blend, the better its synergism with DCP.

## CONCLUSIONS

In this study we have determined the influences of several elastomers, their mixture [CPE/EPDM/ SBS (2/2/2)], and crosslinking agent (DCP) on the morphology and properties of tetra-component blends. This blend system is composed of common plastics with a simulated commingled waste plastics fraction (PE/PVC/PP/PS: mass ratio 70/10/10/ 10). For this tetra-component blend system, CPE, EPDM, SBS, and their mixture were tested such that it was determined that they do not act as compatibilizer for the blend. However, SBS and the mixture have a good phase-dispersion effect for the tetra-component blend. EPDM exhibited

**Figure 1** Scanning electron microscope observations of the impact fracture surface of the blends: (a) PE/PVC/PP/PS (70/10/10/10); (b) PE/PVC/PP/PS/CPE (70/10/10/10/6); (c) PE/PVC/PP/PS/EPDM (70/10/10/10/6); (d) PE/PVC/PP/PS/SBS (70/10/10/10/6); (e) PE/PVC/PP/PS/CPE/EPDM/SBS (70/10/10/10/2/2); (f) PE/PVC/PP/PS/DCP (70/10/10/10/0.6); (g) PE/PVC/PP/PS/CPE/EPDM/SBS/DCP (70/10/10/10/2/2/2).

the most significant impact modification effect for the tetra-component blends. The phase dispersion-crosslinking synergism for binary blends is also adaptable for this tetra-component polymer blend. When SBS or the mixture is added to the blend together with DCP, good synergism can be realized. Therefore, more co-crosslinked products will form between LDPE and the other three components. Meanwhile, the mechanical properties of the blend were remarkably modified. To upgrade the mechanical properties of PE/PVC/PP/PS (70/ 10/10/10) as a model of commingled plastic wastes, the phase dispersion-crosslinking synergism technique is a good technique. Using this technique, it is possible to achieve an acceptable level of qualities in the composite products for the reuse of the materials in demanding applications.

This work was supported by the National Natural Science Foundation of China (Project No. 59773024).

# REFERENCES

- Paul, D. R.; Vinson, C. E.; Locke, C. E. Polym Eng Sci 1972, 12, 157.
- Paul, D. R.; Locke, C. E.; Vinson, C. E. Polym Eng Sci 1973, 13, 202.
- Park, H. D.; Cho, W. J.; Ha, C. S.; Kwon, S. K. Polym Recycl 1996, 2, 277.
- Park, H. D.; Park, K. O.; Cho, W. J.; Ha, C. S.; Kwon, S. K. Polym Recycl 1996, 2, 283.
- De Groote, Ph.; Grodard, P.; Vanharan, G. Chim Nowv 1996, 14, 1635.

- Vanharan, G.; De Groote, Ph.; Grodard, P. Chim Nowv 1997, 16, 1659.
- Liao, B.; Huang, Y. H.; Cong, G. M. J Appl Polym Sci 1997, 66, 1561.
- Fortelny, I.; Krulis, Z.; Michalkova, D.; Horak, Z. Angew Makromol Chem 1999, 270, 28.
- Ha, C. S.; Park, H. D.; Cho, W. J. J Appl Polym Sci 2000, 76, 1048.
- Suarez, J. C. M.; Mano, E. B.; Tavares, M. I. B. J Appl Polym Sci 2000, 78, 899.
- 11. Vivier, T.; Xanthos, M.; Breant, P. ANTEC '93, 1993, 1227.
- Ha, C. S.; Park, H. D.; Cho, W. J. J Appl Polym Sci 1999, 74, 1531.
- Xu, C. W.; Fang, Z. P.; Zhong, J. H. Angew Makromol Chem 1993, 212, 45.
- Fang, Z. P.; Xu, C. W.; Bao, S. J.; Zhao, Y. F. Chin Sci Bull 1995, 40, 1671.
- Fang, Z. P.; Xu, C. W.; Bao, S. J.; Zhao, Y. F. Polymer 1997, 38, 131.
- Xu, C. W.; Fang, Z. P.; Zhong, J. H. Polymer 1997, 38, 155.
- Fang, Z. P.; Zeng, M. F.; Xu, C. W. in Preprints of 6th Pacific Polymer Conference, Guangzhou, China, 1999; p. 528.
- Blom, H. P.; Teh, J. W.; Rudin, A. J Appl Polym Sci 1996, 61, 959.
- 19. Fayt, R.; Jerome, R.; Teyssie, Ph. J Polym Sci Polym Phys Ed 1982, 20, 2209.
- Fang, Z.; Xu, C.; Xiang, Y.; Wu, J.; Xu, H. Chin J Polym Sci 1998, 16, 207.
- Zheng, Q.; Araki, O.; Takigawa, T.; Takahashi, M.; Masuda, T. J Soc Mater Sci Jpn 1996, 45, 1284.
- Zheng, Q.; Zhao, T. Chin J Mater Res 1998, 12, 225.