

# Compatibilizers for Recycling of the Plastic Mixture Wastes.

## II. The Effect of a Compatibilizer for Binary Blends on the Properties of Ternary Blends

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**ABSTRACT:** In this article, we discuss the effect of a compatibilizer for binary blends on the properties of ternary blends composed of high-density polyethylene (HDPE), polypropylene (PP), or polystyrene (PS) and poly(vinyl chloride) (PVC) virgin polymers with a simulated waste plastics fraction. Chlorinated polyethylene (CPE), ethylene-propylene rubber (EPR), and their 1/1 (w/w) mixture were tested as compatibilizers for the HDPE/PP/PVC ternary blend. CPE, styrene-ethylene-propylene block copolymer (SEP), or their 1/1 (w/w) mixture were tested as compatibilizers for the HDPE/PS/PVC ternary blend. The composition of the ternary blends were fixed at 8/1/1 by weight ratio. The amount of the compatibilizer was 3 phr. Rheological, mechanical, and thermal properties were measured. For the 8/1/1 HDPE/PP/PVC ternary blends, the tensile strength was slightly decreased, but the impact strength was significantly increased by adding EPR, CPE, or their mixture. EPR exhibited the most significant impact modification effect for the ternary blends. In a similar way, for 8/1/1 HDPE/PS/PVC ternary blends, on adding SEP, CPE, or their mixture, the tensile strength was slightly decreased, but the impact strength was noticeably increased. It was found that the SEP worked much better as an impact modifier for the ternary blends than CPE or the SEP/CPE mixture did. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 76: 1048–1053, 2000

**Key words:** compatibilizers, plastic waste mixture, polymer blend, recycling

### INTRODUCTION

A substantial part of municipal solid waste streams is composed of mixed polymers from laminated or composite materials. The processing of plastic mixtures for recycling has been attempted with some success, but poor mechanical properties and uncertain economic value limit their more versatile reuse.<sup>1–15</sup>

High-density polyethylene (HDPE), polypropylene (PP), and poly(vinyl chloride) (PVC), as well as polystyrene (PS) are among the most common plastic wastes, because they are among the

most frequently used commercial plastics in our daily lives as well as in industries.<sup>9</sup> We can recycle mixed waste plastics in the form of blends. This approach to reuse is attractive, because it avoids the difficult task of separation. Usually, enhancement of impact strength, dimensional stability, stress cracking, and improvement of processibility have been the prime goals of blending. As a consequence, academic and industrial interest in virgin and recycled polymer blends is rapidly expanding.

One technical problem associated with plastics waste is its heterogeneous composition. The properties of blends are usually inferior because of the lack of compatibility of different polymers, when no addition of a compatibilizing agent has been made.<sup>8–15</sup> Compatibilizers have been developed

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for binary blends. For example, chlorinated polyethylene (CPE) has been investigated as a compatibilizer for HDPE and PVC pairs.<sup>10,14</sup> Styrene-ethylene-propylene block copolymer (SEP) and ethylene-propylene rubber (EPR) have been used for HDPE/PS<sup>11</sup> and HDPE/PP blends,<sup>12,13</sup> respectively. The compatibilizing effect of such a compatibilizer has been known to be attributable to the enhanced interfacial adhesion between two component polymers, because the compatibilizer usually possesses the units of the parent polymers. Our main concern is to investigate whether such a compatibilizer for binary blends can be effective for ternary blends containing either of the parent polymers as one component.

Although a number of studies<sup>16–20</sup> have been made on the effect of a compatibilizer for binary blends, few studies<sup>21,22</sup> have reported on the effect of a compatibilizer on the properties of ternary blends consisting of any of the parent polymers and the third polymer whose compatibility is not affected by the compatibilizer.

In this article, therefore, we report the effect of a compatibilizer for binary blends on the properties of ternary blends composed of HDPE, PP (or PS), and PVC virgin polymers with a simulated waste plastics fraction. CPE, SEP, EPR and their mixtures were tested as compatibilizers.

## EXPERIMENTAL

The materials used in this study were HDPE ( $M.I = 0.89$  (g/10 min) at 190°C,  $M_w = 100,200$ ) supplied by Korea Petrochemicals (M 830), PP ( $M.I = 3.5$  (g/10 min) at 230°C,  $M_w = 60,000$ ) supplied by Honam Petroleum (H 430), PS ( $M_w = 240,000$ ) supplied by Kumho Chemicals (GP 100), PVC ( $DP = 1000$ ) supplied by LG Chemicals (LS 100), CPE (Cl content 36 wt %) supplied by Dow Chemicals (CPE 3615), EPR ( $M.I = 0.89$  (g/10 min) at 190°C,  $M_w = 120,000$ ; ethylene content: 52 mol %) supplied by Korea Petrochemicals, and SEP (styrene/rubber ratio 28/72 by mol %) supplied by Shell Chemical Co. The virgin PVC was used in the form of a powder. To avoid thermal degradation, 3 phr of liquid thermal stabilizers (SONGSTAB TL-700; dioctyltin laurate organic complex, and 3 phr of epoxidized soybean oil plasticizers (SONGSTAB E-700), supplied by Song Won Indust. Co. Ltd., were added to the PVC by dry blending before the blend mixing. Blending was conducted in a Table Kneader internal mixer. The blending time, temperature,

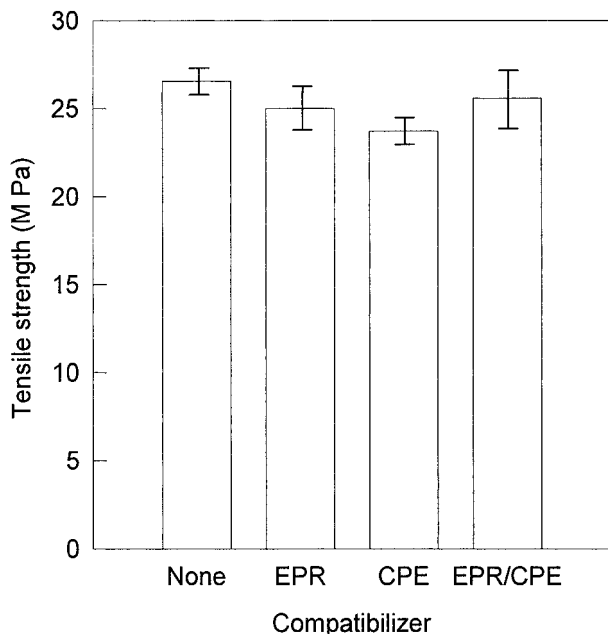
and rotating speed were 8 min, 180°C, and 60 rpm, respectively. The blend composition was fixed at 8/1/1 HDPE/PP (or PS)/PVC by wt %, which is the waste plastics fraction according to the Korea Curbside Tailing Survey data of 1995. The amount of a compatibilizer for ternary blends was fixed at 3 phr based on the total amount of the blend mixture. A mixture of 1/1 CPE and SEP or CPE and EPR by weight ratio was also tested as a compatibilizer. Tensile properties were measured using a tensile tester (Hounsfield H25) at room temperature. The crosshead speed was 10 mm/min. Seven measurements were averaged. For the impact test, notched Izod impact strength was measured with an impact tester (TMI impact testing machine) at room temperature. The specimens for the Izod impact strength measurements had the dimensions of 63 × 12.5 × 3.1 mm with a notch 3 mm in radius. Seven measurements were averaged, and the standard deviation of the measurement was 0.455 J/m. Rheological properties were measured with Rheometrics dynamic analyzer (RDA) at 180°C using parallel plates. Strain was maintained at 10% throughout the experiments. The thermal analysis was performed at a heating rate of 10°C/min using a Perkin–Elmer differential scanning calorimeter (DSC 7) calibrated with pure indium as a standard.

## RESULTS AND DISCUSSION

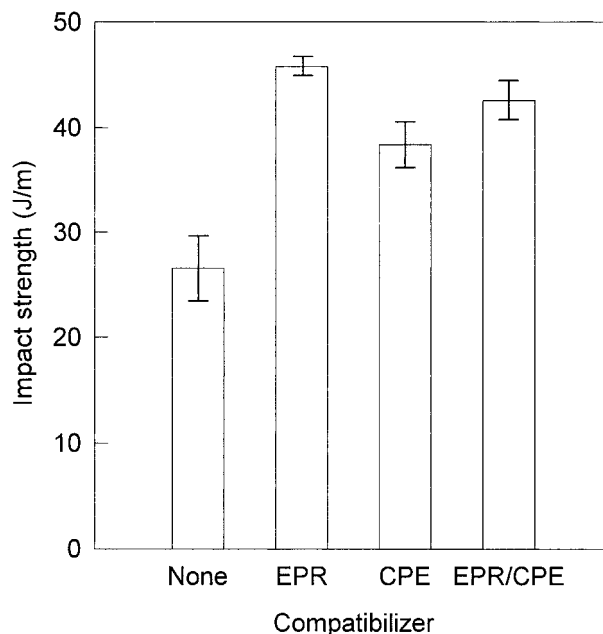
### Properties of HDPE/PP/PVC Ternary Blends

It has been reported<sup>23,24</sup> that the addition of a compatibilizer into a binary blend enhanced such mechanical properties as impact strength, because of the additives locating at the interface between the phases and enhancing the stress transfer, and the particle size reduction–emulsion effect.

Figure 1 shows the tensile strength of the ternary HDPE/PP/PVC blends with or without compatibilizers. By the addition of CPE or EPR, it was expected that interfacial adhesion would be improved, and thus, tensile strength would be increased. This is thought to be attributable to the compatibilization effect of the CPE or EPR at the interface; that is, an increased interfacial adhesion between HDPE and PVC or HDPE and PP, respectively.<sup>21,25</sup> The tensile strength of the ternary blends compatibilized with CPE or EPR or their mixture was, however, not enhanced as compared to the ternary blend without a compatibi-



**Figure 1** Tensile strength of HDPE/PP/PVC (8/1/1) blends with different compatibilizers.

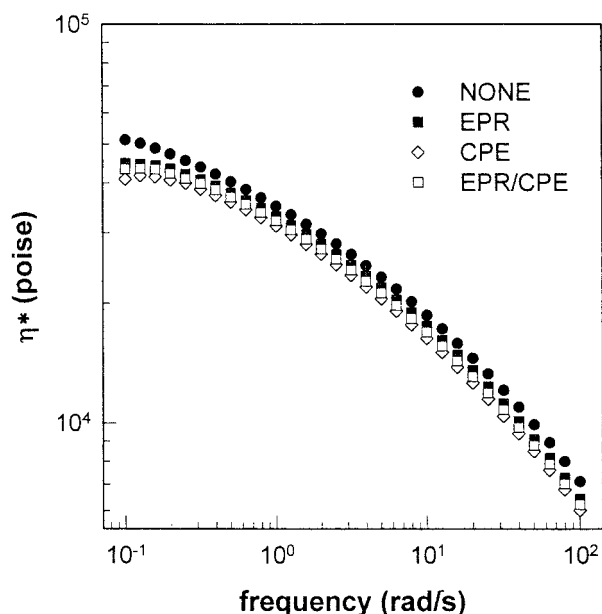


**Figure 2** Impact strength of HDPE/PP/PVC (8/1/1) blends with different compatibilizers.

lizer. The result implies that neither of the compatibilizers for binary blends seems to act as a compatibilizer for ternary blends. Likewise, the mixture of CPE and EPR also did not work as a compatibilizer for HDPE/PP and HDPE/PVC or PP/PVC pairs. The tensile strength of the ternary blends consisting of EPR or EPR/CPE mixture, however, showed slightly higher tensile strength than the ternary blends consisting of CPE only, although the difference is not large. The result may be attributable to the stronger interaction of EPR to HDPE and PP to increase interfacial adhesion between them, rather than that of CPE to HDPE and PVC. On the other hand, it should be noted, as shown in Figure 2, that the impact strength was much improved by addition of either of the compatibilizers, especially EPR. In this case, we can see that CPE, EPR, or their mixture work also as effective impact modifiers for the ternary blend, although either of the compatibilizers is used for impact modification for the binary blends containing any of their parent polymers as a component.

Figure 3 shows the logarithmic plot of the complex viscosity of the HDPE/PP/PVC ternary blends with compatibilizers. All of the complex viscosities of the ternary blends with either of the compatibilizers were lower than that of the ternary blend without a compatibilizer. The complex viscosity of EPR containing ternary blends was

highest among the three ternary blends containing compatibilizers, although the difference is marginal. The result may be related to the fact that the tensile strength of the ternary blends containing EPR showed better tensile strength as



**Figure 3** Complex viscosity of HDPE/PP/PVC (8/1/1) blends with different compatibilizers.

**Table I**  $T_m$  and  $\Delta H_m$  in HDPE/PP/PVC (8/1/1) Blends with Different Compatibilizers

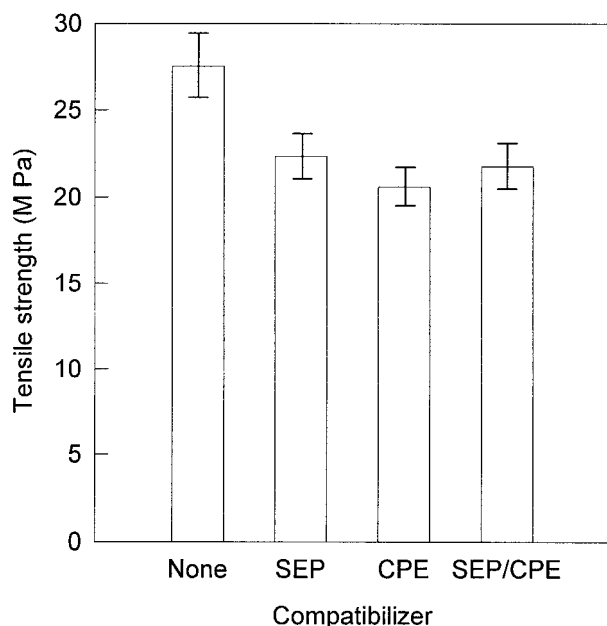
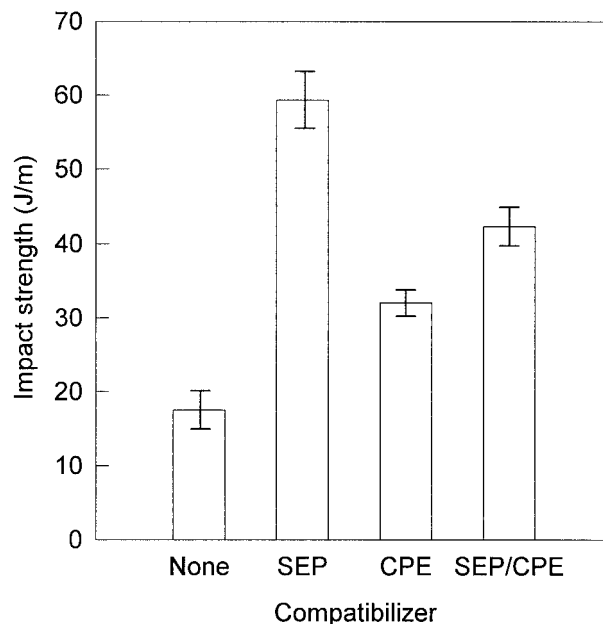
Compatibilizers Used	$T_m$ ( $^{\circ}\text{C}$ )	$\Delta H_m$ (J/g)
None	$132.1 \pm 2.0$	$158.2 \pm 1.5$
EPR	$130.8 \pm 1.5$	$149.7 \pm 1.8$
CPE	$130.8 \pm 2.4$	$149.8 \pm 1.9$
EPR/CPE	$130.9 \pm 1.6$	$150.3 \pm 2.1$

well as impact strength than the CPE containing ternary blends.

The melting point of HDPE in the ternary blend was slightly decreased with adding CPE or EPR or their mixture, as shown in Table I. The heat of fusion of HDPE in blends was also slightly decreased when CPE or EPR or their mixture was added to the blend, meaning that the crystallization of HDPE was retarded by CPE, EPR, or their mixture. Not much difference was observed when the comparison was made based on the kinds of compatibilizers.

#### Mechanical Properties of HDPE/PS/PVC Ternary Blends

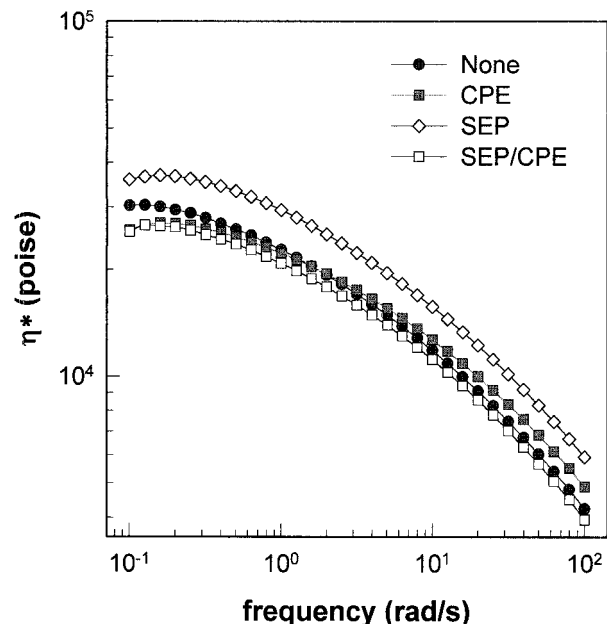
The tensile strength of the HDPE/PS/PVC (8/1/1) ternary blend is presented in Figure 4. The tensile strength of the ternary blends compatibilized with CPE or SEP or their mixture was reduced in

**Figure 4** Tensile strength of HDPE/PS/PVC (8/1/1) blends with different compatibilizers.**Figure 5** Impact strength of HDPE/PS/PVC (8/1/1) blends with different compatibilizers.

comparison to the ternary blend without a compatibilizer. As for HDPE/PP/PVC blends, the result also implies that either of the compatibilizers for binary blends seemed not to act as a compatibilizer for ternary blends. Although CPE and SEP acts as a compatibilizer for HDPE/PVC and HDPE/PS pairs, respectively, the CPE, SEP, or their mixture did not work as good compatibilizers for HDPE/PS and HDPE/PVC or PS/PVC pairs. Not much difference was observed in the change of the tensile strength when SEP and CPE were used as a compatibilizer. Figure 5 shows the impact strength of ternary blends with and without compatibilizers. It should be noted that the impact strength was remarkably increased with the addition of either of the compatibilizers, and the largest increase was observed in the case of the SEP. The result implies that SEP acted as an efficient compatibilizer for HDPE/PS pairs to enhance the impact strength for the ternary blend system more than CPE did for the HDPE/PVC blend system. The increase in the impact strength may be mainly attributable to the enhanced interfacial adhesion between HDPE and PS in the ternary blend by adding SEP.

#### Rheological Properties

A logarithmic plot of the complex viscosity of the HDPE/PS/PVC ternary blends with compatibiliz-



**Figure 6** Complex viscosity of HDPE/PS/PVC (8/1/1) blends with different compatibilizers.

ers is shown in Figure 6. It is seen that the complex viscosities of the ternary blends with a compatibilizer were almost the same as that of the ternary blend without compatibilizers, except for SEP. The complex viscosity of the SEP-containing ternary blends, however, was higher than that of the ternary blend without a compatibilizer. Adding a compatibilizer to this blend increased the interaction of the interface, thereby increasing complex viscosity of the blends.<sup>26</sup> The results in Figure 6 imply that particle interactions were increased by the addition of SEP into the HDPE/PS/PVC blend. The result strongly suggests that the SEP as a compatibilizer for the HDPE/PS blend works well as a compatibilizer for the HDPE/PS/PVC ternary blend, although CPE exhibits a weak compatibilizing effect for the ternary blend at high frequency in the rheological sense.

### Thermal Properties

In general, the addition of a compatibilizer to an immiscible blend causes a melting point depression.<sup>27</sup> In Table II, however, the melting point of HDPE in the ternary blend is not obviously changed, even when adding CPE or SEP. Because the CPE possesses a chemical structure similar to HDPE and has a weak interaction with PVC through its long methylene segments,<sup>28,29</sup> it was

expected that CPE would affect the crystallization behavior of HDPE in blends. Similar  $T_m$  behavior was observed for SEP. The heat of fusion of HDPE in blends was slightly decreased when CPE or SEP was added to the blend. Although the heat of fusion data in Table II are given for the whole blend, it might be assumed that the data of HDPE in blends would be almost identical to those of the whole blend, because the PVC and PS components used in this work were amorphous; moreover, the heat of fusion of CPE used in this work was 0.6 J/g and could be negligible as compared with that of HDPE. The result of Table II indicates that the degree of crystallinity of HDPE was decreased. The retardation of HDPE to crystallize may be explained as a result of the dilution effect of CPE or SEP to reduce the nucleation of crystals.<sup>30</sup> Similar behavior in  $T_m$  and heat of fusion was observed for the 1/1 mixture of CPE and SEP.

### CONCLUSIONS

Recycling of mixed waste plastics in the form of blends is one attractive approach to solving the problems of the municipal solid waste stream, because it avoids the difficult task of separation. High-density polyethylene (HDPE), polypropylene (PP), and poly(vinyl chloride) (PVC) as well as polystyrene (PS) are among the most common plastic wastes, because they are some of the most frequently used commercial plastics in our daily lives as well as in industry.

In this article, we determined the effect of a compatibilizer for binary blends on the properties of ternary blends composed of those common polymers with a simulated waste plastics fraction (8/1/1 w/w). Chlorinated polyethylene (CPE), ethylene-propylene rubber (EPR), and their 1/1 (w/w) mixture were tested as compatibilizers for the HDPE/PP/PVC ternary blend. CPE, styrene-ethylene-propylene block copolymer (SEP), or their

**Table II**  $T_m$  and  $\Delta H_m$  in HDPE/PS/PVC (8/1/1) Blends with Different Compatibilizers

Compatibilizers Used	$T_m$ (°C)	$\Delta H_m$ (J/g)
None	131.5 ± 2.5	158.2 ± 1.5
SEP	131.4 ± 2.3	151.6 ± 2.6
CPE	130.8 ± 1.9	148.9 ± 2.0
SEP/CPE	130.5 ± 2.2	148.2 ± 2.3

1/1 (w/w) mixture were tested as compatibilizers for the HDPE/PS/PVC ternary blend. The amount of the compatibilizer was 3 phr.

For 8/1/1 (by weight) HDPE/PP/PVC ternary blends, the tensile strength was not significantly changed, but the impact strength was significantly increased by adding EPR, CPE or their 1/1 (w/w) mixture. Among the three kinds of compatibilizers, EPR showed much better impact modification for the ternary blend than did the other compatibilizers. The melt viscosity of the ternary blends was decreased when either of CPE, EPR, or their mixture was added. The degree of crystallinity of the HDPE/PP/PVC blend decreased with the addition of any of CPE, EPR, and their mixture. It was found that the SEP and CEP or their 1/1(w/w) mixture as compatibilizers improved the impact strength of 8/1/1 HDPE/PS/PVC blend system; whereas, they did not enhance the tensile strength of the ternary blend. The addition of SEP increased the melt viscosity of the ternary blends, but the other ternary blends containing CPE or CPE/SEP mixture reduced the melt viscosity. The degree of crystallinity of the HDPE/PS/PVC blend was also decreased with the addition of any of CPE, SEP, and their mixture, as for the HDPE/PP/PVC blend when CPE, EPR, or their mixture was added.

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