

# Compatibilization of Regenerated Low Density Polyethylene/Poly(vinyl chloride) Blends

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**ABSTRACT:** The aim of this work is to study the valorization of regenerated low density polyethylene (rLDPE) by blending with PVC in the presence of chlorinated polyethylene (CPE) as compatibilizer. For this purpose, four rLDPE samples coming from neat or dirty wastes were used. They were obtained after milling, washing, and extrusion in a conventional recycling plant. They were first characterized in terms of physicochemical (density, melt flow index, water absorption, and level of oxidation by Fourier transform infrared spectroscopy) and mechanical (tensile and shore D hardness) properties. The effect of the ratio of PVC on these physical and mechanical properties was then investigated.

These binary blends exhibited lower properties than those of the separated polymers. The addition of CPE to the binary blend with weight proportion of 50/50 leads to a substantial improvement of the considered properties which is due to a better interfacial adhesion between rLDPE and PVC as evidenced by the analysis of the morphology of the blends by scanning electron microscopy. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 1750–1755, 2008

**Key words:** PVC; regenerated LDPE; blends; compatibilization; CPE; physicochemical properties; mechanical properties

## INTRODUCTION

Polyethylene (PE) and poly (vinyl chloride) (PVC) are the highest volume thermoplastics consumed in the world, constituting the dominant components of plastics wastes.

As it is well known, PVC/PE blends are immiscible, giving rise to a phase-separated system with a very low degree of adhesion between components.<sup>1</sup> This lack of adhesion causes poor mechanical properties which limits the practical use of these blends. The use of a third polymer to “compatibilize” the blend and to obtain better mechanical properties is a key technology to obtain polymer blends with desirable properties. Many studies have been done on the compatibilization of this pair of polymers.<sup>2–5</sup> It was found that small amounts of chlorinated polyethylene (CPE) increases both ductility and tensile strength of PVC/PE blends.<sup>2</sup> Different rheological techniques were used to elucidate the role played by block and random CPEs in the rheology of practical devoted HDPE/PVC/CPE systems.<sup>5</sup> The morphology and mechanical properties of PVC/PE blends were also modified by adding butadiene rubber (BR) into the blend. This was attributed to the fact that BR can increase the melt viscosity of the PE-rich phase, and hence, decrease the viscosity ratio

between the PVC-rich phase and PE-rich phase.<sup>3</sup> On the other hand, a compatibilization-crosslinking synergism technique was proposed to improve the mechanical properties of PVC and PE.<sup>4</sup> A compatibilizer can promote the phase dispersion of PVC and PE and their interfacial adhesion. Then, the probability of a crosslinking agent existing at the interface will increase and more cocrosslinked products will be formed. Therefore, compatibilization and crosslinking are both exerted sufficiently. It was found that BR or styrene-butadiene rubber (SBR) and the crosslinking agent dicumyl peroxide (DCP) have a good synergism in the improvement of the mechanical properties of incompatible PVC/LDPE blends.<sup>4</sup>

The objective of this work is to recycle regenerated LDPE in the form of blends with PVC in the presence of CPE as compatibilizer. The latter is a commonly used impact modifier of PVC and its compatibility depends on the chlorine content and the distribution of the chlorine atoms on the PE backbone.<sup>6</sup> CPE with 36% chlorine is the optimum composition for obtaining the necessary features of impact, processing and strength<sup>3,7–9</sup> and it is used in this work as the third component of rLDPE/PVC blends.

## EXPERIMENTAL

### Materials

Four regenerated LDPE coming from different sources (Table I) and obtained after milling, washing,

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TABLE I  
Description of the Samples of LDPE Used

Designation of rLDPE	Origin of the LDPE scraps	Nature of the scraps	Final aspect of the granules
R1	Stabilized agricultural films	Neat	Green
R2	Milk pouches	Dirty	Grey
R3	Bags and unstabilized agricultural films	Dirty	Yellow
R4	Carbon black stabilized agricultural films	Dirty	Black

and extrusion in a conventional recycling plant were used. Virgin LDPE (B24/2) and PVC (VICIR S 1200) were commercial products from ENIP (Skikda, Algeria) and CIRES (Estarreja, Portugal). CPE with 36% chlorine was supplied by Aldrich (Taufkirchen, Germany). The PVC additives were dioctyl phthalate (DOP) from SGP (Rades, Tunisia) as plasticizer, Interstable from AKZO (Milan, Italy) as lead based thermal stabilizer, and stearic acid from SOGIS-SPZ (Milan, Italy) as lubricant.

### Samples preparation

- rLPDE/PVC blends: Blends of variable composition from 0 to 100 wt % were prepared in the presence of 3 wt % of heat stabilizer, 1 wt % of lubricant and 30 wt % of plasticizer. The amounts of the three additives were added according to the amount of PVC in the blend. Melt mixing was performed at 125°C on a two-roll mill for 20 min. The blends were then pressed in a hydraulic press at 170°C and 300 KN for 5 min. The specimens were cut from the obtained plates for the mechanical characterization.
- rLDPE/PVC/CPE blends: rLDPE/PVC with a weight proportion of 50/50 was used as the basic blend in this study. It was chosen because it exhibited the lowest tensile properties. The amount of CPE for ternary blends was 5, 10, 15, and 20 wt % based on the total amount of the ternary blend. The amounts of the three PVC additives were as mentioned in the previous section. Concerning the pure materials, they were treated in the same way as the blends to have the same themomechanical history.

### Samples characterization

The density and water absorption were measured, respectively, according to the ISO/R 1183-1970 (F) and ISO 62-1980. The melt flow index (MFI) was measured according to the ISO 1133-1981 (F).

The level of oxidation ( $R_{ox}$ ) was evaluated by FTIR analysis using a SHIMADZU FTIR 8300 apparatus according to the following relation:

$R_{ox} = A_{C=O}/A_{CH_2}$ , where  $A_{C=O}$  and  $A_{CH_2}$  correspond to the absorbances of the carbonyl band at 1720  $cm^{-1}$  and the methylene band at 729  $cm^{-1}$ , respectively.<sup>10</sup>

The tensile properties and shore D hardness were measured according to the ISO 37 F and NFT51-109 (1981). Tensile test was performed on an Adamel Lhomargy tensile instrument at a crosshead speed of 250 mm/min. The results of the tensile test and shore D hardness were obtained by averaging the results of five measurements.

The morphology of the blends was characterized using a Philips XP. 20 Scanning Electron Microscope. The surfaces were metallized with a thin layer of carbon to increase the surface conductivity of the material and thus its sensitivity to this test.

## RESULTS AND DISCUSSION

### Preliminary characterization of virgin LDPE, rLDPE, and plasticized PVC

The physicochemical properties (density, water absorption, MFI, and  $R_{ox}$ ) of virgin LDPE, rLDPE, and plasticized PVC are given in Table II. The densities of the rLDPE are comparable with that of virgin LDPE and to literature.<sup>11</sup> The plasticized PVC density is also conform to literature.<sup>11</sup> Polyolefins are not totally waterproof<sup>12</sup> and the results confirm it. However, as expected, water absorption for all LDPE samples was lower than that of PVC.<sup>12</sup>

Melt flow index increases directly with fluidity and inversely with viscosity. An increased value of MFI means a reduced molecular weight ( $M_w$ ). According to results of Table II, all rLDPE samples presented lower values of MFI in comparison with virgin LDPE which means that their  $M_w$  are higher than that of virgin LDPE. This can be explained by the fact that the initial LDPE scraps were probably subjected to thermo and photooxidative degradation during the fabrication process or the use which leads to some crosslinking. The occurrence of oxidation reactions is confirmed by FTIR analysis ( $R_{ox}$ ). Hence, all rLDPE samples showed a certain level of degradation.

Table III shows the mechanical properties of virgin LDPE, rLDPE and plasticized PVC. Tensile proper-

**TABLE II**  
Physicochemical Properties of Virgin LDPE, rLDPE, and PVC

Sample	Density (g/cm <sup>3</sup> )	Water absorption at 23°C (%)	Melt flow index (g/10 min)	R <sub>ox</sub>
Virgin LDPE	0.902	0.06	0.70 <sup>a</sup>	0.00
R1	0.900	0.04	0.30 <sup>b</sup>	0.60
R2	0.901	0.05	0.44 <sup>a</sup>	0.35
R3	0.905	0.07	0.40 <sup>b</sup>	0.40
R4	0.893	0.06	0.50 <sup>a</sup>	0.70
PVC	1.270	0.13	—	—

<sup>a</sup> Nominal load  $M = 2164$  kg f.

<sup>b</sup> Nominal load  $M = 5$  kg f.

ties (strain and stress at break) show clearly that degradation reactions affected the behavior of rLDPE samples which failed at lower elongations and higher stresses (except for R4) than virgin LDPE. Furthermore, the shore D hardness values of all rLDPE samples are higher than that of virgin LDPE. This fact is directly related to their level of reticulation which affects their rigidity and therefore their hardness. Sample R4 exhibited the most noticeable mechanical properties which is probably because of its higher level of degradation since it was obtained from dirty agricultural films which were submitted to photooxidative weathering during their initial use. This fact is in accordance with FTIR analysis (Table II) which shows that R4 has the higher level of oxidation ( $R_{ox} = 0.70$ ) in comparison with the other rLDPE samples.

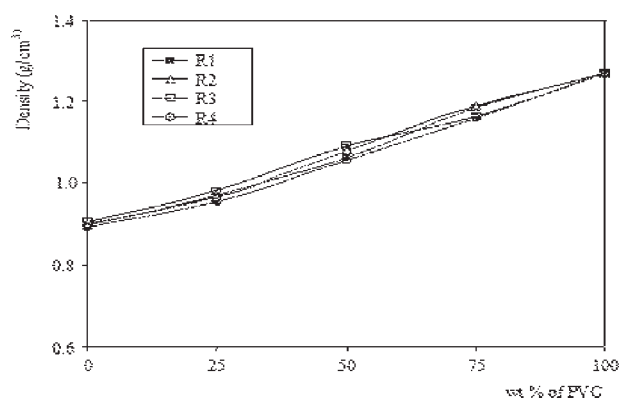
### Characterization of rLDPE/PVC blends

Figure 1 shows the variation of density as a function of the amount of PVC in the blends. Since the density of plasticized PVC is higher than that of LDPE, it is obvious that the addition of plasticized PVC increases the density of rLDPE/PVC blends. The variation of density with blend composition is quasi-linear. Although water absorption of PVC is relatively higher than that of LDPE, the variation of water absorption of rLDPE/PVC blends with blend composition (Fig. 2) does not follow the same behavior than density. It can be seen that water absorption

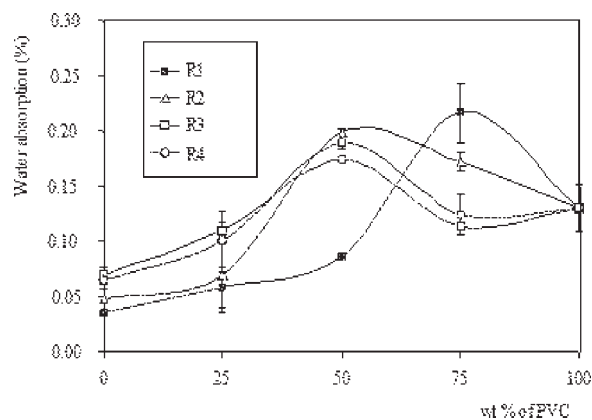
increases with the amount of PVC until a certain threshold [50 wt % of PVC for rLDPE samples coming from dirty scraps (R2, R3, and R4) and 75 wt % of PVC for rLDPE coming from neat scraps (R1)] and then decreases. It seems that the binary blends undergoes a phenomenon of water absorption because of the immiscibility of the two polymers which favors the presence of microcavities and facilitates the water penetration until their saturation. On the other hand, for a same composition, the lowest values of water absorption are obtained with R1 up to about 50 wt % of PVC. This fact shows the influence of the nature of the initial scraps. Indeed, regenerated LDPE coming from dirty scraps may contain residual contaminants which affect the adhesion between the macromolecular chains and result in the presence of a highest free volume (volume existing between macromolecules) compared with regenerated LDPE coming from neat scraps. Then, the size of the microcavities which are present in the case of R2, R3, and R4 is relatively more important due to the combined effect of the immiscibility of the two polymers and the lack of adhesion related to residual contamination and then results in higher values of water absorption in comparison with R1. This water penetration leads to the migration of the PVC additives (plasticizer, heat stabilizer, lubricant) outside the polymer matrix and then explains the

**TABLE III**  
Mechanical Properties of Virgin LDPE, rLDPE, and Plasticized PVC

Sample	Strain at break (%)	Stress at break (MPa)	Shore D hardness
Virgin LDPE	900.00	10.690	52.0
R1	800.00	14.037	53.0
R2	800.00	12.714	53.0
R3	750.00	12.762	53.0
R4	450.00	9.098	54.0
PVC	300.00	23.174	61.0



**Figure 1** Variation of density with blend composition.



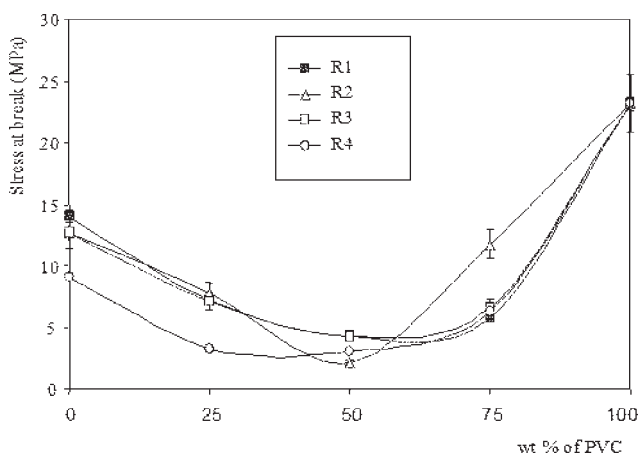
**Figure 2** Variation of water absorption with blend composition.

observed decrease in water absorption.<sup>13–15</sup> In the case of R1, water absorption decrease started at about 75 wt % of PVC while it has already attained minimal values for R2, R3, and R4. This fact can be explained by the contribution of water absorption of PVC which is three times higher than that of R1 (Table II).

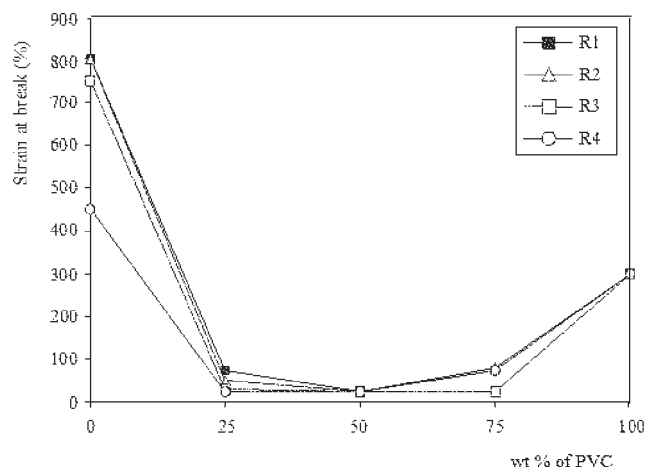
Figures 3 and 4 show, respectively, the variation of stress and strain at break with blend composition. It can be seen that all the blends failed at lower values than that of rLDPE and plasticized PVC alone. These results confirm that the binary blends of rLDPE and plasticized PVC are typically immiscible with a poor phase structure and mechanical properties.<sup>1</sup> Therefore, the incorporation of a third component is necessary to enhance the compatibility of this pair of polymers.

#### Characterization of rLDPE/PVC/CPE blends

The density of rLDPE/PVC/CPE blends versus CPE content is plotted in Figure 5. All the densities of the ternary blends are lower than that of the correspond-



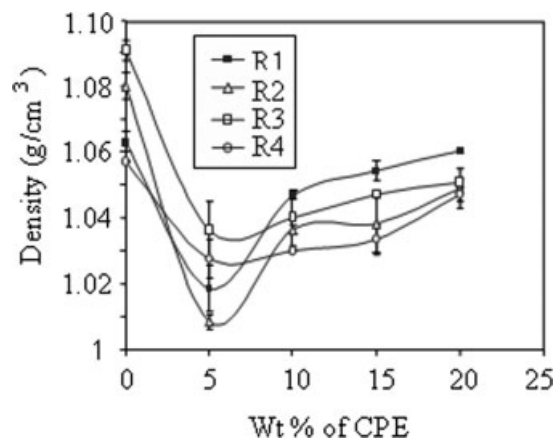
**Figure 3** Variation of stress at break with blend composition.



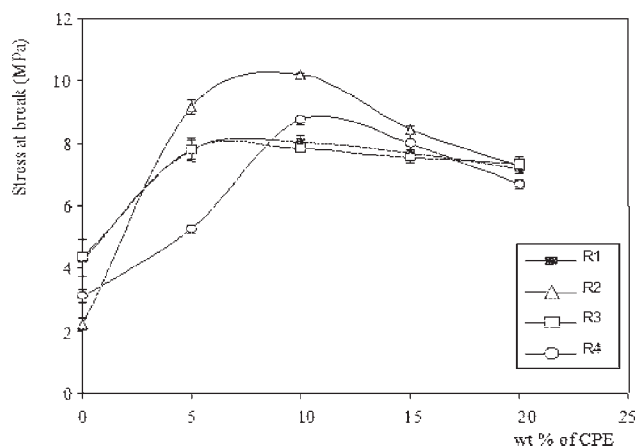
**Figure 4** Variation of strain at break with blend composition.

ing binary blends. Furthermore, the density decreases for a level of CPE of about 5 wt % and then increases slowly and seems to reach a plateau. These observations are related to the presence of the compatibilizer which increases the interfacial adhesion between the two incompatible polymers and then reduces the microcavities in the blend.

Figures 6 and 7 show, respectively, the variation of stress and strain at break with CPE content. The effect of the addition of CPE on the tensile properties of 50/50 (wt) rLDPE/PVC blends is very noticeable in our case. It can be seen that all the ternary blends underwent broke at higher stresses and elongations than the binary blends. Both stress and strain at break increase with increasing CPE content and reach a maximum value for about 8–15 wt % of CPE. These results can be explained by the fact that CPE addition increases adhesion via interactions with the blend components which enhances tensile properties. However, it seems that the increase of CPE level reduces the adhesion with rLDPE samples



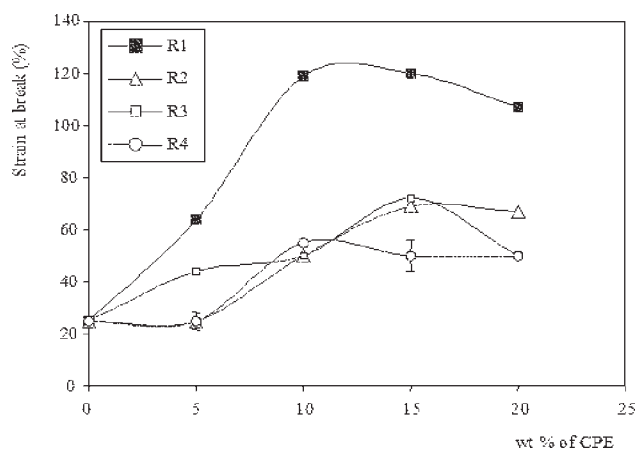
**Figure 5** Variation of density with the level of CPE.



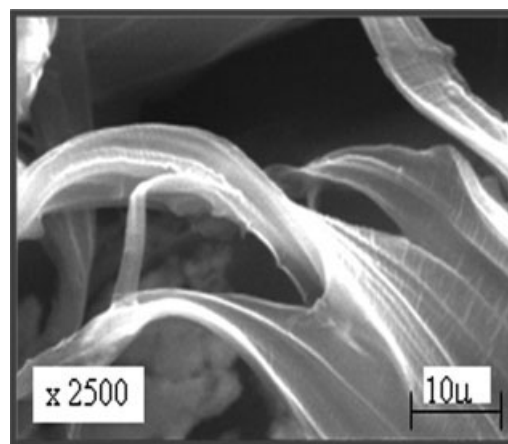
**Figure 6** Variation of stress at break with the level of CPE.

which explains the observed decrease in tensile properties after the maximum reached. Furthermore, the effect of the residual contamination is clearly shown in Figure 7. The better strains at break are obtained with R1 which is coming from neat scraps. This tensile property increased from 25 to 120% for R1 while it increased more lightly for the others (from 25 to 69% for R2, 25 to 72% for R3, and 25 to 55% for R4). These results are related to residual contamination which affects the adhesion between the blends components and leads to lower strains at break.

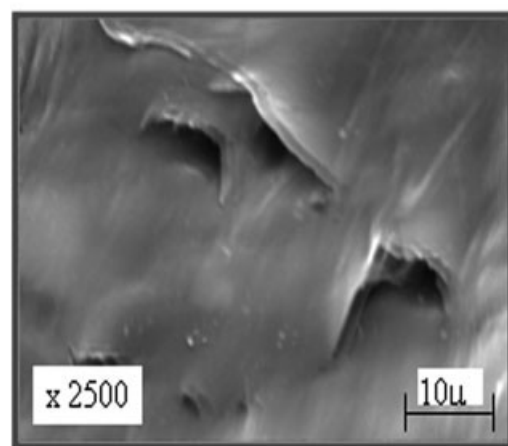
Scanning electron micrographs of R1/PVC (50/50) and R1/PVC/CPE (45/45/10) are shown in Figure 8. They reveal a morphological change when CPE is added to the binary blend. Indeed, as can be seen in Figure 8(a), in the case of nonmodified R1/PVC blend, the presence of micro cavities is clearly shown. This means that the association ability of PVC and rLDPE particles is weak which leads to poor mechanical properties. However, when CPE is incorporated [Fig. 8(b)] a much better dispersion is



**Figure 7** Variation of strain at break with the level of CPE.



a- R1 /PVC (50/ 50)



b- R1 /PVC /CPE (45 /45/ 10)

**Figure 8** Scanning electron micrographs of binary and ternary blends (Grx 2500).

obtained with a better cohesion and a decrease of the micro cavities size which explain the improvement of the mechanical properties.

## CONCLUSIONS

The preliminary characterization of the rLDPE samples showed that their physicochemical (density, water absorption, MFI, Rox) and mechanical properties (tensile and shore D hardness) depend on the level of degradation of the corresponding wastes.

The characterization of rLDPE/PVC blends showed that this pair of polymers is immiscible with a poor phase structure and tensile properties.

The experimental data showed that the addition of CPE to binary polymer blends resulted in a significant increase of tensile properties (stress and strain at break) which is due to a better interfacial adhesion as evidenced by scanning electron microscope. Then, from the point of view of practical applications, the

addition of CPE as a compatibilizer is an effective way of recycling LDPE wastes in presence of PVC.

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