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PREPARATION, CHARACTERIZATION AND VALORIZATION OF REGENERATED LOW DENSITY POLYETHYLENE/POLYPROPYLENE BLENDS

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Abstract

The aim of this work is to study the valorisation of two regenerated low density polyethylene (LDPE) coming from dirty wastes by blending with polypropylene (PP) in presence of compatibilizer. The first step was the determination of the physico-chemical properties of the two regenerated samples (density, melt flow index, water absorption, melting temperature and oxidation index) and the mechanical properties (tensile and shore D hardness).
The second step was the characterization of the physical (density, water absorption) and mechanical properties (tensile and shore D hardness) of LDPE / PP blends. In the third and last step, the effect of compatibilizer on the properties of LDPE / PP blends in the same proportion was considered. Physical characterization (density, water absorption and morphology analysis by scanning electron microscopy) and mechanical properties were carried out.
The results showed that this kind of blending has contributed considerably in performing properties of regenerated LDPE.

Key words: compatibilization, LDPE/PP blends, recycling, valorization

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1. Introduction

The mechanical recycling of post - consumer plastics remains one of the preferred recycling options for ecology and energy reasons, as far as this way is economically profitable.

Low density polyethylene (LDPE) is a widely used commodity thermoplastic and it represents an important weight percentage of the polyolefins found in waste stream. Alone, LDPE is not really interesting for particular applications because its mechanical properties are influenced by the aging of the product (Bertin et al., 2002). Blending of different plastic resins has long been practiced to tailor blends for specific processing and performance requirements.

Blends of PE and polypropylene (PP) are among the binary systems that have attracted much attention (Munaro and Akcerlrud, 2008; Shurman et al., 1998; Tai et al., 2000; Yang et al., 2003; Strapasson et al., 2005; Lai et al., 2005). However, preparation of LDPE/PP blends is hindered by the low compatibility of this polymer pair (Ali et al., 2005; Radonjic and Gubeljak, 2002).

In the present work two regenerated LDPE coming from dirty wastes were used. Their physico-chemical and mechanical properties were first evaluated.

The preparation and characterization of rLDPE / PP blends was carried out. As expected, the binary blends exhibited poor mechanical properties. Finally, the effect of a compatibilizer on the properties of rLDPE / PP in the same proportion was considered. Physical, mechanical and morphological characterizations were carried out..

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2. Experimental

2.1. Materials

Two regenerated LDPE (rLDPE) coming from dirty wastes (Table 1) and obtained after milling, washing and extrusion in a conventional recycling plant were used. Virgin LDPE (B24 / 2) and PP (Lacqrene 7240) were commercial products from ENIP (Algeria) and Atofina (France). Maleic anhydride functionalized elastomeric ethylene copolymer (Exxelor VA 1801), a commercial product from Exxon Mobil (France) was used as compatibilizer.

Table 1. 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	Carbon black stabilized agricultural films	Dirty	Black
R2	Milk pouches	Dirty	Grey

2.2. Samples preparation

- **rLDPE/PP blends**: blends of variable composition from 0 to 100 wt % PP were prepared by injection moulding.
- **rLDPE/PP/ Compatibilizer blends**: rLDPE / PP with a weight proportion of 50/ 50 was used as the basic blend in this work. The amount of compatibilizer was 5, 10, 15 and 20 wt %. Ternary blends and pure materials were treated in the same way as the binary blends in order to have the same thermo mechanical history.

2.3. Samples characterization

The density and water absorption were measured respectively, according to the standards T 51-561 1990 (F) and ISO 62- 1980. Melting temperature (Tm) was measured by using a Dupont DSC apparatus at a heating rate of 10 °C/min.

The melt flow index (MFI) was measured according to the ISO 1133- 1999 using a plastometer CEAST 6542/000. The level of oxidation (R_{ox}) was evaluated by FTIR analysis using a FTIR 830 Matsini Matson apparatus according to Eq. (1).

$$R_{ox}= A_{c=o} / A_{CH2} \quad (1)$$

where: $A_{c=o}$ and A_{CH2} correspond to the absorbances of the carbonyl band at 1720 cm^{-1} and the methylene band at 729 cm^{-1}, respectively (Irinislimane et al., 2007; Kabdi et Belhaneche- Bensemra, 2009).

The tensile properties and shore D hardness were measured according to the ISO 527- 1993 and ISO 868- 1978 (F), respectively. Tensile test was performed on a Zwick/ Roell tensile instrument at a crosshead speed of 250 mm/ mn. The results of the tensile test and shore D hardness were obtained by averaging the results of five measurements. The morphlogy of the metallised samples was investigated by using a Jeol apparatus.

3. Results and discussion

3.1. Preliminary characterization of virgin LDPE, rLDPE and PP

The physico- chemical properties (density, water absorption MFI, Tm and R_{ox}) of virgin LDPE, rLDPE and PP are given in Table 2. The densities of R_1 and R_2 are comparable to that of virgin LDPE and to literature (Irinislimane et al., 2007; Kabdi and Belhaneche- Bensemra, 2009). The density of PP is also comparable to literature (Lai, 2005). As expected, the results show that all the studied samples are not totally waterproof (Kabdi and Belhaneche- Bensemra, 2009). R1 and R2 showed a relatively higher value of water absorption tan LDPE. This fact may be due to the existence of a free volume existing between macromolecule which favours water absorption.

It is known that melt flow index increases directly with fluidity and inversely with viscosity. An increased value of MFI means a reduced molecular weight (Mw). According to the results of Table 2, regenerated samples presented higher values of MFI in comparison with virgin LDPE. These results can be explained by the fact that the initial LDPE scraps were probably subjected to thermo and photo- oxidative degradation during the fabrication process or the initial use which leads to some crosslinking. Furthermore, it can be suggested that R2 is the more crosslinked. FTIR analysis (R_{ox}) confirmed the occurrence of oxidation reactions (Table 2).

It seems that R1 is the most oxidized as it comes from agricultural films that were subjected to photo and thermo-oxidative degradation during their initial use. Mechanical properties (Table 3) showed clearly that these degradation reactions affected the behaviour of regenerated samples which failed at lower stresses and higher strains than virgin LDPE. It is to be noted that R1 exhibited lower strain at break due to the highest level of oxidation which means that chains scissions occurred during the initial use as agricultural films.

On the other hand, the shore D hardness values of the two regenerated samples were slightly higher than that of virgin LDPE. However, R1 exhibited a higher shore D hardness value which confirms its higher rigidity due to its higher level of reticulation as it comes from agricultural films.

3.2. Characterization of rLDPE/ PPblends

The density decreased with the increase of PP content (Fig. 1a). This is obvious since PP has less density as compared to LDPE. Water absorption (Fig. 1b) increased with the increase of PP content in the blends. This is due to the immiscibility of this pair of polymers which favours the presence of cavities in the blends and facilitates the wáter penetration. This result was already observed (Kabdi et Belhaneche-Bensemra, 2009).

The variation of stress at break with blends composition is given in Fig. 2a. It can be noted that stress at break increased in the case of the two rLDPE samples used. The strain at break (Fig. 2b) decreased with blend composition. It reached a minimal value up to about 20 wt % of PP and remained constant for higher compositions.

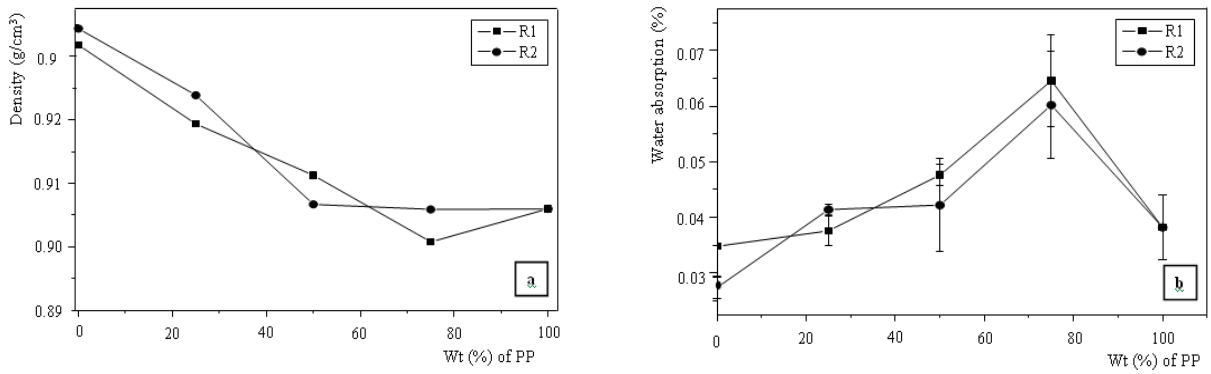
These poor mechanical performances are related to the immiscibility of the two polymers. Therefore, the incorporation of a third component is necessary to enhance the compatibility of this pair of polymers. In order to investigate the effect of the compatibilizer, the rLDPE/ PP (50/50) which exhibited the lowest elongations at break was chosen as the basic blend in the third step of this work.

Table 2. Physico- chemical properties of LDPE, rLDPE and PP

Samples	*Density (g/cm^3)*	*Water Absorption (%)*	*MFI (g/10min)*	*Tm(°C)*	*Rox*
LDPE	0.928	0.034	0.262	118.73	0.00
R1	0.932	0.048	0.420	115.90	0.40
R2	0.934	0.042	0.334	125.83	0.35
PP	0.906	0.039	/	/	/

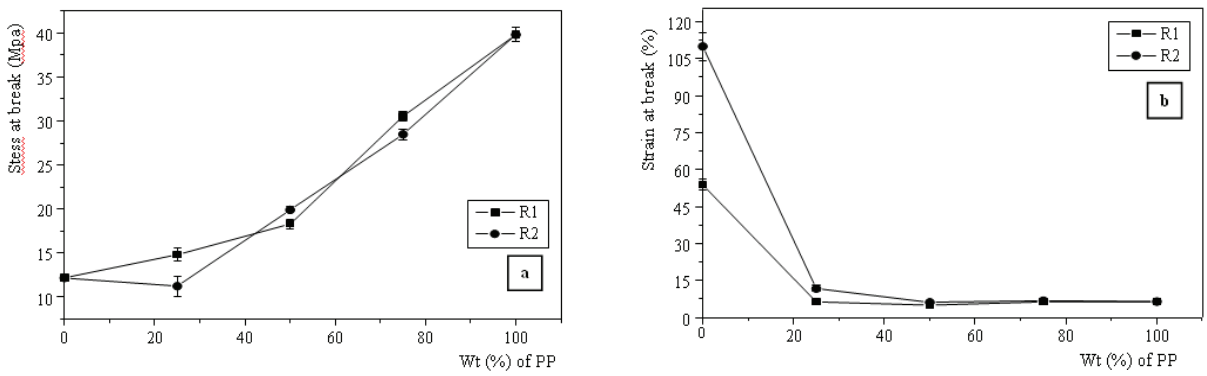
Table 3. Mechanical properties of LDPE, rLDPE and PP

Sample	*Strain at break (%)*	*Stress at break (MPa)*	*Young modulus (MPa)*	*Shore D hardness*
LDPE	54.61	13.74	225.24	51.2
R1	56.31	12.14	172.76	52.4
R2	105.74	11.98	250.38	51.4
PP	6.5	39.78	607.19	70.0



(a) (b)

Fig. 1. Variation of physical properties with blend composition: (a). Density; (b).Water absorption; R1: Carbon black stabilized agricultural films; R2: Milk pouches



(a) (b)

Fig. 2. Variation of tensile properties with blend composition: (a) Stress at break; (b) Strain at break; R1: Carbon black stabilized agricultural films; R2: Milk pouches

3.3. Characterisation of rLDPE / PP/ compatibilizer blends

Fig. 3a shows a decrease of density for about 5 wt % of compatiblizer wich denotes a certain amelioration of interfacial adhesion between the two polymers. Fig. 3 b shows also a decrease of the water absorption for the same level of compatibilizer.

These results can be explained by the fact that the compatibilizer increased the interfacial adhesion between the two incompatible polymers which reduced micro cavities in the ternary blends. Furthermore, the presence of micro- cavities facilitates the water penetration until their saturation. This water penetration leads to the migration of the present additives like antoxidants outside the polymer matrix and then explains the observed negative values of water absorption (Atek et al., 2005; Belhaneche-Bensemra et al., 2002; Boussoum et al., 2006).

The variations of stress and strain at break with the level of compatibilizer are given in Figs 4. Fig. 4a shows clearly that stress at break decreased with the level of compatibilizer while Fig. 4b shows that the strain at break increased. These results indicate the amelioration of mechanical properties by incorporation of the compatibilizer. Fig. 5 shows the morphology of the realized blends with R1.

In the case of the blend R1/PP, the presence of microcavities indicates the weak association capacity between the particles of rLDPE and PP and explains the weak performances obtained in tensile properties.

A change in the morphology is obtained when the compatibilizer is added. A decrease in the microcavities size and a better cohesion between the blends components is obtained. These effects are more pronounced with the level of compatibilizer.

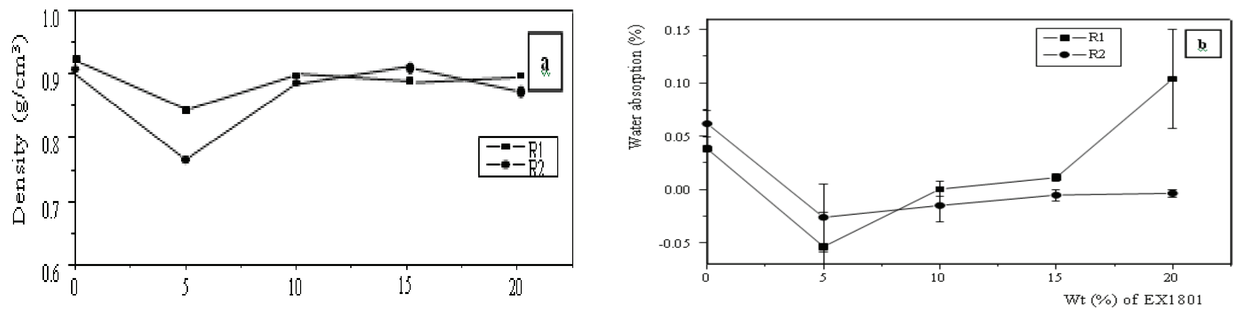
Then, the amelioration of tensile properties is due to a better interfacial adhesion between these two incompatible polymers.

4. Conclusions

From the above results, the following conclusions can be drawn.

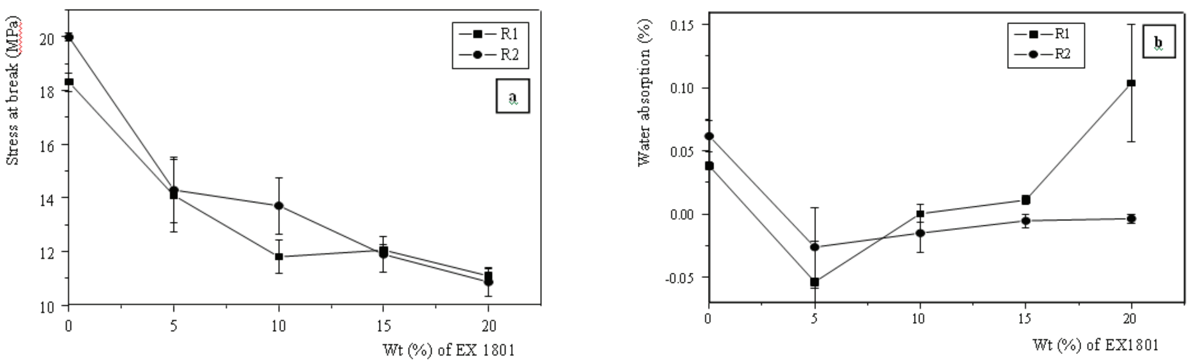
The preliminary characterization of the rLDPE samples showed that their physico- chemical and mechanical properties depend on the level of degradation of the corresponding wastes. The characterization of rLDPE / PP blends showed that this pair of polymers has poor tensile properties due to their incompatibility.

A relative amelioration of the tensile properties is obtained when maleic anhydride functionalised elastomeric ethylene copolymer is added to the blends. From the point of view of practical applications, the addition of the compatibilizer is an effective way of recycling LDPE wastes in presence of PP. Finally, the results showed that this kind of blending has considerably contributed in performing properties of rLDPE.



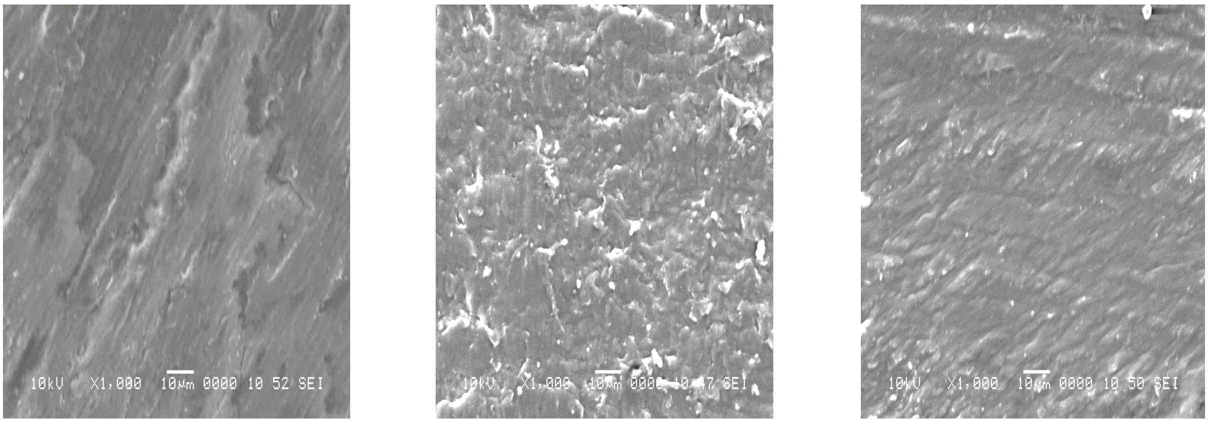
(a) (b)

Fig. 3. Variation of physical properties with the level of compatibilizer. (a) Density; (b) Water absorption
R1: Carbon black stabilized agricultural films; R2: Milk pouches



(a) (b)

Fig. 4. Variation of tensile properties with the level of compatibilizer. (a) Stress at break; (b) Strain at break;
R1: Carbon black stabilized agricultural films; R2: Milk pouches



a. R1/PP (50/50)

b. R1/PP/EX1801 (50/50/5)

c. R1/PP/EX1801 (50/50/15)

Fig. 5. Analysis of R1/PP with and without compatibilizer by scanning electron microscopy (Gr ×1000).
R1: Carbon black stabilized agricultural films

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