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Polyethylene Blends: Better Formulations for Recycled Polyethylene

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Summary: To evaluate the performance of recycled high density polyethylene for pipe applications, blends containing 10, 30 and 50% of recycled in a PE T100N matrix were prepared and characterized using several instrumental analysis methods. The thermal analysis was performed by differential scanning calorimetry and thermogravimetry in the temperature ranges [23 °C, 300 °C] and [25 °C, 550 °C], respectively. Other technics as DMA, FTIR, XRD and MFR were used. The experimental results have showed that incorporation of recycled PE in PE T100N leads to poorer properties and it requires an upgrading with suitable additives.

Keywords: blends; pipes; polyethylene; properties; recyclates

Introduction

The reuse of recycled polymers in engineering applications, as plastic pipes for drainage and sewerage, is desirable due to the environmental and economic concerns. However, they need to be characterised[1] and blended with virgin materials and additives to assure the maintenance of requested properties for such applications.

The performance of the blends depends on the properties of components, their content and morphology.[2] Preparation of individual ratios of blended polymers requires many combinations and each has to be individually characterized.[3] The final properties must be evaluated, aiming to assess their suitability for specific applications.

In this work a set of blends containing recycled PE and virgin PE T100N was compounded by extrusion. Subsequently they were analysed by thermal, mechanical and FTIR spectroscopy methods. Mass flow rate was also measured.

Experimental Part

Materials

The HDPE used in this research study was supplied by Repsol (T100N). The PE T100N has a density near of 0.96 and a melt flow rate of 0.4 g/10min (190 °C/5 kg). Two PE recyclates, RPE1 and RPE3, were supplied by AMBIENTE industries with densities 0.97 and 0.96, respectively. All the PE materials were in pellets and contain carbon black. The PE blends were prepared with 10, 30 and 50% of the two recyclates in a HDPE polymer matrix in a Prism Eurolab twin screw extruder at temperature of 190–210 °C and at speed of 300 rpm. The raw materials and the compounds were conditioned during 24 h at 23 °C and 50% of relative humidity before testing.

Instrumental Methods of Analysis

The thermal analysis was performed by differential scanning calorimetry (DSC), thermogravimetry (TGA) and dynamical mechanical analysis (DMA), according to the international standards ISO 11357, ISO 11358 and ISO 6721.[4–6] Other instrumental method of analysis used was infrared spectroscopy (FTIR). Melt flow rate measurements (MFR) were also performed according international standard EN ISO 1133.[7]

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The differential scanning calorimeter apparatus used was a DSC NETZCH 200 F3 Maia. The transition temperatures and respective enthalpies were determined using a scanning rate of 10 °C/min, in nitrogen atmosphere. Oxidation induction time and oxidation induction temperature were determined in oxygen atmosphere, at 200 °C and in the range [23 °C, 300 °C], respectively. In all tests it was adopted a gas flow of 50 ± 5 mL/min.

The TGA was performed in a SETARAM TG 92-1750 with alumina crucibles, between 25 and 550 °C, in argon atmosphere with a flow of of 50 ± 5 mL/min at a heating rate of 10 °C/min. Dynamical mechanical analysis was performed in a DMA TA Instruments Q800, using a 3-point bending clamp, for a frequency of 1 Hz and an amplitude of 15 μm, at a rate of 3 °C/min in the temperature range [−150 °C, 90 °C]. For FTIR analysis it was used a Thermoscientific Nicolet Magna-IR 550 Series II in the range [4000–400 cm^{-1}] with a spectral resolution of 4 cm^{-1}. The films used for analysis were prepared by compression moulding at 170 °C until uniform thickness. The MFR was obtained at 190 °C/5 kg condition, using an Extrusion Plastometer DTS.[7]

Results and Discussion

The results of the blend characterization containing HDPE T100N and different contents of two recyclates are presented in the Table 1. The information obtained helps to understand the influence of recycled PE on the properties of blends.

The thermal oxidation resistance of polyolefin is mainly illustrated by the oxidation induction parameters (OIT). The incorporation of recycled PE in the blend induces faster degradation and a decrease in the normal HDPE properties, not satisfying the requisites of good performance for typical applications selected. Other properties, like transition temperatures (melting (Tm) and decomposition temperatures (DT) and initial decomposition temperatures (IDT) give, also, indications of previous degradation. The value of isothermal OIT, measured at 200 °C, for the recycled samples RPEn (0 and 1 min) is an indicator of the low thermal resistance when compared with the excellent thermal stabilization of the PE T100N (OIT = 78 min). The incorporation of 10% recycled in PE T100N decreases the isothermal OIT values in 60–70%, depending on the type of recycled PE used and its homogeneity. The presence of only 30% of recyclate induces 90% of loss in thermal stability. The blend of 50% showed a decrease of 28 °C in the induction oxidation temperature when compared with the correspondent value of virgin PE T100N. These results indicate that the addition of an antioxidant is necessary to improve the thermoxidative resistance of the blends. The initial decomposition temperature decreases with the

Table 1.
Properties of polyethylene blends.

Material	Content in RPE, %	OIT (200°C), min	OIT, °C	IDT, °C	DT, °C	Tg, °C	Tm, °C	Tn, °C	MFR (190/5), g/10min
PE T100	0	78	248	409	477	−116	137	118	0.4
RPE1	100	1	205	397	479	−128	131	119	2.0
	50	4	221	399	478	n.m.	131	121	0.7
	30	10	227	n.m.	n.m.	−125	135	120	0.5
	10	22	235	n.m.	n.m.	−120	134	120	0.4
RPE3	100	0	197	406	480	n.m.	137	119	2.6
	50	4	226	n.m.	470	n.m.	138	119	0.6
	30	9	225	n.m.	n.m.	n.m.	136	120	0.5
	10	31	236	347	n.m.	n.m.	136	119	0.4

n.m. = not measured.

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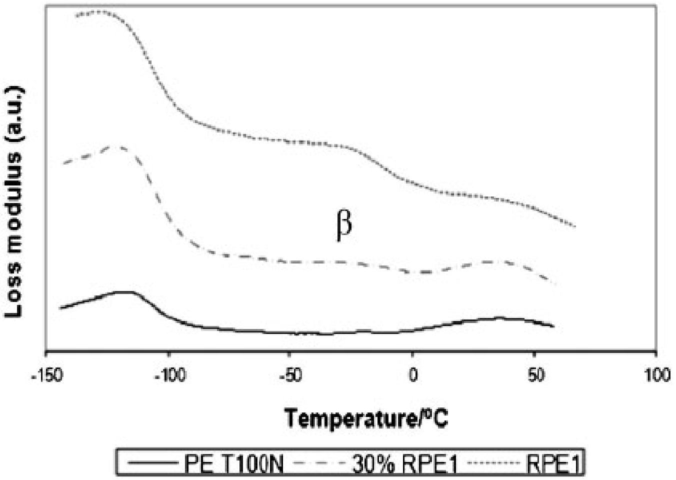

Figure 1.
Intensification of the β relaxation with the recycled content.

incorporation of recycled PE. The mass loss of HDPE T100N begins at 406 °C and the two blends containing 50% of recycled PE show starting of mass loss 60 °C below this value. The incorporation of recycled PE lowered also the glass transition temperature (Tg), suggesting an increase in the blend plasticity, which confirms that recycled PE has a plasticizer effect. The intensification of the β relaxation increases with the recycled content (Figure 1), and assign this transition with the onset of movements in the amorphous phase, either in branching[8] or main chain segments.[9]

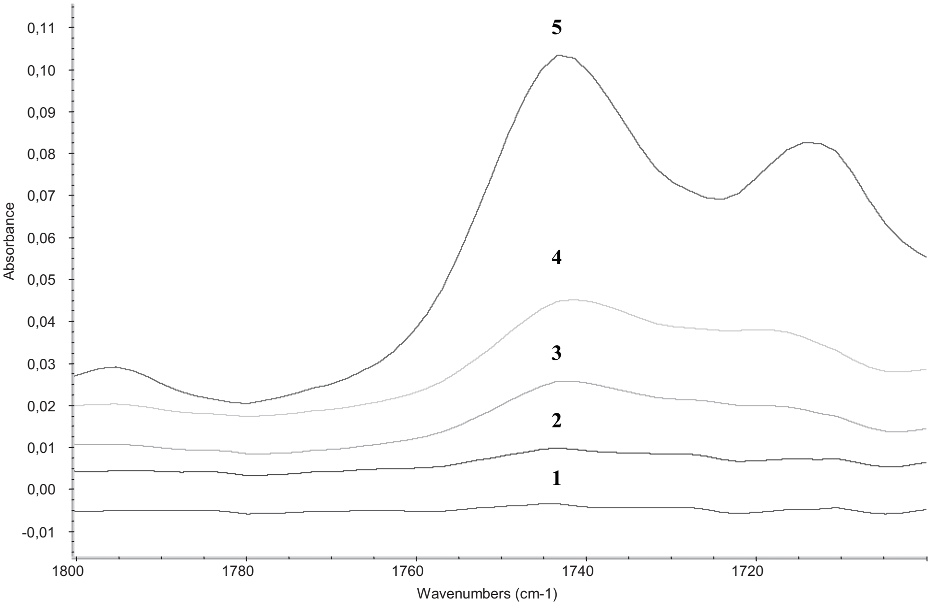

Figure 2.
Absorbance in the wavenumber region 1800–1700 cm^{-1} of samples and blends: 1- PE T100N; 2–10% recycled; 3–30% recycled; 4–50% recycled; 5- Recycled.

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All the blends and the recyclates showed the C=O bonds in the FTIR spectra. Indeed, the progressive incorporation of recycled PE in the blends induces the appearance of bands in the carbonyl region (Figure 2), near 1742 cm^{-1} and 1714 cm^{-1}, which magnitude increases with recycled PE content, confirming the presence of oxidation products resulting from previous degradation of such polymers.

Melt flow rate (MFR) is an indirect measure of molecular weight of a polymer and illustrates the ability of the material's melt to flow under pressure. The addition of 10% of recycled PE to the PE T100N didn't induce changes on this property. The presence of 30% and 50% of recycled PE in the blend (independently of its type) increased the MFR in 25% and 75%, respectively.

Conclusion

The incorporation of recycled PE in the polymeric blends based on PE T100N leads to poorer properties, namely the thermoxidative resistance, decomposition and glass transition temperatures. The melt flow rate increases significantly for high contents of recycled PE, but the melting and nucleation temperatures are not affected. The carbonyl group is detected in FTIR spectra of the blends, which is indicative of previous oxidative degradation on the recycled PE. To improve the properties of the blends it may be useful to upgrade them with suitable additives, as nanoclays and antioxidants. The results of such upgrading will be presented in a following work.

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