

# Study of Compatibilization of HDPE–PET Blends by Adding Grafted or Statistical Copolymers

N. TORRES,<sup>1</sup> J. J. ROBIN,<sup>2</sup> B. BOUTEVIN<sup>2</sup>

<sup>1</sup> CERE MAP, route des Salins, B.P. 118, 34140 Meze, France

<sup>2</sup> UMR 5076, Laboratoire de Chimie Macromoléculaire, Ecole Nationale Supérieure de Chimie de Montpellier, 8 rue de l'École Normale, 34296 Montpellier Cedex 5, France

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**ABSTRACT:** The reactive compatibilization of blends of HDPE–PET [high-density polyethylene–poly(ethylene terephthalate)] was investigated in this study. The compatibilizers used were two grafted copolymers prepared by reactive extrusion containing 1.20–2.30 wt % GMA such as HDPE-*g*-GMA and one statistical copolymer containing 1 wt % GMA such as Lotader AX8920. HDPE was successfully functionalized using a melt free-radical grafting technique. Grafting was initiated in two ways: adding an initiator in the polymer–monomer mixture or activation by ozone of polymer. Ozonization of HDPE by the introduction of a peroxide lead to a better grafting yield and to better grafting efficiency of the samples. The effects of the three compatibilizers were evaluated by studying the morphology and the thermal and mechanical properties of HDPE–PET (70/30 wt %) blends. Significant improvements were observed, especially in morphology, elongation at break, and Charpy impact strength of the compatibilized blends. A more pronounced compatibilizing effect was obtained with the statistical copolymer, for which the elongation at break and the impact strength were increased by 100%, while the uncompatibilized blends showed a 60% decrease in the Young's modulus and the strength at break. We also were able to show that the grafting yield increase of 1.20–2.30 wt % of GMA did not affect the properties of the blends because the grafted copolymers possess very similar chemical structures. However, compatibilization of blends with grafted copolymers is an interesting method, particularly for recycled blends, because the synthesis of these compatibilizers is easy and cheap in comparison to statistical copolymer. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 81: 2377–2386, 2001

**Key words:** high-density polyethylene; poly(ethylene terephthalate); compatibilization; grafted copolymer; reactive extrusion

## INTRODUCTION

Because polyolefins and polyesters are widely used thermoplastic polymers, they constitute a significant portion of postconsumer wastes. For

example, an useful method for upgrading the properties of polyolefins is the blending of high-density polyethylene (HDPE) with engineering plastics such as poly(ethylene terephthalate) (PET). However, the immiscibility of HDPE and PET in the melt state leads to blends possessing large interfacial tension, poor interfacial adhesion, and mediocre mechanical properties<sup>1,2</sup>. As shown in several studies, the compatibility and

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Correspondence to: J. J. Robin.

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**Table I** Characteristics of Polymers

Polymers	References	$M_w$ (g/mol)	$M_n$ (g/mol)	Density (g/cm <sup>3</sup> )	MFI (g/10 min)	$[\eta]$ (dl/g)	$T_m$ (°C)
HDPE (Dow)	10262A, pellets	64,000	27,000	0.962	10 (190°C, 2.16 kg)	—	130
HDPE (Solvay)	K38-20P, powder	235,000	8500	0.938	0.90 (190°C, 5 kg)	—	126
PET (Akzo)	MO3-300, pellets	44,000	—	1.40	—	0.76	248

adhesion of polyolefin–polyester blends can be improved by the addition of suitable copolymers, which act as emulsifying agents at the interfaces. The most widely used reactive functional groups to compatibilize these blends are acrylic acid,<sup>3</sup> maleic anhydride,<sup>4–9</sup> epoxy groups,<sup>2,10–14</sup> and oxazoline<sup>15</sup> which can react with the hydroxyl or carboxyl end groups of PET. However, the epoxy groups are more reactive than the others functions because they can react with the carboxyl (preferentially) and hydroxyl end groups of PET.

GMA-functionalized polyolefins synthesized by solution copolymerization are relatively expensive to be used in large quantities in such products as recycled HDPE–PET blends. Therefore, other methods of preparing compatibilizers, such as the radical grafting of GMA onto polyolefins, have been investigated. For example, Sun et al.,<sup>16,17</sup> Yao et al.,<sup>13</sup> Cartier et al.,<sup>18</sup> and Champagne et al.<sup>14</sup> grafted GMA onto polyolefins (PE or PP) using a peroxide as the initiator. The grafting, carried out in a batch mixer or in an extruder, led to copolymers containing a maximum of 1.2 wt % GMA. In this study we also prepared grafted copolymers by melt free radical grafting of glycidyl methacrylate (GMA) onto HDPE. Two methods were used to initiate the grafting: the adding of an initiator in the polymer–monomer system or ozonization of the polymer. We ozonized the polymer to increase the grafting yield of copolymers because it has been shown that the treatment of polymer with the ozone followed copolymerization with a vinylic or acrylic monomer is an interesting way to prepare grafted copolymers.<sup>19–26</sup>

Because it has been shown that the compatibilizing effect depends on the structure of copolymer used, the object of this study was to evaluate the effect of grafted copolymers prepared by reactive extrusion, such as HDPE-*g*-GMA containing 1.20–2.3 wt % GMA and statistical copolymer such as Lotader AX8920 containing 1 wt % GMA, on the compatibilization of HDPE–PET blends. Gaylord<sup>27</sup> has shown that block copolymers, particularly diblock copolymers, are more effective

than the grafted copolymers. Fayt et al.<sup>28,29</sup> have shown that triblock copolymers are less effective than diblock copolymers. So the effects of various compatibilizers on the morphology, the thermal properties, and such mechanical properties such as Young's modulus, strength, and elongation at break and impact strength of HDPE–PET (70:30 wt %) blends were investigated. The HDPE–PET (70:30 wt %) blends without compatibilizer were also prepared, and their properties were compared with those of the compatibilized blends. The uncompatibilized and compatibilized blends were prepared directly by injection molding. This melt processing is an interesting industrial method because it is more economical than the processing of polymer blends in two steps (extrusion and injection molding).

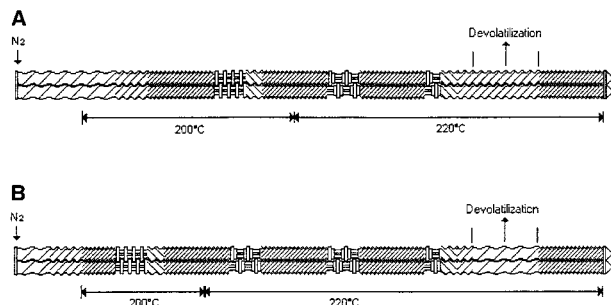
## EXPERIMENTAL

### Materials

The polymers used in our study are listed in Table I. Glycidyl methacrylate (GMA), with a purity of 97%, was purchased from Aldrich. *tert*-Butyl cumyl peroxy (trigonox T), from Akzo, is a peroxide with a purity of 98% that is soluble in the GMA, and its estimated half-lifetime is close to 30 sec at 190°C. PolyGMA was synthesized by polymerization of GMA in the presence of AIBN at 60°C in the THF. It has a number-average molecular mass ( $M_n$ ) close to 25,000 g/mol and a polydispersity index of 5.20 (determined by gel permeation chromatography). The statistical copolymer used as compatibilizer was Lotader AX8920: ethylene–methyl acrylate–glycidyl methacrylate (70:29:1 wt %), purchased from Atochem. This product, achieved by a high-pressure polymerization process, possesses a melt index of 6 g/10 min, close to 20,000 g/mol, and is mainly amorphous (crystallinity < 5%).

### Ozonization of Polymer

HDPE powder was ozonized in a fluidized bed at 20°C for 40 min through an ozone current of 8.3



**Figure 1** Screw configurations A and B and temperature profiles used for the grafting procedures.

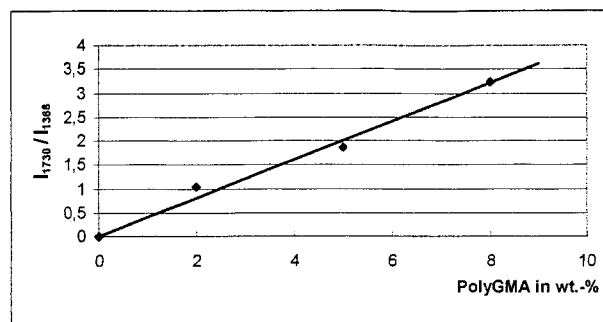
$10^{-3}$  g/l. The power of the ozone generator was 240 W, and the air/ozone gas flow was 1500 L/h.

### Grafting Procedure

Grafting in the molten state was carried out in a corotating intermeshing twin-screw extruder (Cletral BC21) with a length-to-diameter ratio of  $L/D = 36$ . The systems (HDPE-peroxide-monomer or ozonized HDPE-monomer) were introduced into the hopper of the extruder. This process was carried out under nitrogen current with a screw rotation speed of 150 rpm and a throughput of 3.5 kg/h. The screw configurations A and B and the temperature profiles used are shown in Figure 1. The screw profile was made up of conveying and kneading elements and also used opposite pitch to ensure melting, mixing, shearing, and good dispersing of the components. The extrudates were cooled in a water bath trough, pelletized, and dried at 60°C for 8 h. Then 5 g of the modified HDPE samples was purified by dissolution in hot xylene and precipitated into acetone at room temperature. This solvent permits the precipitation of the modified HDPE, the dissolution of the excess of monomer that did not react, and the elimination of homopolymer. The purified copolymers were dried under vacuum at 80°C, for 8 h.

### Determination of Grafting Yield

Infrared spectrometry was the most suitable method for determining the grafting yield of the synthesized copolymers. The analysis was performed with Fourier transform infrared (FTIR) spectrometry (Nicolet 510P). Thirty-two scans, each with a  $4\text{ cm}^{-1}$  resolution, were recorded. To determine the grafting yield of GMA onto HDPE, we used a calibration curve established with mechanical blends from HDPE and polyGMA (syn-

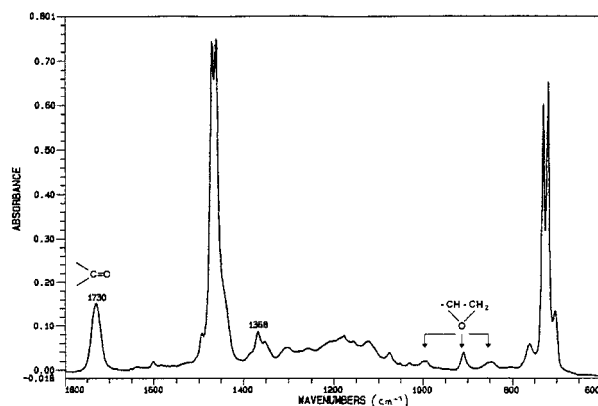


**Figure 2** Calibration curve for the determination of the grafting yield of GMA onto HDPE by FTIR.

thesized in our laboratory) in known proportions (2%, 5%, and 8% in weight). From these blends we prepared thin films (60 and 80  $\mu\text{m}$ ) by compression molding at 190°C for 2 min under a pressure of 120 bar. We calculated the ratio  $I_{1730}/I_{1368}$  ( $I_{1730}$ : intensity corresponding to the carbonyl group of polyGMA,  $I_{1368}$ : intensity corresponding to the symmetrical deformation of methyl groups in HDPE<sup>30</sup>) for every sample which we reported on a graph. The results enabled us to establish the calibration curve [ $I_{1730}/I_{1368} = f(\% \text{ polyGMA})$ ], which is shown in Figure 2. To determine the grafting yield of GMA in the purified copolymers, we calculated their ratio of intensity ( $I_{1730}/I_{1368}$ ) (Fig. 3) and reported these values on the calibration curve. This method can be applied because the infrared absorption of free polyGMA blended with HDPE is the same as that of the grafted GMA.

### Melt Processing

Before blending, pellets of PET were dried in a dehumidifying drier (5 h at 160°C). Blends of



**Figure 3** FTIR spectrum of copolymer HDPE-g-GMA.

**Table II** Grafting Yield and Grafting Efficiency of GMA onto HDPE

Experiment No.	Screw Configuration	Time of Ozonization (min)	[TrigonoxT] (wt %)	[GMA] <sub>i</sub> (wt %)	[GMA] <sub>g</sub> (wt %)	[GMA] <sub>g</sub> /[GMA] <sub>i</sub> (%)
1	A	—	0.20	3	0.60	20
2	A	—	0.20	6	0.80	14
3	A	—	0.40	3	0.90	30
4	A	—	0.60	3	1.00	34
5	B	—	0.40	3	1.20	40
6	B	40	—	3	2.30	77

HDPE–PET at composition (70:30 wt %) with or without compatibilizer (5 wt %) were injection-molded in the shape of ISO 1 test bars using a Billion 90-ton injection-molding machine. The typical molding conditions were as follows:

Barrel temperature: 250–280°C  
 Back pressure: 10 bar  
 Screw rotation speed: 100 rpm  
 Mold temperature: 40°C  
 Overall cycle: 40 s

### Characterization

To study the morphology of the blends, the samples were fractured in liquid nitrogen. The fractured surfaces were examined by scanning electron microscopy (SEM), using a Cambridge Stereoscan 260, after they have been coated with a thin conductive layer of palladium alloy. The enlargements are specified in the legends.

A PerkinElmer DSC-4 calorimeter was used to study the thermal properties of HDPE–PET blends with or without compatibilizer after injection molding. The temperature used was 50–280°C with a helium atmosphere, and the samples (7–10 mg) were heated at 10°C/min. The percent of crystallinity ( $\chi_c$ ) for HDPE was calculated from this equation:

$$\chi_c(\text{wt } \%) = 100 \times \frac{\Delta H_m}{\Delta H_m^0}$$

where  $\Delta H_m^0$  is the heat of fusion of 100% crystalline HDPE ( $\Delta H_m^0 = 288 \text{ J/g}$ ).<sup>31</sup>

Prior to being tested, the injection-molded specimens were conditioned at 20°C in a laboratory atmosphere for a minimum of three days. The reported values for all properties are the average of at least 10 determinations. Tensile

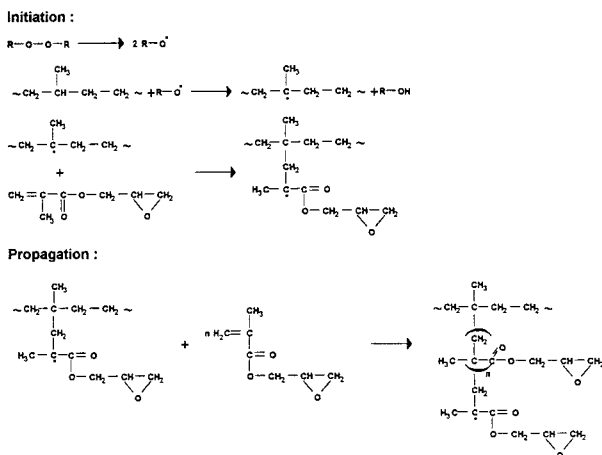
tests were performed according to ISO 527, using a Zwick tensile tester model 5101. Young's modulus measurements were made at a crosshead speed of 1 mm/min, while elongation measurements were made at 50 mm/min. Impact tests were performed according to Charpy ISO 179, notched specimens, using a Zwick pendulum impact tester model 5102 (2 J).

## RESULTS AND DISCUSSION

### Synthesis of Grafted Copolymers

The infrared spectra of purified samples show a peak at 1730 cm<sup>-1</sup> (Fig. 3) and three peaks of weak intensity—typical of deformation vibrations (symmetrical and asymmetrical) of the epoxide function—at 990 cm<sup>-1</sup>, 908 cm<sup>-1</sup>, and 850 cm<sup>-1</sup>,<sup>32</sup> confirming that grafting has occurred. The quantity of grafted GMA was calculated from the ratio of the intensities of the absorbance peaks at 1730 cm<sup>-1</sup> and 1368 cm<sup>-1</sup> by using the previously established calibration curve (Fig. 2). Table II gives the grafting yield and grafting efficiency of the grafted copolymers prepared in the twin-screw extruder.

Under the applied conditions the grafting yield of GMA onto HDPE varied from about 0.60 to 2.30 wt %, depending on the initiator (peroxide concentration or ozone), the monomer concentration, and the screw configuration. The increase in GMA concentration (experiments 1 and 2) led to a higher grafting yield (from 0.60 to 0.80 wt %) but a lower grafting efficiency (from 20% to 14%). These results show that no developments of interest if the initial GMA concentration is multiplied by two. As shown in experiments 1, 3, and 4, a higher initial concentration of peroxide involves a higher grafting yield and a higher grafting effi-



**Figure 4** Scheme of radical grafting process onto polyolefins.

ciency. The grafting mechanism is based on the formation of free radicals, released by the thermal decomposition of peroxide. These free radicals generate macroradicals by hydrogen abstraction on HDPE chains, which will react with the GMA (Fig. 4). However, the instability of these macroradicals can lead to crosslinking rather than grafting. A higher initial concentration of the peroxide generates numerous macroradicals, leading to grafting and/or crosslinking. Because the samples are totally soluble in hot xylene, crosslinking reactions are avoided, and the process of grafting is predominant.

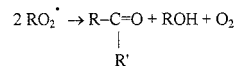
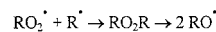
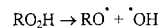
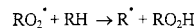
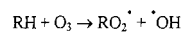
Table II shows that screw configuration B allows the obtaining of samples possessing higher grafting yield and grafting efficiency (experiment 5) than those prepared with screw configuration A (experiment 3). The estimated residence time, varying from about 60 s (screw configuration A) to 120 s (screw configuration B), is responsible for these improvements. All these results show that residence time is a determining parameter for the grafting yield of the samples. To compare ozonized polymer to the HDPE-initiator system, we prepared grafted copolymers from ozonized HDPE. When polyethylene is oxidized with ozone, radicals come from active oxygen atoms formed upon the decomposition of ozone. Each radical immediately reacts with an oxygen molecule, producing a peroxy radical. Some peroxy radicals make hydroperoxide by pulling out the hydrogen atom out of polyethylene, and other peroxy radicals make alcohol or ketone by recombination. The breaking by heating of the O—O bond of the hydroperoxide leads to alkoxy and hydroxyl rad-

icals that will produce copolymers and homopolymers,<sup>33,34</sup> respectively (Fig. 5). Ozone oxidation has been used to graft vinyl monomers onto polyethylene or polypropylene when the grafting was carried out in a Brabender batch mixer. To determine peroxide concentration, we used the iodometric method described by Johnson and Siddiqui.<sup>35</sup> The peroxide concentration was about  $10^{-5}$  mol/g of polymer. The initial concentration of the monomer was fixed at 3 wt %, and screw configuration B offered the best compromise between grafting yield and grafting efficiency. The ozonization of polymer led to a doubling of the grafting yield and the grafting efficiency of the samples (experiment 6). As shown in previous studies,<sup>13,14,17</sup> the grafting of GMA onto PE or PP leads to copolymers containing 0.20–1.20 wt % of reactive functions. Consequently, the samples of experiments 5 and 6 were used to compatibilize *in situ* HDPE-PET blends.

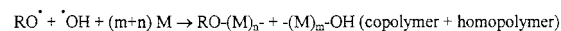
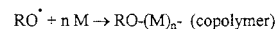
### Compatibilization of HDPE-PET Blends

Torres et al.<sup>2</sup> have shown that mechanical properties of HDPE-PET blends prepared by injection molding, particularly elongation at break and impact strength, are improved by adding small quantities (5 wt %) of GMA-functionalized polyolefins. The best mechanical properties are obtained when the emulsifying agent has a weak rate of reactive functions (1 wt %), that is, in the presence of Lotader AX8920. Recently, Champagne et al.<sup>14</sup> have shown that polypropylene grafted with 0.20–1.20 wt % GMA is an effective

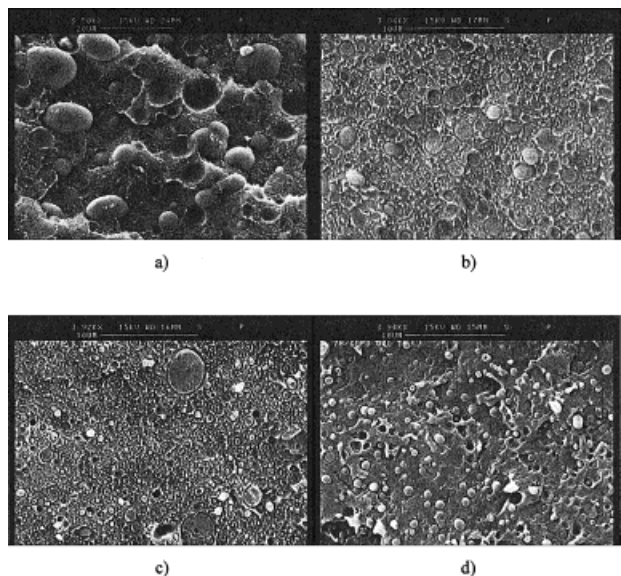
#### Initiation :



#### Propagation :



**Figure 5** Scheme of radical grafting process onto polyolefins oxidized with ozone.



**Figure 6** Photomicrographs of HDPE–PET (70:30 wt %) blends with various compatibilizers prepared by injection molding: (a) 70:30 wt % ( $\times 3500$ ); (b) 70:30 (wt %) with 5 wt % AX8920 ( $\times 3860$ ); (c) 70:30 (wt %) with 5 wt % HDPE-*g*-GMA (experiment 5) ( $\times 3920$ ); (d) 70:30 (wt %) with 5 wt % HDPE ozonized-*g*-GMA (experiment 6) ( $\times 3940$ ).

reactive compatibilizer for PP–PET blends. Their studies<sup>2,14</sup> have shown that low rates of reactive functions are sufficient to compatibilize polyolefin–polyester blends.

We thus studied the morphology and thermal and mechanical properties of compatibilized HDPE–PET (70:30 wt %) blends. The compatibilizers tested were HDPE-*g*-GMA ([GMA]<sub>g</sub> 1.20 wt %, experiment 5), HDPE ozonized-*g*-GMA ([GMA]<sub>g</sub> 2.30 wt %, experiment 6), and Lotader AX8920 (1 wt % GMA). In this work we compared the effects of copolymers grafted with a statistical copolymer on compatibilization of the blends. These blends were prepared directly by injection molding. Melt processing is interesting from an industrial point of view because it permits the transformation of polymer blends in one step.

### Morphology

SEM photomicrographs of HDPE–PET blends with compatibilizers are shown in Figure 6. The HDPE–PET (70:30 wt %) blends without compatibilizer possess a coarse morphology [Fig. 6(a)], with larger-size domains, as compared to compatibilized blends (Table III). The large particle size, with no evidence of adhesion between the

matrix and dispersed phase, confirms the incompatibility of the two components. In comparison, compatibilized blends with only 5 wt % compatibilizers added [Fig. 6(b–d)] show well-dispersed PET particles inside the HDPE matrix, smaller-size domains, and an improvement of interfacial adhesion. Under the applied conditions the morphologies of compatibilized blends depend on the interfacial tension, which is controlled by the structure of compatibilizer. It seems that the Lotader AX8920 is more effective than grafted copolymers in reducing interfacial tension because the blend with 5 wt % AX8920 possesses a better interfacial adhesion and a better dispersion of PET particles in relation to blends with 5 wt % grafted copolymers (Table III).

### Thermal Properties

The thermal properties of compatibilized blends during the first heating run were compared using differential scanning calorimetry (DSC). That permitted some indications of the microstructure of HDPE and PET in the test bars. These indications can be directly related to the mechanical properties of material. The samples were taken from the core of the test bars. Appearing on the thermograms are two endothermic peaks associated with the fusion of the crystalline fractions of HDPE and PET. The melting temperatures ( $T_{m,max}$ ) of HDPE and PET in the blends with or without compatibilizer are close to those of pure HDPE and PET (Table IV). This behavior is normal for immiscible polymers in the melt state.<sup>36,37</sup> Park et al.<sup>4</sup> obtained similar results on the compatibilization of HDPE–PET blends. The enthalpy of melting ( $H_m$ ) and the degree of crystallinity ( $\chi_c$ ) of HDPE decrease when PET is added to the blends (Table IV). This tendency is more pronounced when the blends are compatibilized with 5 wt % AX8920 or 5 wt % HDPE ozonized-*g*-GMA (experiment 6). These results show that interactions have been created between HDPE and PET in presence of these two compatibilizers. But these decreases can also come from the degree of crystallinity of the compatibilizers [ $\chi_c = 5$  wt % for AX8920,  $\chi_c = 47$  wt % for HDPE ozonized-*g*-GMA (experiment 6)] because it is very inferior to these of the HDPE matrix ( $\chi_c = 75$  wt %). Because these two compatibilizers are compatible with HDPE, they disturb its crystallization. The enthalpy of melting ( $H_m$ ) of PET increases strongly when HDPE is added to the blends. This tendency is more pronounced when

**Table III Photomicrograph Observations of HDPE-PET (70:30 wt %) Blends with Various Compatibilizers**

Composition of Blends	Diameter of Nodules ( $\mu\text{m}$ )	Dispersion	Interfacial Adhesion	Photo No.
70 HDPE-30 PET	4-10	Poor	Poor	6a
70 HDPE-30 PET with 5 wt % AX8920	0.8	Good	Good	6b
70 HDPE-30 PET with 5 wt % HDPE-g-GMA (experiment 5)	0.6-3	Medium	Medium	6c
70 HDPE-30 PET with 5 wt % HDPE ozonized-g-GMA (experiment 6)	0.6	Good	Medium	6d

the blends are compatibilized. The HDPE and compatibilizers can act as nucleating agents, facilitating the crystallization. We did not calculate the degree of crystallinity of pure PET ( $\chi_c$  (wt %) = 100%).

$$\frac{\Delta H_m - |\Delta H_c|}{\Delta H_m^0}$$

because we did not know its enthalpy of crystallization. Indeed, the absence of the exotherm of cold crystallization<sup>38</sup> arising from the crystallization of the amorphous phase located between 130°C and 160°C comes from either of the endotherms of melting of HDPE, coinciding with the cold crystallization of PET or the ordering of chains of PET, which does not occur. The results recorded during the first heating run show that the microstructural characteristics of HDPE and PET in the uncompatibilized or compatibilized blends are modified with regard to those pure polymers. It would seem that compatibilizers such as Lotader AX8920 or HDPE ozonized-g-

GMA (experiment 6) have a much greater effect on the crystallinity of polymers than do HDPE-g-GMA (experiment 5).

The thermal properties of compatibilized blends during the first cooling run were studied by DSC. On the thermograms of the blends appears an exothermic peak that arises from the crystallization of HDPE. The absence of the exotherm of crystallization of PET in the uncompatibilized or compatibilized blends containing high concentrations of polyolefin has been observed by Jabarin<sup>39</sup> and by Xanthos.<sup>3</sup> They maintain that the absence of the exotherm of crystallization of PET comes from a very slow crystallization that occurs over a large temperature domain, leading to a flattened peak undetectable by DSC. To confirm this hypothesis, we recorded the thermal properties of compatibilized blends (70:30 HDPE-PET with 5 wt % AX8920) during a second heating run at 10°C/min. On the thermogram recorded during this run, we observed the melting peak of PET, also indicating that crystallization of PET occurred during the first cooling run. The

**Table IV Thermal Properties of HDPE-PET (70:30 wt %) Blends with Various Compatibilizers Recorded During First Heating Run**

	HDPE			PET	
	$T_{m,\max}$ (°C)	$\Delta H_m$ (J/g of HDPE)	$\chi_c$ (wt %)	$T_{m,\max}$ (°C)	$\Delta H_m$ (J/g of PET)
HDPE	134	216	75	—	—
PET	—	—	—	247	9
70 HDPE-30 PET	132	210	73	247	26
70 HDPE-30 PET + 5 wt % AX8920	133	199	69	244	34
70 HDPE-30 PET + 5 wt % HDPE-g-GMA (experiment 5)	134	210	73	247	29
70 HDPE-30 PET + 5 wt % HDPE ozonized-g-GMA (experiment 6)	133	200	69	248	35

crystallization temperature ( $T_{c,\min}$ ) of HDPE in the blends with or without compatibilizer is close to that of pure HDPE (Table V). The presence of PET in the blends decreases the enthalpy of crystallization of HDPE because the growth of crystalline units of HDPE perturbed by PET domains of different sizes; therefore, a part of the energy usable for crystallization is lost.<sup>40</sup>

Tables IV and V show that the crystallization of HDPE decreases in the blends while that of PET increases and that this tendency is more pronounced when the blends are compatibilized with Lotader AX8920 and HDPE ozonized-*g*-GMA (experiment 6) because these two compatibilizers unlike HDPE-*g*-GMA (experiment 5) possess degrees of crystallinity very inferior to the degree of crystallinity of HDPE matrix.

### Mechanical Properties

Table VI gives the mechanical properties of HDPE–PET (70:30 wt %) blends with various compatibilizers. Lotader AX8920 allows multiplying by two the elongation at break and impact strength of the blends. Inversely, this compatibilizer leads to a decrease of Young's modulus and the strength at break about 60% as compared to uncompatibilized blends. The grafted copolymers, HDPE ozonized-*g*-GMA (experiment 6) and HDPE-*g*-GMA (experiment 5), decrease the strength at break of about 60% and increase the elongation at break about 50% compared to uncompatibilized blends. It can be noted that the decrease of Young's modulus and the increase of impact strength compared to the uncompatibilized blends are more pronounced when the blends are compatibilized with HDPE ozonized-*g*-GMA (experiment 6). The good interfacial adhesion of blends compatibilized with Lotader AX8920 can explain the increase in the elongation at break and the impact strength, while the decrease in crystallinity of the HDPE matrix leads to a loss of rigidity and an improvement in impact strength.

### CONCLUSIONS

This study has shown that the structure of the compatibilizer determines the morphology and the mechanical properties of the blends. As we have shown, it can be seen that statistical copolymer is more effective in compatibilizing HDPE–PET blends than grafted copolymers. These results are in disagreement with the literature,<sup>41</sup>

**Table V Thermal Properties of HDPE–PET (70:30 wt %) Blends with Various Compatibilizers Recorded During First Cooling Run**

	$T_{c,\min}$ (°C)	$\Delta H_c$ (J/g of HDPE)
HDPE	118	218
70 HDPE–30 PET	118	206
70 HDPE–30 PET + 5 wt % AX8920	118	196
70 HDPE–30 PET + 5 wt % HDPE- <i>g</i> -GMA (experiment 5)	118	211
70 HDPE–30 PET + 5 wt % HDPE ozonized- <i>g</i> -GMA (experiment 6)	118	195

which had found that a hanging group is more reactive than a group inside a macromolecular chain. Thereby, it is necessary to take into account parameters other than the conformation (statistical or grafted) of compatibilizer. Grafted copolymers formed mainly of HDPE are miscible in the matrix and so are not located preferably at the interface, unlike statistical copolymer, which is compatible but not miscible with HDPE. Being preferably placed at the interface, Lotader AX8920 ensures a better anchorage of the dispersed phase, leading to materials with better mechanical properties compared to blends compatibilized with the grafted copolymers. An additional explanation for the properties of blends compatibilized with grafted copolymers might be the relatively high amount of free monomer (GMA) and homopolymer (PGMA) present in the blends. They could prevent the compatibilization reaction by reacting quickly with the end groups of PET.<sup>15,42</sup> These two reasons could explain why grafted copolymers prepared as described here are less effective than statistical copolymer in compatibilizing HDPE–PET blends. This work has shown that all the grafted copolymers improved morphology, elongation at break, and impact strength of the blends compared to uncompatibilized blends. Moreover, they led to blends higher possessing Young's moduli than those blends compatibilized with Lotader AX8920. The increase in grafting yield of 1.2–2.3 wt % does not affect the properties of blends because these two grafted copolymers possess very close chemical structures. The introduction of grafted copolymers to compatibilize the HDPE–PET blends is



**Table VI Mechanical Properties of HDPE-PET (70:30 wt %) Blends with Various Compatibilizers**

	Young's Modulus (N/mm <sup>2</sup> )	Strength at Break (N/mm <sup>2</sup> )	Elongation at Break (%)	Charpy Impact Strength (Notched, 20°C, kJ/m <sup>2</sup> )
70 HDPE-30 PET	1266 (±100)	19.0 (±1.5)	40 (±4)	3.0 (±0.2)
70 HDPE-30 PET + 5 wt % AX8920	761 (±56)	12.0 (±1.0)	80 (±7)	6.2 (±0.4)
70 HDPE-30 PET + 5 wt % HDPE- <i>g</i> -GMA (experiment 5)	1176 (±129)	13.4 (±1.4)	64 (±6)	5.0 (±0.2)
70 HDPE-30 PET + 5 wt % HDPE ozonized- <i>g</i> -GMA (experiment 6)	1000 (±81)	12.1 (±1.3)	60 (±7)	5.7 (±0.3)

an interesting method because the synthesis of these compatibilizers is not complex and not expensive compared to statistical copolymers. It could be interesting to prepare grafted copolymers from a polymer compatible but not miscible with HDPE because it would preferably place it at the interface as with Lotader AX8920. This method will be examined in a future article.

## NOMENCLATURE

*i* initial  
*g* grafted

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