Polymer Bulletin 41, 91–98 (1998)

Polymer Bulletin

Influence of a copolymer on the mechanical properties of a blend of PP and recycled and non-recycled HDPE

C. Albano*, G. Sánchez, A. Ismayel

Universidad Central de Venezuela, Facultad de Ingeniería, Escuela de Ingeniería Química,
P.O. Box 48146, Los Chaguaramos, Caracas 1041-A, Venezuela

Received: 23 May 1997/Revised version: 5 May 1998/Accepted: 14 May 1998

Summary

The mechanical and morphological behavior of polypropylene (PP) and high-density polyethylene (HDPE), recycled and non-recycled, by the addition of an ethylene-propylene block copolymer, was studied. In the non-recycled samples, the effect of the copolymer on the mechanical and morphological properties is negligible. In the recycled samples, it was shown that with 5% copolymer, both particle size in the dispersed phase and interface thickness decreased, and that 5% copolymer composition is optimal in improving the adhesion and flexibility of the blend.

Introduction

Blends are commercially interesting due to the possibility of combining the attractive features of the different polymers and improving deficient properties. Almost all blends are immiscible and have poor physical and mechanical properties in comparison to pure components. This is due to the little interaction between phases, produced by interfacial tension in the melted blends. This phenomenon makes deformation of the disperse phase in the blend during the mixing process difficult, and results in poor interfacial adhesion in the solid state (1).

The addition of a third component, which reduces interfacial tension and improves compatibility, and the mixing of functional polymers able to interact through chemical reactions are two of the methods used to improve interfacial adhesion.

The efficiency of a compatibilizing agent in a polymeric matrix is analyzed through the affinity with the matrix, the distribution in the melted material related to the miscibility and grade of dispersion, and the stability of the microstructure.

D’Orazio et al. (2,3) observed that the random addition of an ethylene-propylene copolymer affects the binary morphology of PP/HDPE blends, improving their mechanical properties. Vicki-Flaris et al. (4) studied PP/LLDPE (Polypropylene/Linear Low-Density Polyethylene) polyblends with a block copolymer and concluded that impact strength is enhanced.

The use of recycled components increases the possibility of immiscibility. Consequently, the use of a compatibilizing agent is important.

---

* Corresponding author

Therefore, this research is aimed at analyzing the influence of a block copolymer on polyblends of PP with non-recycled and recycled HDPE.

Experimental

To fulfill the objectives of this study, the recycled material was degraded by 15-20% in wt. The degree of degradation was determined by measuring the variation in viscosity of HDPE exposed to 280°C for varying lengths of time (5). Blend of PP (Profax, MFI: 6.5g/10min, p: 0.903g/cm^3) with 30% non-recycled HDPE (Polímeros del Lago, MFI: 5.0g/10min, p: 0.953g/cm^3) or with 30% recycled HDPE were mixed in an extruder with different weight compositions of an ethylene-propylene block copolymer (Propilco, IF: 8.0g/10min p: 0.899g/cm^3) containing 8% ethylene.

Subsequently, the specimens were compression-molded. Tensile testing was carried out using a Universal Instron test machine. Impact testing (Izod) was also performed.

The samples were treated with ruthenium tetroxide so their microstructure could be studied by TEM, and were fractured in liquid N_2 and coated with gold for SEM study.

Results and Discussion

Figures 1 and 2 show the mechanical properties of the blends containing different concentrations of block copolymer. A blend of 70% PP with non-recycled and recycled (r) HDPE was chosen (Table 1), since analysis of the mechanical properties of different compositions of both polymers showed that at this concentration good mechanical properties were obtained, and that the deviation of properties with respect to PP is relatively small. For the PP/recycled-HDPE blend, the same composition was chosen, for similar reasons, and because a high percentage of recycled material is used (6).

HDPE(%)	E (Kg/mm^2)	σ_R (Kg/mm^2)	ε_R(%)	IR(J/m)
0	64±8	3.6±0.1	12. 6±0.7	7.0±1.0
5	69±4	3.2±0.3	8.0±2.0	6.0±1.0
10	78±6	3.4±0.3	12.0±2.0	6.2±0.9
20	88±6	3.1±0.4	8.0±2.0	8.1±0.5
30	99±5	3.7±0.2	9.0±1.0	8.5±1.0
50	85±4	2.7±0.3	6.5±0.6	2.6±0.2
100	48±5	2.3±0.3	960.0±50.0	28.0±4.0
5 r	68±8	3.6±0.2	10.1±0.8	9.3±1.0
10 r	73±5	3.4±0.2	8.6±0.9	8.5±1.1
20 r	72±5	3.1±0.3	7.1±0.9	9.3±0.9
30 r	68±9	3.2±0.2	6.8±0.9	8.7±0.5
40 r	65±3	3.1±0.2	8.3±0.8	7.5±0.8
50 r	69±10	2.9±0.1	7.1±0.3	7.0±0.5
100 r	55±4	1.6±0.7	560.0±100.0	38.3±5.0

Table 1: Variation of the mechanical properties of PP with non-recycled and recycled HDPE. (E: Young's modulus, ε_R: Elongation at break, σ_R: Final tensile strength, IR. Impact strength).

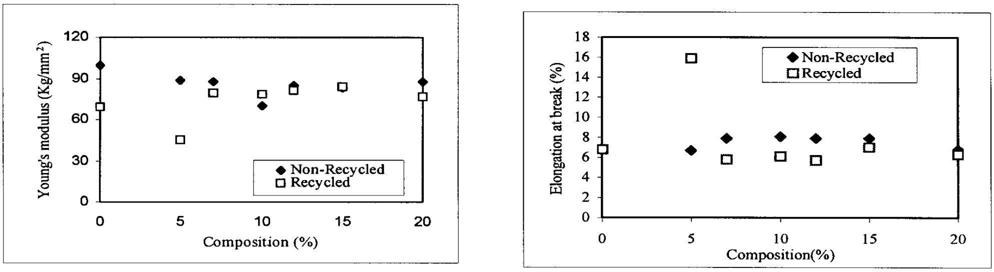

Figure 1: Variation of a) Young's modulus and b) Elongation at break as a function of compatibilizing agent composition.

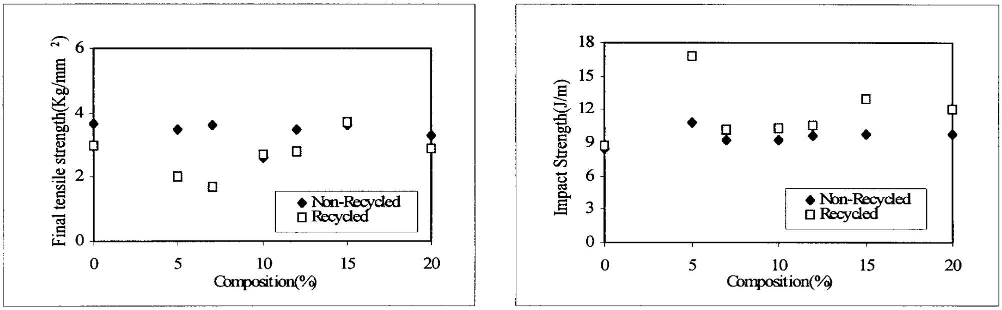

Figure 2: Variation of a) Final tensile strength and b) Impact strength as a function of compatibilizer composition.

The elongation at break of the PP/recycled-HDPE blend (Figure lb) is atypical in its behavior. Impact strength shows characteristics (Figure 2b) similar to the elongation at break, i.e. maximum values are reached at 5% copolymer composition, and subsequently values similar to those of a blend without copolymer are maintained.

In the case of Young's Modulus (Figure 1a), a drastic decrease to 5% copolymer can be observed for the PP/recycled-HDPE blend and then the modulus values are similar to those of the blend without copolymer.

Final tensile strength for the compound with recycled HDPE (Figure 2a) slightly decreases to 5% copolymer, and then at higher copolymer compositions it increases until reaching the value of the blend without compatibilizing agent.

The action of the copolymer was found to be negligible in the PP/non-recycled-HDPE blend for almost all of the mechanical properties, because no significant variations were observed (Figures 1-2). This indicates that the agent has not been able to increase either the adhesion or the flexibility of the composite material, or to modify the brittleness.

In the case of the PP/recycled-HDPE blend, analyses showed that mechanical properties vary only at 5% copolymer. Elongation at break and impact strength, properties related to the tenacity and flexibility of the material, increase at this point, probably because the blend absorbs a greater amount of energy. As a result, the material shows greater resistance to crack propagation under impact conditions, implying an increase in adhesion and flexibility. It can be deduced that at this same composition, Young's modulus decreases, indicating a reduction in the brittleness of the polyblend.

According to Barlow et al. (7), block copolymers can increase ductility to a greater extent than random types, and simultaneously decrease Young's modulus and resistance at break. In contrast, Dumoulin et al. (8) found that the addition of block copolymers to PP/LDPE results in an increase in rigidity and brittleness, and thus in an increase in the values of Young's modulus and final tensile strength.

Figure 3: SEM micrograph of PP/non-recycled-HDPE blend without copolymer. Scale bar: 5μm (_____)

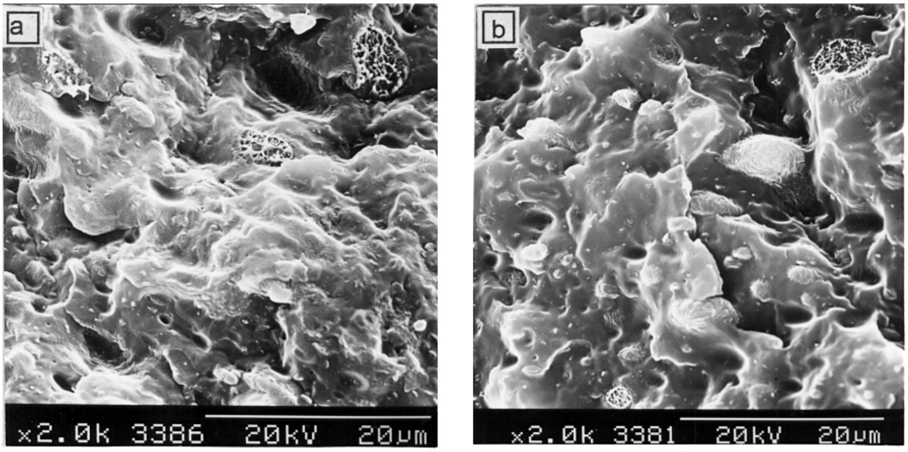

Figure 4: SEM micrographs of the PP/ non-recycled-HDPE blend with copolymer a) 5%; b) 15%.

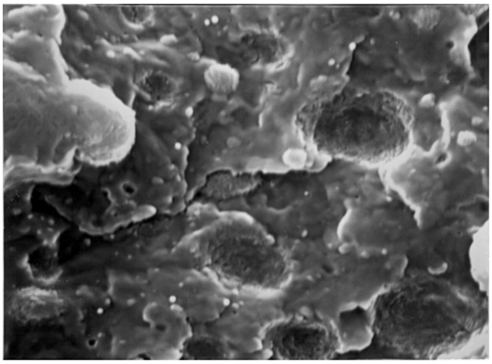

Figure 5: SEM micrograph of the PP/recycled-HDPE blend without copolymer. Scale Bar: 5μm (_____).

Micrographs of fracture surface for blends of 70% PP with non-recycled HDPE (Figures 3 and 4) show high uniformity of the domain size when studied as a function of the compatibilizing agent concentration.

In mixtures of PP with recycled HDPE without copolymer, fracture surface micrographs (Figures 5 and 6) show recycled-HDPE domains with diameters over 3μm.

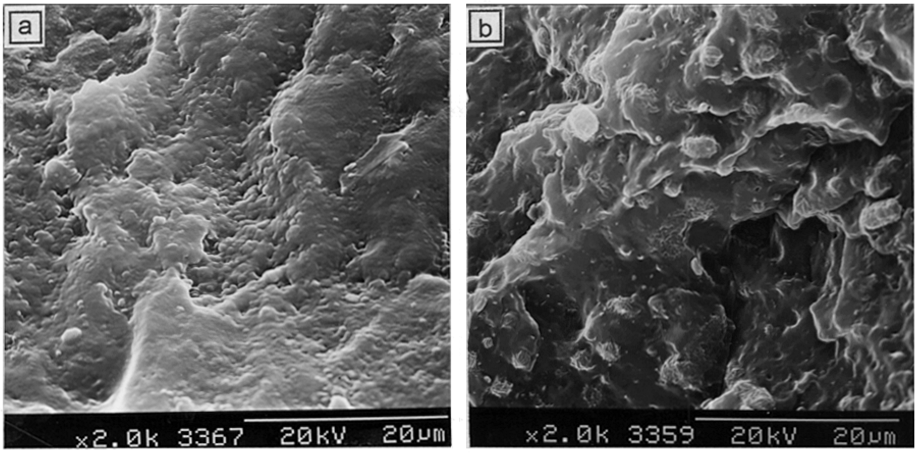

Figure 6: SEM micrographs of the PP/recycled-HDPE blend with copolymer.
a) 5%; b) 15%.

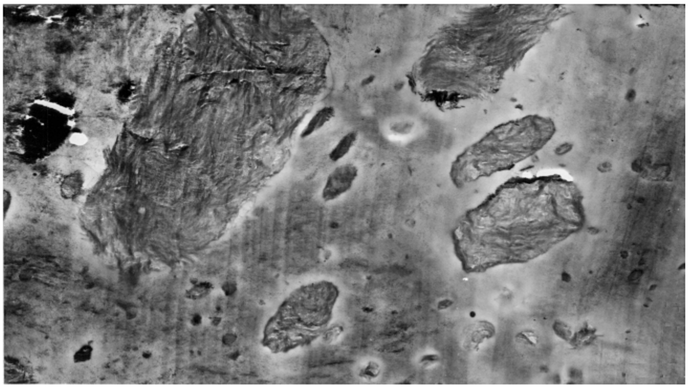

Figure 7: TEM micrograph of PP/non-recycled-HDPE blend. Scale bar : 1.4µm (______).

The addition of 5% copolymer gives rise to a large change in morphology, to the extent that no disperse phase is observed in the PP matrix. This proves that the copolymer acts as a compatibilizing agent, enhancing properties such as elongation at break and impact strength. Experimentation additionally shows that when the concentration is increased up to 15%, these properties show no further changes.

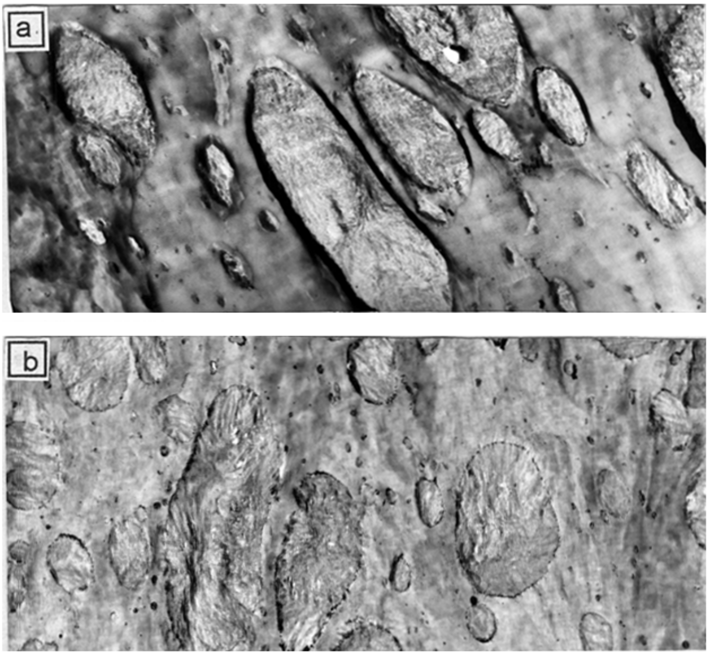

Figure 8: TEM micrographs of the PP/non-recycled-HDPE blend with copolymer.
a) 5%; b) 15%. Scale bar: a) 1.7µm, b) 2.1µm. (______).

Figure 9: TEM micrograph of the PP/recycled-HDPE blend. Scale bar: 2.8µm. (______).

TEM micrographs of PP/non-recycled-HDPE blends, both with and without copolymer, (Figures 7 and 8) show no appreciable variations in the sizes of HDPE domains (0.5-3µm), as well as no significant changes in the interface. This results are similar to those obtained from the analysis of their mechanical properties.

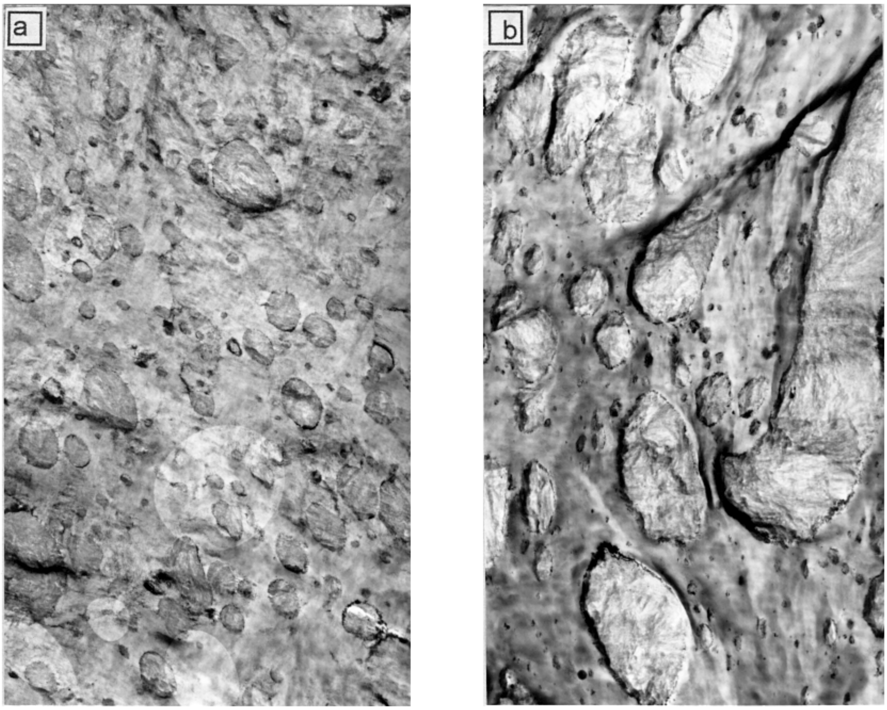

Figure 10: TEM micrographs of the PP/recycled-HDPE blend with copolymer. a) 5%; b) 15%. Scale bar: 1.7µm (______).

Analysis of the micrographs of the blend of 70% PP with 30% recycled HDPE (Figures 9 and 10), with and without copolymer at different compositions, reveals that the addition of a compatibilizing agent at 5% composition decreases both particle size as well as interface thickness. At 15% copolymer composition results similar to those shown for the blend without copolymer are obtained, with interface thickness varying between 400 and 600 Å, whereas in the blend using 5% copolymer composition, the interval obtained varies between 165 and 200 Å.

It can be concluded that the addition of this agent at the above mentioned composition (5%) is the optimal concentration for improving the final properties and the finished product. From these results, it can be deduced that at 5%, the compatibilizing agent reaches its saturation level.

The action of a copolymer in immiscible polymer blends is ruled by interface width and interfacial tension (9,10), hence the importance of attaining an optimal interface thickness level, which is obtained through a balance between entropy, enthalpy, and the corresponding interfacial tension.

According to Noolandi (11), a decrease in interface thickness leads to a reduction in interfacial tension, due to the action of the block copolymer, its molecular weight, and the orientation of the blocks in the interface.

In the micrographs, disperse particles of this compatibilizing agent can be seen in the matrix phase, located at the interface between the two homopolymers.

Transmission electronic microscopy was used to establish disperse phase particle size for the blends under study. Values fluctuate between 0.5 and 5 pm, that is, within the ‘appropriate’ range (0.32-1µm), as designated by Taylor (12) and Lee et al. (13), so that major deterioration of mechanical properties can be avoided.

Conclusions

As a result of the present research, it can be concluded that the optimal composition of this copolymer for PP/recycled-HDPE blends is 5%, due to the increase in adhesion and flexibility obtained. In PP/non-recycled-HDPE blends, the action of this copolymer is not significant.

This research was sponsored by CDCH. We gratefully acknowledge their financial support.

References

1. Lui NC, Baker WE (1992) Advances in Polymer Technology 11(4): 249.
2. D’ Orazio L, Greco R, Martuscelli E, Ragosta G (1982) Polym Eng Sci 22(9): 536.
3. D’ Orazio L, Greco R, Martuscelli E, Ragosta G (1983) Polym Eng Sci 23(9): 486.
4. Vicki F, Zbigniew Stachurski H (1992) Journal Appl Polym Sci 45: 1789.
5. Arribas G, Milanovich A, Sánchez G, (1981) Acta Científica 32:64.
6. Albano C, Reutilización de Plásticos: estudio térmico, mecánico y termodegradativo de PP con PEBD y PEAD degradado, PHD. UCV. Caracas, Venezuela, 1995.
7. Barlow JW, Paul DR (1984) Polym Eng Sci 24(8): 525.
8. Dumoulin MM, Farha C, Utraki LA (1984) Polym Eng Sci 24(17):1319.
9. Wu S (1987a) Journal Polym Sci Part B: Polym Phys 25: 2511.
10. Wu S (1987b) Polym Eng Sci 27(5): 335.
11. Noolandi J (1991) Makromol Chem Rapid Commun 12: 517.
12. Taylor GI (1954) Proc R Soc London 226A: 34.
13. Lee K, Jeong T, Kim Ch (1991) Polym Eng Sci 31(13): 944.