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A. G. Andreopoulos[a]; P. A. Tarantili[a]; P. Anastassakis[a]
[a] Department of Chemical Engineering, National Technical University of Athens, Zografou, Greece

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COMPATIBILIZERS FOR LOW DENSITY POLYETHYLENE/POLYPROPYLENE BLENDS

A. G. Andreopoulos,* P. A. Tarantili, and P. Anastassakis

Department of Chemical Engineering
National Technical University of Athens
9 Iroon Polytechniou Str.
15780 Zografou, Greece

ABSTRACT

The role of polyolefin elastomers as compatibilizers in Low Density Polyethylene/Polypropylene (LDPE/PP) blends, in the presence of di-cumyl peroxide (DCP) has been studied. Mixtures of 90/10 LDPE/PP ratio, were prepared in a Brabender plasticorder and tested for their mechanical properties and calorimetric response. Then the elastomers ethylene-propylene-diene copolymer (EPDM) and polybutadiene (BR) were added, alone or together with 0.2% DCP at concentrations up to 2%. The mixing torque and gel content of the above products were recorded as a function of the blend consistency. Also, the mechanical properties of specimens were measured as an additional evidence to explore the capabilities of these additives to promote compatibility of the blend components. It was found that EPDM and BR can be easily incorporated into polyolefin blends and appear suitable as potential compatibilizers for those materials, probably acting within the PP phase. Both elastomers result in an increase of strength and modulus, the BR having more enhanced effect. The latter gives low elongation, which allows its use in applications where high tensile properties are desired and flexural behavior is not critical.

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* Author to whom correspondence should be addressed.

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INTRODUCTION

It is well known that polyblends represent a very important class of polymeric materials because they can be used in a broad spectrum of applications. Surprisingly, polymeric blends and alloys often present far better properties compared to those of their components and therefore they display great versatility.

The improvement of properties which usually takes place after mixing two or more polymers, is the basis for a series of new materials. For instance, the rubber toughened polymers contain small particles of elastomers dispersed into a plastic matrix. This combination displays better impact strength compared to that of the initial polymer alone [1].

Polyblends have also been studied in the context of recycling, since waste materials usually contain polyolefins and vinyl polymers and their miscibility is of great importance. It should be noted that miscibility is associated with the solubility of the components to each other whereas compatibility is rather estimated via the overall mechanical response of the heterogeneous mixture [2].

Recent reports on polymeric blends include the effect of polypropylene contamination on the properties of recycled high density polyethylene [3] and the development of the related modelling [4]. Blends based on polyolefins, containing filers or reinforcements have also been studied [5-8]. Many parameters must be taken into consideration when predicting compatibility of the components of a blend. Such parameters are molecular weight and molecular weight distribution, rheological properties, morphology and mechanical properties [9]. However, it is common practice to use additives which are specified for improving miscibility of the components of the blend, the so called compatibilizers [10]. These additives are either chemicals acting at the interface or low molecular weight copolymers, which contain the suitable units and groups capable of promoting compatibility among the polymers of a certain mixture which otherwise would not display a synergistic effect [11, 12]. Thus, copolymers of maleic anhydride have been reported [13] along with alkyl carboxyl-substituted polyolefins [14], ethylene-co-vinyl acetate and ethylene-co-acrylic copolymers [15]. Other attempts focus on the mixing techniques and the content of compatibilizer [16, 17]. In our previous work, [18] a reactive compatibilizer for blends of polyolefins, namely di-cumyl peroxide (DCP), was studied as an attempt to develop a new kind of means for improving properties of polymeric blends. It was found that the incorporation of DCP restricts the thermoplastic characteristics of the melt, which was primarily attributed to chain extension and branching which occur in LDPE. This results in an enhancement in the adhesive bonding between the two polymers mainly due to chain entanglements. This was further supported by the fact that mechanical properties of the treated blend were significantly improved.

In the present work, the role elastomers, such as ethylene-propylene-diene copolymer (EPDM) and polybutadiene (BR), as compatibilizers for LDPE/PP blends was investigated. EPDM was reported to be an efficient compatibilizer for PE, PVC and PET blends (19) and was successfully introduced in polyolefin mixtures for the preparation of various products (20). Our attempt was to study the combined action of the above elastomers with dicumyl peroxide and evaluate the efficiency of these systems as compatibilizers for LDPE/PP blends.

EXPERIMENTAL

Materials

LDPE, with the trade name Alcudia, was supplied by Repsol Quimica, Spain. The density of the polymer was 0.917 g/ml and its melt flow index 7 g/10 min. Similarly the PP, supplied by Appryl (Spain), had a density of 0.905 g/ml and a melt flow index of 6 g/10 min. As to the elastomers, the EPDM with the trade name KELTAN 512 was supplied by DSM (Holland) and had 38% PP content whereas BR (trade name SKD) with a density of 0.91 g/ml and 90% cis-1,4 content, was supplied by WURFBAIN (Germany). The di-cumyl peroxide Merck (Germany) was 98% pure.

Preparation of the Blends

A Brabender plasticorder was employed for preparing the blends. The plasticorder is capable of indicating and recording the mixing torque of the melt and was adjusted at the following working parameters: temperature of mixing head: 200°C, speed: 20 rpm, time: 20 minutes. A hydraulic press equipped with the suitable molds was used for molding the mixtures obtained from the plasticorder into rectangular sheets with 3 mm thickness. Molding was carried out at 200°C and 100 atm for 5 minutes. Tensile specimens were cut from the above sheets according to ASTM D-638, with a suitable die Zwick (Germany).

Characterization and Testing of Samples

The gel content of the blends containing DCP was determined by Soxhlet extraction with boiling toluene, for 4 hours. The procedure is described in detail in reference 21 and provides us with information about the extent of crosslinking.

The heat of fusion and melting temperatures for the obtained blends was determined by Differential Scanning Calorimetry (DSC), using a Perkin-Elmer (Switzerland) apparatus, type DSC 4. Samples of about 10 mg were used and the scan was carried out in the range 50 to 200°C at a heating rate of 10°C/min.

The tensile properties of the LDPE/PP specimens were determined with a Zwick (Germany) tensometer according to ASTM D 638.

RESULTS AND DISCUSSION

The variation of the mixing torque, when the elastomers EPDM and BR are incorporated in 90/10 LDPE/PP blends are shown in Figures 1 and 2, respectively. In both cases, an increase of torque is recorded with increasing elastomer

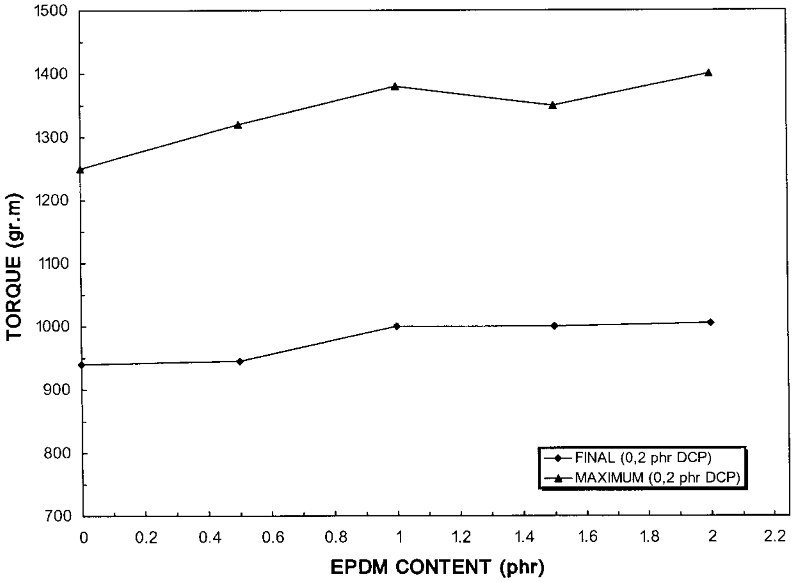

Figure 1. The mixing torque in systems LDPE/PP containing EPDM and DCP.

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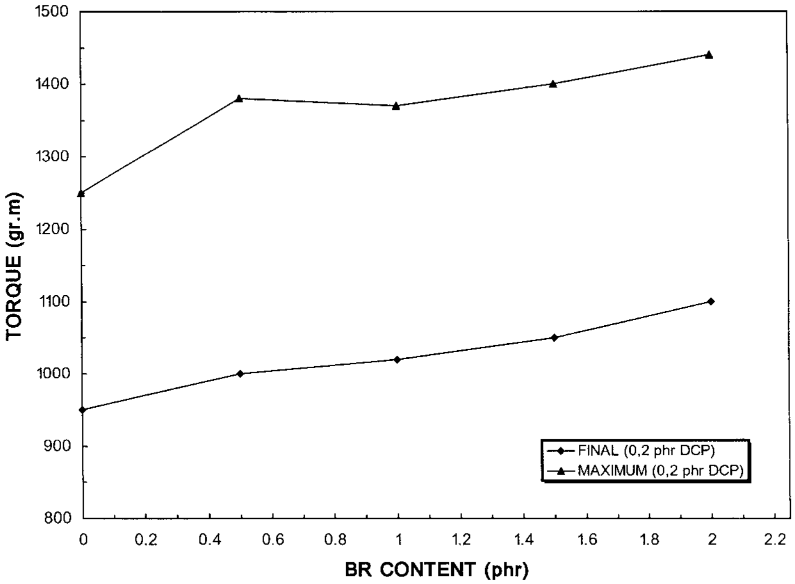

Figure 2. The mixing torque in systems LDPE/PP containing BR and DCP.

concentration, which could be attributed either to the rheological characteristics of the blend itself or to a possible involvement of the elastomers added in the reactions initiated by di-cumyl peroxide and more specifically, the cross-linking reaction (18). In order to further explore the above possibilities, the gel content of some of these blends was determined, as presented in Figure 3. From the curves of this figure, it can be seen that the gel content does not display a clear dependence on the concentration of EPDM or BR and therefore, it can be concluded that the viscosity characteristics of these elastomers are responsible for the observed increase in the torque that the blend displays during melt mixing of the blend. The effect of EPDM and BR on the structure of LDPE/PP blend, in terms of induced changes in crystallinity and melting points was also studied. The results for samples containing elastomers with or without DCP, are presented in Table 1.

From the data in this table, it can be concluded that the incorporation of the elastomers studied reduces the heat of fusion of the blend and this effect is more enhanced in the presence of peroxide. This appears reasonable since the action of peroxide clearly results in a reduction of crystallinity content in polyolefins, as chain mobility and possibilities of packing are restricted. Very inter-

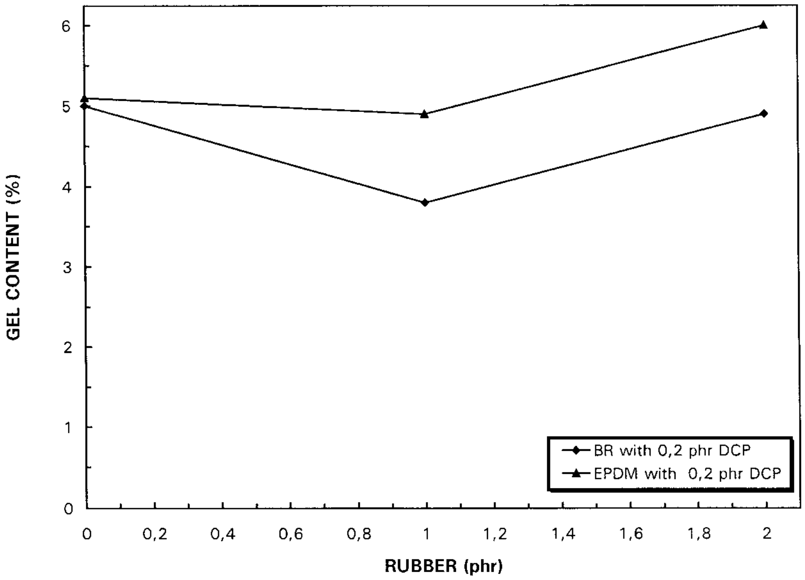

Figure 3. The gel content of blends containing elastomers and DCP.

estingly, the reduction seems to be similar for the two elastomers, and this can be due to the interaction of its chains mainly with the PE lattice, which greatly contributes to the enthalpy of fusion. As to the melting points (m.p.), it is clear again that the presence of di-cumyl peroxide causes, in general, a depressive effect. On the other hand, in the case of PP an increase of the m.p. is recorded upon the incorporation of the elastomers alone. This effect could probably be attributed to some nucleation occurring within the PP phase when the elastomers

TABLE 1. Results from DSC Measurements for LDPE/PP Blends Containing EPDM, BR and DCP

Additive	PE Melting Point (°C)	PP Melting Point (°C)	Heat of Fusion (cal/gr)
None	113,6	166,7	7,1
2phr EPDM	113,2	167,1	6,7
2phr EPDM/0.2phr DCP	112,3	166,3	6,3
2phr BR	113,5	167,8	6,8
2phr BR/0.2 phr DCP	112,1	166,9	6,3

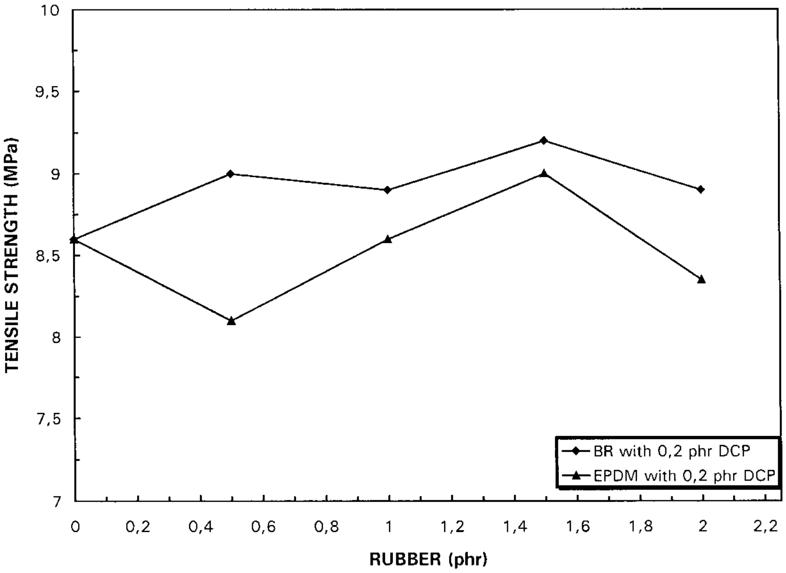

Figure 4. The tensile strength at break for various LDPE/PP blends containing EPDM and BR.

are blended. This results in an increase in m.p. which is not accompanied by an increase in the overall heat of fusion, since PP does not contribute significantly to the crystallinity content.

The mechanical properties of specimens molded from 90/10 LDPE/PP blends containing EPDM and BR, were recorded in terms of tensile strength and modulus. Figure 4 shows the strength at break for specimens containing the above elastomers in concentrations up to 2%. It is evident that some improvement in strength takes place for concentrations in the area of 1.5%, for both elastomers. On the other hand, the modulus of elasticity presents a maximum at concentrations of about 1%, as in Figure 5, can be seen.

As far as the elongation at break of the same specimens is concerned, Figure 6 shows its dependence on the elastomer concentration, and a considerable decrease in the case of BR is observed whereas with the incorporation of EPDM there are no significant changes in elongation. It should be noted that the reduced elongation of specimens containing BR denotes poor flexibility.

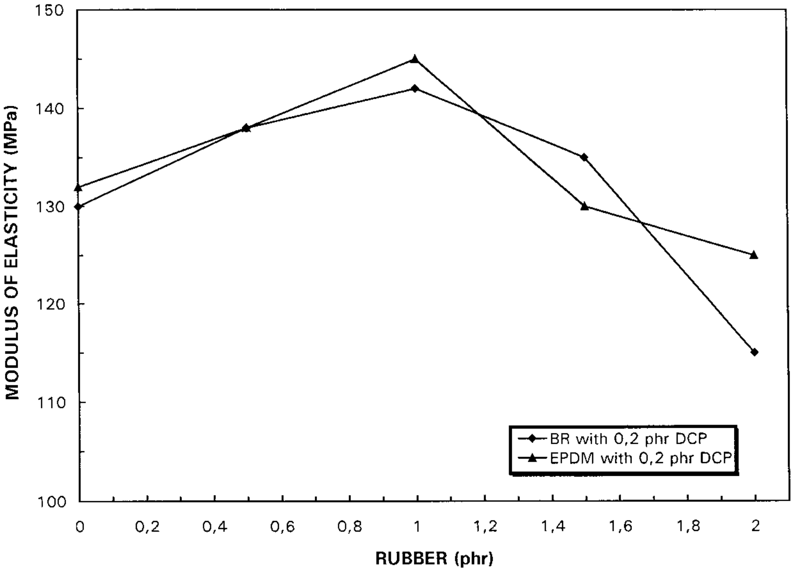

Figure 5. The modulus of elasticity for various LDPE/PP blends containing EPDM and BR.

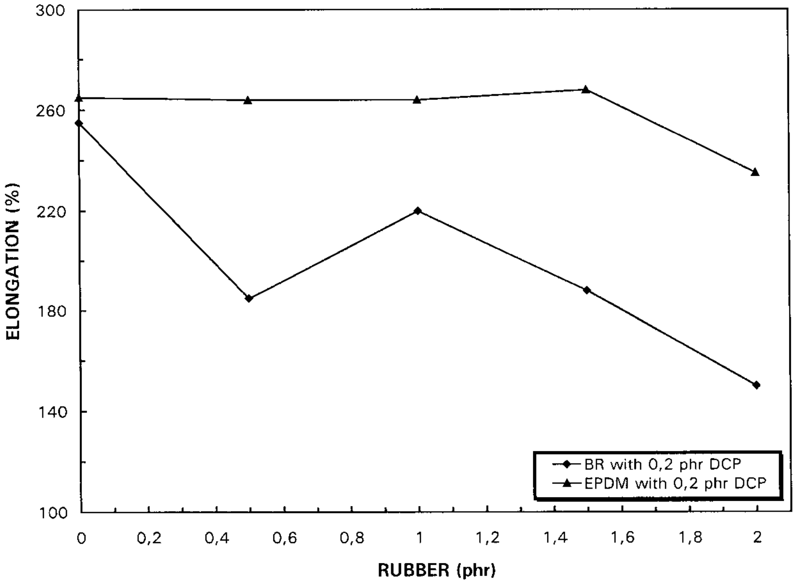

Figure 6. The elongation at break for various LDPE/PP blends containing EPDM and BR.

CONCLUSION

From the above results, the following conclusions can be drawn: EPDM and BR are elastomers with interesting performance in LDPE/PP blends and appear suitable to be used as compatibilizers. They are easily incorporated into those blends and their main function seems to be located in the PP phase. Both elastomers result in an increase of strength and modulus, the BR having more enhanced effect. The latter gives low elongation, which allows its use in applications where high tensile properties are desired and flexural behavior is not critical.

REFERENCES

[1] A. G. Andreopoulos and G. C. Papanicolaou, *J. Mat. Sci., 22*, 3417 (1987).

[2] L. A. Utraki, *Polym. Eng., Sci. 22*, 1166 (1982).

[3] A. A. Adewole, K. Dackson, and M. D. Wolkowicz*, Antec '94 Conference Proceedings*, San Fransisco CA, May 1-5, 1994, 012 Corp. Ed., SPE Vol. III, p. 3044-9.

[4] M. J. Orroth and R. A. Malloy, *Antec '94 Conference Proceedings*, San Fransisco CA, May 1-5, 1994, 012 Corp. Ed., SPE Vol. III, p. 3059-63.

[5] W. G. Ma and F. P. La Mantia, *Polymer Recycling, 1*, 69 (1995).

[6] S. A. R. Hashmi, A. K. Majumdar, and N. Chand, *J. Mat. Sci. Lett., 15*, 1343 (1996).

[7] M. M. Miller, J. M. G. Cowie, J. G. Tail, D. L. Brydon, and R. R. Mather, *Polymer, 36*, 3107 (1995).

[8] W. Zhu, X. Zhang, B. Huang, and Z. Feng, *J. Appl. Polym. Sci., 58*, 515 (1995).

[9] S. H. Hamid and M. Atiqullah, *J. Macromol. Sci., C35*, 495 (1995).

[10] D. Vesely, *Polym. Eng. Sci., 36*, 1574 (1996).

[11] M. Xanthos, *Polym. Eng. Sci., 28*, 1392 (1988).

[12] M. Xanthos and S. S. Dagli, *Polym. Eng. Sci., 31*, 929 (1991).

[13] H. Chang-Sik, P. Hae-Dong, K. Youngkyoo, K. Soon-ki, and C. Won-Jei, *Polym. Adv.Technol., 7*, 438 (1996).

[14] J. T. Yeh, C. C. Fan-Chiang, and M. F. Cho, *Polym. Bull., 35*, 371 (1995).

[15] R. L. Mc Envoy and S. Krause, *Macromoleules 29*, 4258 (1996).

[16] A. R. Nesarikar, S. H. Carr, K. Khait, and F. M. Mirabella*, J. Appl. Polym. Sci., 63*, 1179 (1997).
[17] K. Cho, T. K. Ahn, B. H. Lee, and S. Choe, *J. Appl. Polym. Sci., 63*, 1265 (1997).
[18] P. K. Sen Gupta and J. C. Bevington, *Polymer, 14*, 527 (1973).
[19] F. P. La Mantia and C. Perrone, *Poliplasti Plast. Rinf. , 40*, 58 (1992).
[20] J. J. Scobbo, *Makromol. Chem. Macromol. Symp., 57*, 345 (1992).
[21] E. M. Kampouris and A. G. Andreopoulos, Biomaterials, 10, 106 (1989).

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