Optimization of Aramid Powder Reinforced PP/EPDM Blends

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Summary: The effect of twaron powder on the tensile behavior and abrasion resistance of polypropylene (PP) and an ethylene-propylene diene terpolymer (EPDM) and their blends has been investigated by means of an experimental design. The stiffness and strength of the composites with EPDM as continuous phase hardly varies with the addition of twaron powder, however, at high percentages of PP (above 50%) in the matrix, these properties decrease as powder content increases. In all cases the elongation at fracture decreases in the presence of the powder. A maximum in the abrasion resistance is observed in the 14% twaron powder filled polypropylene composite. From the results and the graphs of the response surfaces a better affinity between the twaron powder and the EPDM can be suggested. It can be also deduced that as twaron content in the composite increases, the phase inversion in the polymer matrix takes place at lower PP percentages. A good correlation between mechanical characteristics and morphological observations has been observed.

Introduction

The protection of the environment and new recycling legislation has become of paramount importance to plastic manufacturers, processors and consumers: how to eliminate the plastic scrap and products once they have been used. So, thermosets and rubbers have been reduced for use in particular applications where their role cannot be carried out by other plastic materials. This was one of the reasons that encouraged the development of thermoplastic rubbers and within this large range are ethylene-propylene copolymers (EPM) and ethylene-propylene-diene terpolymers (EPDM) that display excellent impact resistance as well as very good stress-strain properties in a similar manner as rubbers[1–8]. In fact, elastomers based on blends of EPDM and polypropylene (PP) have increased dramatically in popularity over recent years. These blends commonly referred to as TPO´s, are particularly well suited for those applications requiring outstanding ageing and weathering characteristics, ozone and heat

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resistance, ease of fabrication and low cost. Due to the low cost, improved impact properties, and capability of recycling, TPO compounds are expanding their usage in automotive applications. However, the modulus [9, 10] as well as the yield stress[11] of the PP decrease with EPDM content. Consequently, the research on methods for reinforcing the PP/rubber blends has received considerable attention[12 - 15].

The aim of the present study is to carry out a systematic investigation on polypropylene/EPDM physical blends filled with aramid powder to analyze its effect on the behavior of these composites. The final goal is to get products with balanced properties and to analyze their behavior/morphology relationship.

Experimental

Materials

Isotactic polypropylene (PP) (Density: 0.905 g/cm^3 and MFI: 6 g/10 min) and ethylene-propylene-diene terpolymer (EPDM), obtained via "metallocene" (Density: 0.87 g/cm^3, Mooney Viscosity: ML-125°C: 25; ethylene/propylene ratio = 70/25, and ethylene-norbornene content: 5%, in pellets) were supplied by Repsol Química and DuPont Dow Elastomers, under the trade names of Isplen PP-050 and Nordel 4725 P, respectively.

Table 1: Properties for Twaron 5011

Bulk density		405 Kg/m^3	Measured according to DIN 53194
Density		559 Kg/m^3	Measured according to DIN 53194
Particle size distribution	d_{10}	30±5 μm	Measured with laser diffraction using ethanol (Sympatec laser)
	d_{50}	70±10 μm	
	d_{90}	135±15 μm	
Moisture Content		< 1.5	%
Moisture Absorption		< 8	% (20°C, 65% rel. Humidity)

Aramid powder, whose physical characteristics are compiled in Table 1, was supplied by Cordis under the trade name of Twaron 5011.

Planning the experiments

In order to analyze the influence of the matrix composition and Twaron powder percentage on the behavior of these composites, response surface methodology[16 - 18] was used. In this case the two-variables method was applied and the experimental mixtures were planned on the basis of an Uniform Net of Doehlert[19]. This type of design defines the minimum number of experimental combinations in the domain under consideration (100 to 0 % for the PP percentage in the matrix composition, and 20 to 0 for the twaron powder percentage in the composite) to maximize the information obtained. A second-degree equation has been postulated for this experimental design. From the results of the experiments, the equations of the response surfaces were deduced by means of multivariable analysis techniques and, from the equations, the graphs for each of the properties of the different composites were obtained.

From the graphs, different compositions can be selected to obtain a composite with specific mechanical properties and the relative behavior of different materials may be compared. The advantage of this type of experimental design is that besides the individual effect of each variable, their combined effect can also be analyzed. The composition of the samples utilized in this study are compiled in Table 2.

Table 2: Experimental combinations

Experiment no.	% PP in the polymer matrix	% twaron in the composite
1	100	10
2	75	18.66
3	25	18.66
4	0	10
5	25	1.34
6	75	1.34
7	50	18.66
8	50	1.34
9-11	50	50

Methods

The materials were compounded in a roll-mill at 180° C. Once the PP was melted, the appropriated amount of EPDM was added and homogeneously blended. Finally, the aramid powder was incorporated. The blending time was 30 min. After pelletization and drying the compounds were injection molded to prepare dog-bone specimens. The temperatures in the three zones of the injection molding machine were 210, 220 and 230° C, respectively. The period of time for the packing and cooling stages were 30 and 20 s, respectively. Tensile tests were carried out on an Instron T-4301 model, at room temperature and a cross-head speed of 5 and 50 mm/min for measuring the tensile modulus and ultimate tensile strength, respectively. An abrasion test was used to evaluate the resistance of the composites to wear. Standardized specimens for abrasion testing were obtained by compression molding of the injected materials, at 200° C in a Collins Press. The abrasion testing procedure followed ISO 4649: 1985 standard specifications.

Morphological study

In order to analyze the morphology of the composites, fracture surfaces of several samples were observed in a Jeol T330A scanning electron microscope.

Results and discussion

Tensile behavior

The tensile properties of the twaron powder composites are graphically represented in Figures 1 to 3. From Figure 1 it is deduced that the stiffness of the composites mainly depends on matrix composition and the twaron powder hardly affects the modulus of these materials. Only a slightly negative effect of this filler is observed at very rich PP matrices (above 50%).

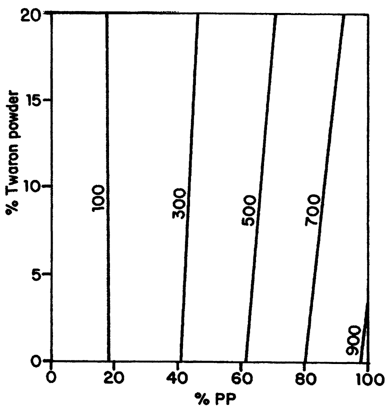

Figure 1. Tensile modulus (MPa)

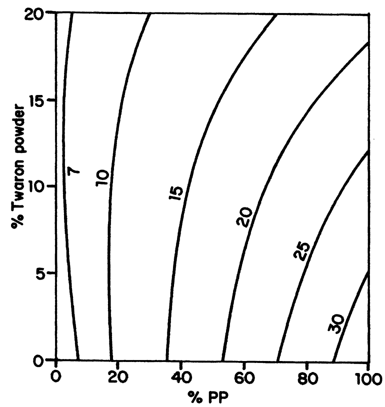

Figure 2. Ultimate tensile strength (MPa)

A similar behavior is shown by the surface responses of the ultimate tensile strength of the composites (Figure 2). In this case, the decrease of this characteristic is more noticeable at high PP percentages in the polymer matrix, which could be an indication of a poor affinity between the twaron powder and the polypropylene. A noticeable increase in the tensile strength is obtained with the increase of the PP percentage in the matrix composition.

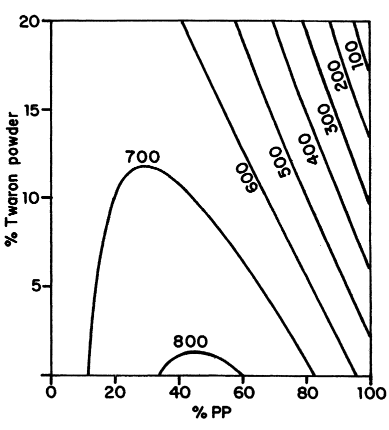

Figure 3. Elongation at break (%)

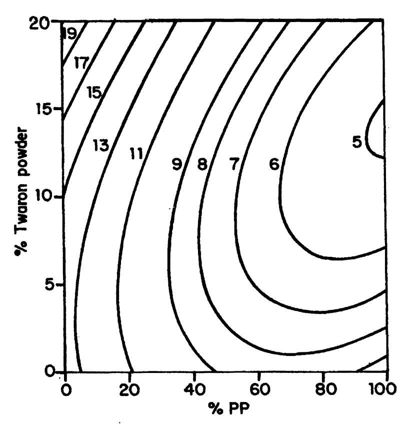

Figure 4. Abrasion resistance (mm^3)

The elongation to break (Figure 3) shows a very special behaviour which could be an indication that as twaron powder content in the composite increases, the phase inversion in the polymer matrix takes place at lower PP percentages. It is also observed that the effect of the twaron is more negative and sensible in PP rich matrices. From these results it can be confirmed that the aramid powder shows a better affinity with EPDM than with PP.

Abrasion tests

As can be seen in Figure 4, the abrasion resistance of the aramid powder composites normally increases as PP content in the matrix increases which is attributed to the higher hardness of this polymer. However, this effect depends on matrix composition. So, only in the case of very rich PP matrices, an increase in the abrasion resistance is observed as powder content increases, but above 13% powder content in the composite an opposite effect is observed. The lowest abrasion resistance is obtained with high EPDM percentages in the matrix and high powder contents in the composite. It can be suggested that powder particles may take the polymer matrix with them when scratched off the surface. Therefore the higher the powder content and the stronger the interaction at the interface, the higher the loss of volume produced in the abrasion test.

Morphological study

Fracture surfaces of samples based on 10% aramid powder filled polypropylene and EPDM are shown in Figures 5 and 6, respectively. As can be deduced from Figure 5, a very good dispersion and incorporation of aramid particles in the PP matrix is obtained. However, it seems that the surface of the holes, due to the extraction of small aramid particles during the breaking of the material, are very clean. In the case of the EPDM

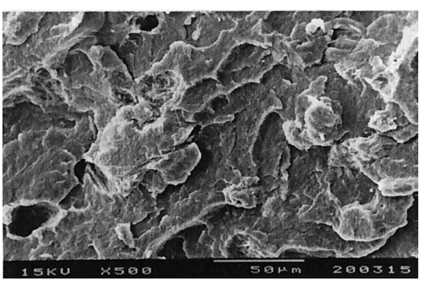

Figure 5. Fracture surface of 10% twaron filled PP

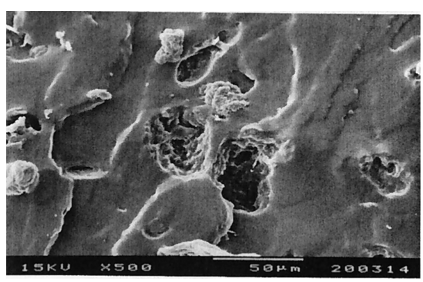

Figure 6. Fracture surface of 10% twaron filled EPDM

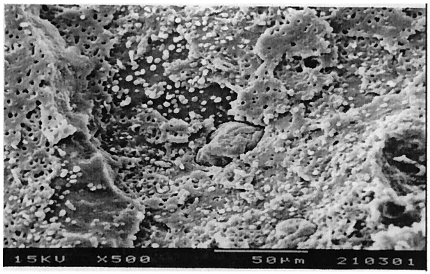

Figure 7. Fracture surface of 25/75 PP/EPDM blend matrix with 1.8% twaron

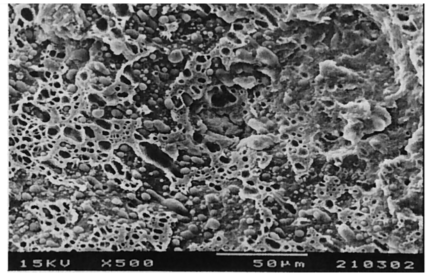

Figure 8. Fracture surface of 75/25 PP/EPDM blend matrix with 1.8% twaron

(Figure 6), these holes show a very rough surface which suggests that a better adhesion between the EPDM and aramid particles exists.

The fracture surface of the 25/75 PP/EPDM blend matrix with low aramid powder content is represented in Figure 7. As it can be seen, near the fracture notch there are many particles on the surface. In the center of this photograph a particle of twaron can clearly be distinguished The smallest particles and voids belong to PP dispersed material. On the other hand, the sample constituted by 75/25 PP/EPDM ratio polymer matrix and 1.8% of aramid powder, shows EPDM particles dispersed on the PP continuous phase. Many of these particles have been strained during the processing of these materials. Generally, they have very small diameters but with a wide particle size distribution. It is not easy to distinguish the aramid powder but one particle is in the center of the picture and big holes on the fracture surface can also be attributed to twaron particles. In general, a homogeneous dispersion of powder particles has been obtained in the powder composites.

Conclusions

From the above results the following conclusions can be deduced:

- Twaron powder hardly affects the modulus and strength of composites with EPDM as continuous phase, however at PP percentages above 50% these characteristics decrease as powder content in the compound increases.

- The affinity between the twaron powder and the EPDM is higher than between twaron and PP as indicated by the tensile behavior of the composites and scanning electron microscope observations.
- In all cases the addition of the powder gives rise to a decrease of the elongation to break as expected.
- The abrasion of twaron powder results in an increase of the abrasion resistance of PP rich matrices until a certain level of powder content is reached in the composite.

References

[1] A. Van der Wal, F. R. Nijho, R. J. Gaymans, *Polymer* **1999,** *40,* 6031.
[2] V. Choudhary, H. S. Varma, I. K. Varma, *Polymer* **1991,** *32,* 2534.
[3] L. D´Orazio, C. Mancarella, E. Martuscelli, F. Polato, *Polymer* **1991** *30,* 1186.
[4] K. C. Dao, *J. Appl. Polym. Sci.* **1982,** *27,* 4799.
[5] J. Karger-Kocsis, V. N. Kuleznev, *Polymer* **1982** *23,* 699.
[6] D. L. Faulkner, *J. Appl. Polym. Sci.* **1988,** *36,* 467.
[7] N. K. Kalfoglou, *Angew. Makromol. Chem.* **1985,** *129,* 103.
[8] L. D´Orazio, R. Greco, C. Mancarella, E. Martuscelli, G. Ragosta, C. Silvestre, *Polym. Engng. Sci.* **1982,** *22,* 536.
[9] B. Pukánszky, F. Tüdös, A. Kalló, G. Bodor, *Polymer* **1989** *30,* 1407.
[10] T. Yu, *SPE ANTEC Tech. Papers* **1995** *41,* 2358.
[11] A. K. Gupta, S. N. Purwar, *J. Appl. Polym. Sci.* **1984,** *29,* 3513.
[12] B. Pukánszky, F. Tüdös , J. Kolarik, F. Lednicky, *Polym. Compos.* **1990,** *11,* 98.
[13] H. Zhang, L. A. Berglund, *Polym. Eng. Sci.* **1993,** *33,* 100.
[14] J. Jancar, A. T. Dibenedetto, *J. Mater. Sci.* **1995,** *30,* 1601.
[15] P. R. Hornsby, K. Premphet, *J. Mater. Sci.* **1996,** *32,* 4767.
[16] K. E Peng, in: *"The Design and Analysis of Scientific Experiments",* Addison Wesley, Reading, MA, 1966.
[17] A. Barella, J. M. Tura, J. P. Vigo, H. O. Esperon, *J. Text. Inst.* **1976,** *67,* 10.
[18] G. Duménil, *Appl. Microbiol. And Biotechnol.* **1988,** *27,* 405.
[19] D. H. Doehlert, *Appl. Statistics* **1970,** *19,* 231.