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# Effect of the incorporation of pet fibers on the properties of thermoplastic elastomer based on PP/elastomer blends

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**Abstract**

Ternary composites based on isotactic polypropylene (iPP), thermoplastic elastomer, such as ethylene–octhene copolymer (EOC) and poly(ethylene-terephtalate) (PET) textile fibers, have been processed and their properties analyzed. The effect of matrix composition and fiber content on the final properties of the composite has been investigated by means of an experimental design based on a Doehlert Uniform Net. The results have shown that PET fibers act as an effective reinforcing agent, giving rise to a sensible improvement in the tensile and flexural behavior, mainly in matrices with high copolymer percentages. It must be pointed out that the analyzed mechanical properties depend more on the matrix composition than on the fiber percentage. So, as PP content increases, the blend becomes more rigid and stable and a noticeable increase in tensile and flexural modulus and strength, as well as a sensible decrease in tensile elongation are observed. The dynamic-mechanical analysis strongly supports the assumption that the PET fibers behave as a reinforcing agent, showing a sensible increase of the storage modulus in the presence of the fibers. Moreover, this increase is more noticeable as fiber content in the composite is increased. The morphology of the composites has been also analyzed through scanning electron microscopy (SEM). 

*Keywords*: Ethylene–octhene copolymer; Ternary composites; PET fibers

## 1. Introduction

Thermoplastic olefinic elastomers (TPOs) combine the excellent properties of vulcanized elastomers at service temperature [1,2] with the processing advantages of thermoplastics at elevated temperatures [3–5]. Because of their unique molecular configuration they may be processed through the same techniques used with conventional thermoplastics materials, but the mechanical properties of the final product are very similar to those of conventional vulcanized elastomers. The most typical industrial TPOs are those based on isotactic polypropylene (iPP) and ethylene–propylene-diene (EPDM) blends. In fact, because of its high impact strength over a wide temperature range, EPDM is considered as one of the most effective impact modifiers for iPP [6–10]. The main objective of the incorporation of the elastomeric phase to the PP is to improve its flexibility and behavior at low temperatures. However, the elastomeric phase gives rise to a sensible decrease in some mechanical properties, such as stiffness and hardness, so restricting the fields of application to these materials. In order to improve these properties, several studies about the influence of different fillers on these blends have been carried out. In fact, ternary composites based on particulate filler filled or fiber reinforced thermoplastic/elastomer blends have increased dramatically in popularity over recent years [11–13]. It is expected, that the incorporation of reinforcing agents, such as short fibers, will permit to reduce the cost of the material and to improve some properties of these systems. So, they have become attractive materials for numerous engineering applications. In addition, it is well known that the mechanical properties of these systems depend not only on the properties of the components and their proportion in the composite, but also on the morphology of the material, especially on the adhesion at the matrix/reinforcement interface.

Recently, a new type of thermoplastic rubbers synthesized via methallocene catalyst has been tested very successfully as PP impact modifier [14–16], and it has also been shown that a higher crosslinking density was reached with these new polymers [17]. This is what gave rise to the development of new TPOs based on PP/ethylene–octhene copolymer (EOC) blends for the conduction of the present study.

For the above mentioned, the main goal of the present

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Table 1
Physical and mechanical properties of PET fibers

| | |
|---|---|
| Density (g/cm$^3$) | 1.38 |
| Melt temperature (°C) | 250–260 |
| Tensile strength (cN/dtex) | 2.9–3.7 |
| Elongation at break (%) | 85–105 |
| Humidity absorption (%) | 0.4 |

study is to analyze the effect of the incorporation of short poly(ethylene-terephtalate) (PET) fibers on mechanical behavior and morphology of thermoplastic elastomers based on PP-EOC blends. It is our intention to get composites with balanced properties and to analyze their behavior/morphology relationship.

## 2. Experimental

### 2.1. Materials

Commercially available grades of polypropylene (iPP) (MFI: 2.9 at 190°C and 5 Kg, and density 0.905 g/cm$^3$), kindly supplied by Repsol Química, under the trade name of Isplen PP-050 and EOC (MFI: 1 dg/min and Mooney viscosity, ML 1 + 4 of 23 at 121°C) supplied by Du Pont-Dow Elastomers, under the trade name Engage 8450, and short PET fibers, furnished by Velutex-Flock, S.A. were used in this work. The physical characteristics of the fibers are listed in Table 1.

### 2.2. Sample preparation and testing procedures

The composites were processed by means of hot-rolls, at a temperature of 180°C, for 20 min. Once the polymers were melted, the appropriate percentage of fiber was added into the polymer blend. Immediately after the mixing, the material was finely cut into pellets, and then injection molded in a Margarit M50/125 injection molding machine to obtain standardized dog-bone specimens. The temperatures in the three zones of the injection molding machine were 210, 220 and 230°C, respectively. A mold temperature of 60°C, and an specific injection pressure of 750 Kg/cm$^2$ were used. The period of time for the packing and cooling stages were 30 and 20 s, respectively. Since all of the composite materials have been obtained following the same procedure it is assumed that the fiber orientation is the same in all samples.

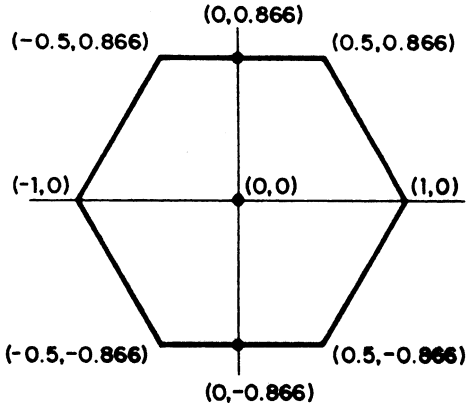


Fig. 1. Schematic of a design based on a Doehlert Uniform Net.

The tensile and flexural testing were performed at room temperature on an Instron mod. 4301 dynamometer, according to ASTM D-638 M and ASTM D-790 M, respectively. Tensile tests were carried out at a cross-head speed of 5 mm/min and 50 mm/min, for measuring the tensile modulus, and ultimate strength and elongation, respectively. Flexural tests were performed at a cross-head speed of 2 mm/min and with a span of 64 mm. The ratio of support span to specimen depth was 32:1. All the results are the average of at least five measurements.

Dynamic mechanical analysis (DMA) was carried out by means of stress–strain oscillation measurements using a Metravib dynamic mechanical thermoanalyzer (Mark 03 model). Tests were carried out in tension-compression mode, over a wide frequency range (5, 10, 25 and 50 Hz) and the temperature programs were run from −80°C to 125°C under a controlled sinusoidal strain, at a heating rate of 2°C/min, under a flow of nitrogen. An oscillating dynamic strain of 0.15% was used. The nominal dimensions of the specimen were $25 \times 10 \times 1$ mm$^3$ The viscoelastic properties, such as, the storage modulus ($E'$) and the mechanical loss factor, damping factor ($\tan \delta = E'/E''$), were recorded as a function of temperature and frequency.

The morphology of the samples was also analyzed using a JEOL T330A scanning electron microscope. Samples were cryogenically fractured in liquid nitrogen and sputtered with gold to be observed in the scanning electron microscopy (SEM).

### 2.3. Experimental design

In order to analyze the influence of matrix composition and fiber percentage on the tensile behavior of these composites, response surface methodology [18–20] was applied to the experimental results in a wide range of both variables and with a small number of experiences. In this case, the two variables method was used and the experiences were planned on the basis of an Uniform Net of Doehlert [21]. This methodology imposes to define the experimental domain to be explored, so PP percentage in the matrix varies from 0 to 100% and the fiber content in the composite ranges from 0 to 20%. A second-degree equation has been postulated for this experimental design. This type of design defines the minimum number of experimental combinations in the experimental domain to be explored to get the maximum information for adjusting the proposed model. Among the Doehlert design properties, one involves a uniform distribution: a set of point is uniformly distributed in the space following a rhombic lattice, which allows exploration of the whole experimental domain. A design of this type is

Table 2
Codified and uncodified values of the experimental combinations

| Experiment | Codified values | | Uncodified values | |
|---|---|---|---|---|
| | $x_1$ | $x_2$ | $X_1$ (%) | $X_2$ (%) |
| 1 | 1.0 | 0 | 100 | 10 |
| 2 | 0.5 | 0.866 | 75 | 18.66 |
| 3 | −0.5 | 0.866 | 25 | 18.66 |
| 4 | −1.0 | 0 | 0 | 10 |
| 5 | −0.5 | −0.866 | 25 | 1.34 |
| 6 | 0.5 | −0.866 | 75 | 1.34 |
| 7 | 0 | 0.866 | 50 | 18.66 |
| 8 | 0 | −0.866 | 50 | 1.34 |
| 9 | 0 | 0 | 50 | 10 |
| 10 | 0 | 0 | 50 | 10 |
| 11 | 0 | 0 | 50 | 10 |

show in Fig. 1. Three equal experiments are carried out in the center of the design to know the experimental error. In our case, two more experimental points (points 7 and 8) have been added in order to prepare three composite materials with the same matrix composition at three different fiber contents.

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 physical values (or uncodified values) of the variables $x_1$ and $x_2$ were obtained from Eq. (1):

$$x = \frac{x_A + x_B}{2} + X\frac{x_A - x_B}{2} \quad (1)$$

where $X$ is the codified value and $x$ is the physical value of the variables. $A$ and $B$ refer to the high and low levels of the variables, in our case, 100 and 0 for the PP percentage in the matrix composition, and 20 and 0 for the PET fiber percentage in the composite. The composition of the samples used in this study, as well as the codified values of the variables, are compiled in Table 2.

## 3. Results and discussion

In order to evaluate the effect of the matrix composition and the percentage of fiber on the mechanical properties of the composite, the Doehlert Uniform Net method has been applied. From experimental data, the equations of the response surfaces were obtained by means of multivariable technique analysis, following the general equation:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_{11}X_1^2 + b_{22}X_2^2 + b_{12}X_1X_2 \quad (2)$$

Where $Y$ is the value of the characteristic or property of the composite, $X_1$ and $X_2$ are, in our case, the code level of PP percentage in the matrix and PET fiber content in the composite, respectively. As it is expected, a different equation was obtained for each property studied, as shown in Table 3. It may be pointed out, that in general, high correlation factors between experimental values and those calculated from theoretical equations have been observed. Only in the case of the composite tensile strength, the correlation coefficient of the corresponding theoretical equation is lower than 0.9, which indicates that there is a more sensible difference between the obtained experimental values and those calculated from the theoretical equation. The graphics obtained from these equations are shown in Figs. 2–6, and in each curve the level or value of the corresponding property is indicated. From these graphics, pairs of values of the experimental variables can be selected to obtain a composite with specific mechanical properties and also, the relative behavior of different materials can be easily compared.

### 3.1. Mechanical properties

Tensile modulus and strength and elongation at fracture as well as the flexural modulus and maximum strength were determined and the results summarized in Table 4. From these experimental data, and through a multiple regression analysis, the equations of the response surfaces were calculated and then the iso-level curves were graphically represented, as shown in Figs. 2–6. The tensile Young's modulus as a function of matrix composition and fiber percentage in the composite is graphically represented in Fig. 2. The individual and combined effect of both variables on the composites stiffness can be analyzed. It can be observed that the incorporation of PET fibers give rise to a more rigid material, showing a gradual increase of the modulus value as fiber content in the composite is increased. This reinforcing effect depends on the matrix composition. In fact, increments of about 25% are obtained at low copolymer percentages in the matrix, while a 50% increase is observed at higher elastomer content. These results seem to suggest that the PET fiber used in this work behave as effective reinforcing agent of these systems based on PP-EOC blends. It is also important to note, that this effect is more sensible at

Table 3
Response surface equations of the composites

| Property | Equation of the response surface | Correlation coefficient |
|---|---|---|
| Tensile modulus | $Y = 529.39 + 418.55X_1 + 70.0807X_2 + 46.12X_1X_1$ | 0.99 |
| Tensile strength | $Y = 19.38 + 4.17X_1 - 4.57X_2 + 9.66X_1X_1$ | 0.89 |
| Elongation at break | $Y = 843.17 - 327.47X_1 - 370.17X_2$ | 0.93 |
| Flexural modulus | $Y = 686.49 + 584.84X_1 + 166.12X_2$ | 0.99 |
| Flexural strength | $Y = 22.73 + 17.60X_1 + 3.80X_2 + 2.19X_1X_1$ | 0.99 |

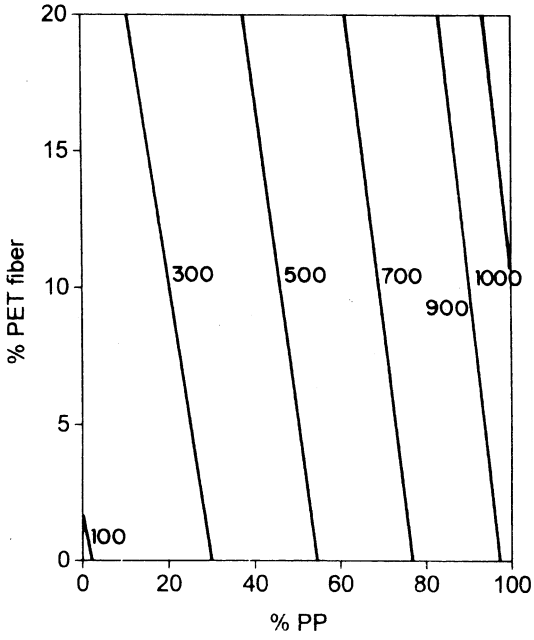


Fig. 2. Tensile modulus as a function of PP percentage in the matrix and fiber content in the composite.

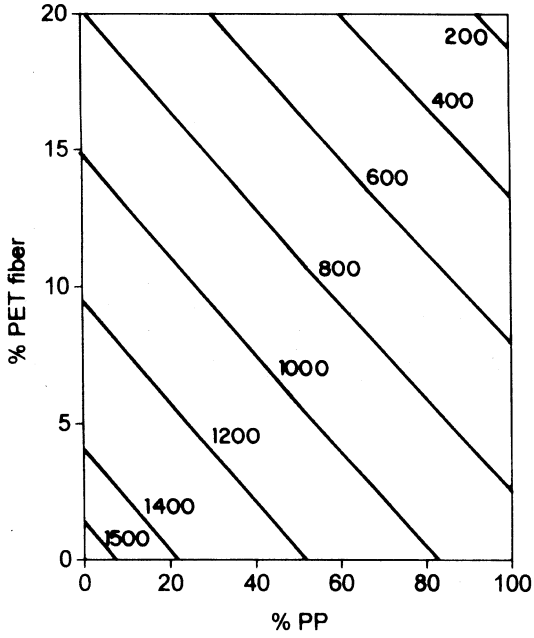


Fig. 4. Elongation at break as a function of PP percentage in the matrix and fiber content in the composite.

higher elastomer contents in the thermoplastic which can be attributed to a better interaction or adhesion between the fibers and the elastomeric phase. However, the Young's modulus of the material depends more on matrix composition than on fiber percentage in the composite. As could be expected, a gradual increase of the elastomeric behavior of the blends is achieved as elastomer content is increased, thus, a dramatically decrease in the Young's modulus (about 150%) is obtained, when the blend composition goes from (75-25) PP-EOC to (25-75) PP-EOC. The combined effect of both variables on this characteristic is very noticeable.

The iso-level curves of the tensile maximum strength, as a function of both matrix composition and fiber content in the composite, are graphically represented in Fig. 3. The addition of the fibers gives rise to a decrease in the maximum

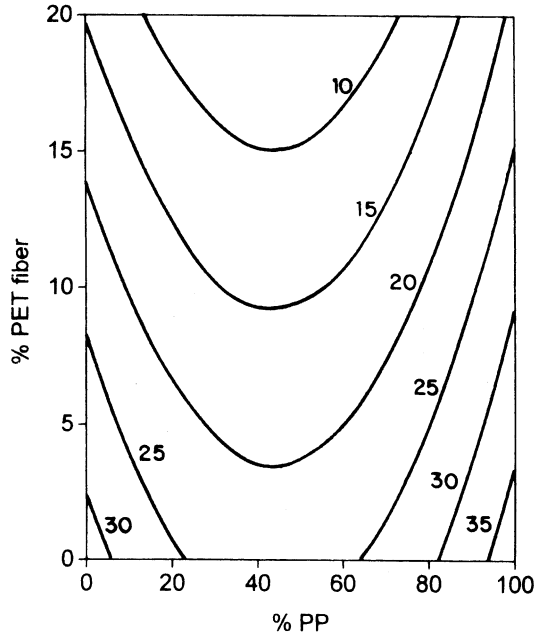


Fig. 3. Iso-level curves of the tensile maximum strength, as a function of matrix composition and fiber content in the composite.

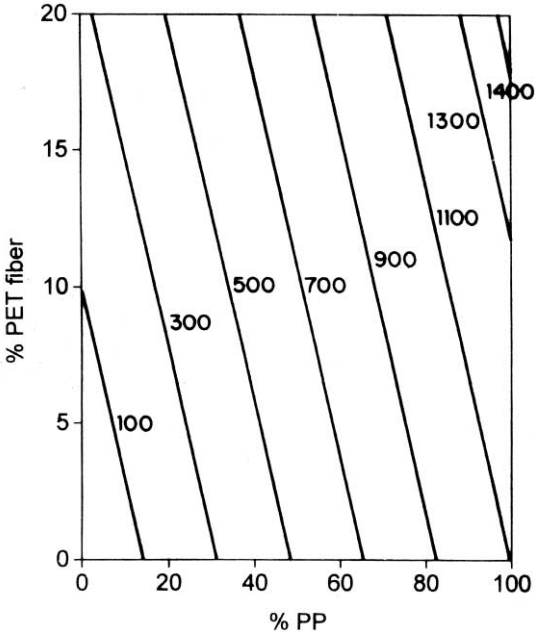


Fig. 5. Iso-level curves of the flexural modulus as a function of PP percentage in the matrix and fiber content in the composite.

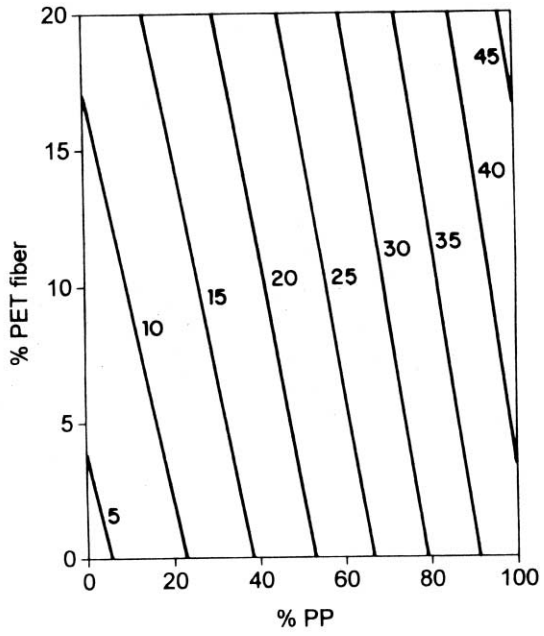


Fig. 6. Maximum flexural strength as a function of PP percentage in the matrix and fiber content in the composite.

strength of the composite, independently of matrix composition. However, the stiffness of the material strongly depends on matrix composition. So, in general, the strength of PP rich matrices is higher than EOC rich matrices, and an inflexion in the response surface curves is observed at PP contents in the matrix about 45–50% which suggest that a phase inversion is taken place.

The main difference between the modulus and the strength during the test is the deformation produced on the material. In the first case, the modulus is measured as the slope of the strain–stress curve at very low elongation (less than 1%), whereas the strength of a material to break, implies an elongation above the elastic limit where the material shows a viscoelastic behavior, and defects in their structure may appear due to stress concentration in the fiber ends, and overlapping of fibers. Moreover, the strength of a material depends on both matrix and reinforcement characteristics, as well as, on the interaction at the fiber/matrix interface. According to the obtained results, it can be assumed that, in general, at high elongation, and when there is not a good adhesion at the interface, the fibers give rise to a decrease in composite strength.

The variation of the elongation at break as a function of the material composition is graphically represented in Fig. 4. As it was expected, a gradual increase in the elastomeric behavior of the blends is obtained as elastomer content is increased, thus, the elongation at break of the material with high EOC percentages in the matrix composition, is sensibly higher. In addition, the incorporation of the fibers gives rise to a more rigid and stable material, showing a marked decrease in the material elongation, independently of the matrix composition.

The iso-level curves of the flexural modulus and maximum strength are graphically represented in Figs. 5 and 6, respectively. Flexural modulus presents a similar behavior to that of the tensile modulus, and thus, a sensible increase is observed, as both PP and PET fiber contents in the composite increase. As it was previously mentioned, the reinforcing effect of PET fibers is more evident on matrices with high engage percentages (>50%), showing an increment in the modulus of about 90%. These results are in agreement with those previously observed in flexural testing, where a sensible increase in the material stiffness was obtained as both PP and PET fiber percentages in the matrix and in the composite, respectively, were increased. In addition, the reinforcing effect of the fibers is more evident at higher elastomer content in the matrix.

On the basis of the mechanical results, it can be concluded that the PET fibers behave as an effective reinforcement for these polymers, being this effect more evident at higher percentages of elastomer in the composite.

### 3.2. Dynamic mechanical analysis

In order to analyze the effect of fiber content on the behavior of the composites, the dynamic mechanical properties

Table 4
Mechanical properties of the composites

| Experiment | Tensile behavior | | | Flexural behavior | |
|---|---|---|---|---|---|
| | Modulus (Mpa) | Maximum strength (Mpa) | Deform. break (%) | Modulus (Mpa) | Maximum strength (Mpa) |
| 1 | 1053 | 29.3 | 16.2 | 1454 | 45.5 |
| 2 | 836 | 21.9 | 12.6 | 1269 | 37.6 |
| 3 | 379 | 14.4 | 845 | 497 | 17.4 |
| 4 | 130 | 20.9 | 1109 | 155 | 7.1 |
| 5 | 257 | 24.6 | 1081 | 265 | 11.4 |
| 6 | 662 | 24.5 | 908 | 846 | 29.1 |
| 7 | 533 | 15.6 | 230 | 810 | 25.5 |
| 8 | 452 | 27.3 | 1105 | 544 | 19.3 |
| 9 | 496 | 20.9 | 975 | 684 | 22.5 |
| 10 | 527 | 18.9 | 814 | 710 | 23.8 |
| 11 | 575 | 17.5 | 772 | 674 | 22.2 |

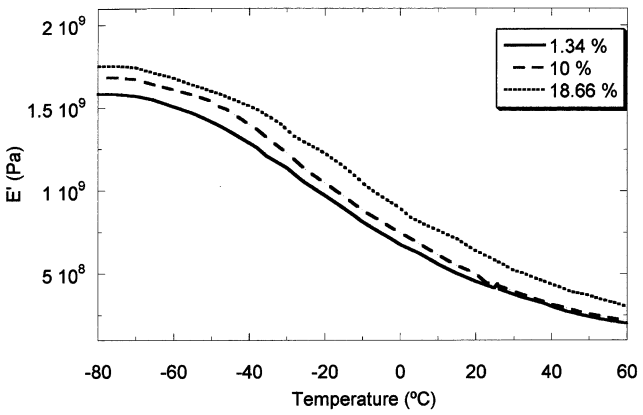


Fig. 7. Storage modulus of samples 7, 8, and 9, as a function of temperature.

of samples 7, 8 and 9 (PP/EOC ratio = 50/50, fiber content: 18.66, 1.34, and 10%, respectively) were measured under a sinusoidal controlled strain and the resultant displacement was measured. The variation of the storage modulus ($E'$) as a function of the temperature at 5 Hz, is reported in Fig. 7. From this Figure, can be deduced that the incorporation of the PET fibers gives rise to a more rigid and tough material, which is reflected in a considerable increment of the modulus value. This increase is more significative as the fiber percentage in the composite increases. According to these results, it can be assumed that the PET fibers behave as effective reinforcement of these systems.

### *3.3. Morphological study*

Fracture surfaces of compression molded specimens 75/25 and 25/75 PP/octhene and 10% fiber content were observed through SEM and the micrographs are reported in Figs. 8 and 9, respectively. There are hardly any voids on the fracture surfaces which predominantly show fiber fracture rather than fiber pull-out. This indicates that a good adhesion at the fiber-matrix interface exists.

The observed morphology is in agreement with the mechanical properties of the studied composites and permit to conclude that the PET fibers used in this work act as reinforcing agent of these polymers.

## 4. Conclusions

The properties of ternary composites based on iPP and EOC blends reinforced with short PET fibers have been studied in this work. In order to analyze the combined effect of the matrix composition and the PET fiber in the composite on the mechanical properties of these systems, an experimental design based on a Doehlert Uniform Net has been applied.

The mechanical properties show a strong dependence on matrix composition. So, as rubber content in the thermoplastic is increased, the material becomes more elastomeric, giving rise to a marked decrease of the modulus and strength of the material. On the other hand, the incorporation of the PET fibers generates a more rigid and stable material, with better mechanical properties. These results strongly support the assumption that the PET fibers behave as a reinforcing agent for PP/octhene blends. Furthermore, it is important to note that this reinforcing effect is more significative at higher elastomer content in the thermoplastic.

These results are in concordance with those obtained by dynamic mechanical analysis, where an increment of the storage modulus was observed in presence of the PET fibers. In addition, the modulus gradually increases as increasing the fiber percentage in the composite.

Moreover, the microscopy study confirmed the results of the composite behavior, where a good adhesion at the fiber-matrix interface was observed. So, it was possible to

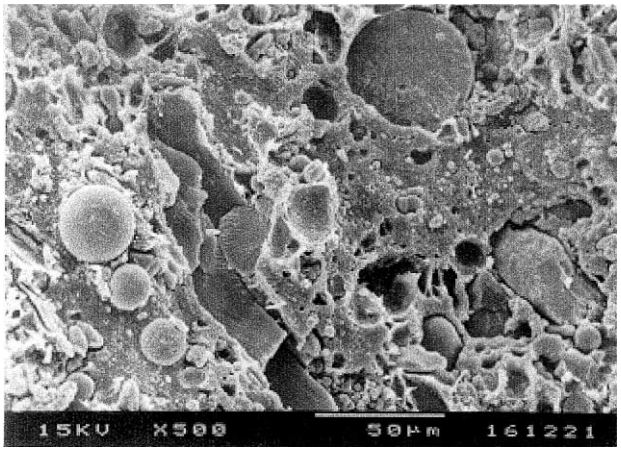


Fig. 8. Fracture surface of a composite based on PP/copolymer (75/25) blend matrix and 10% PET fiber.

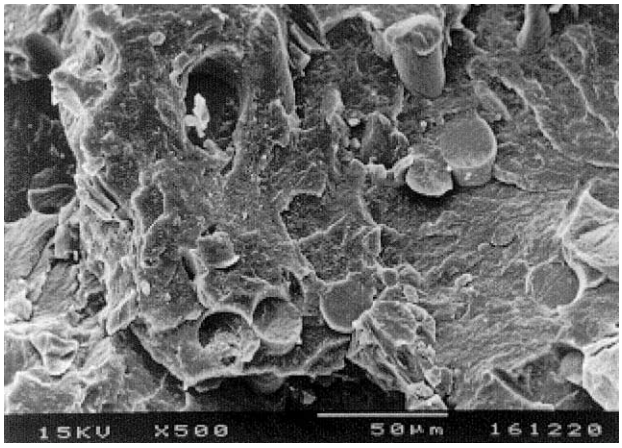


Fig. 9. Fracture surface of a composite based on PP/copolymer (25/75) blend matrix and 10% PET fiber.

correlate a good adhesion at the fiber-matrix interface with an improve of the composites properties.

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