Effect of the Elastomer Type on the Microstructure and Mechanical Properties of Polypropylene

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ABSTRACT: In this study, the effects of the elastomer type—ethylene–propylene–diene monomer (EPDM), three kinds of ethylene vinyl acetate (EVA 9, EVA 18, and EVA 28, where the number is the vinyl acetate concentration), and styrene–butadiene–styrene—and content on the microstructure and mechanical and thermal properties of isotactic polypropylene (*i*-PP) blends were investigated. Five different elastomer concentrations (3, 6, 9, 12, and 15 wt %) were added to *i*-PP to produce polypropylene/elastomer blends. The yield and tensile strengths, elastic modulus, impact strength, hardness, melt flow index (MFI), and structural properties of the blends were investigated. The tensile and yield strengths, elastic modulus, and hardness decreased gradually, whereas the impact strength and MFI increased as the elastomer content increased. As a result, with respect to the impact strength, the most effective elastomers were EPDM with 15 wt % and EVA 28 with 15 wt % for higher impact strength values. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 98: 1445–1450, 2005

Key words: compounding; elastomers; mechanical properties; poly(propylene) (PP)

INTRODUCTION

Polymer blending is a simple and efficient method for designing and controlling the performance of polymeric materials with easily available polymers. The procedure makes it possible to develop a new polymeric material of synergetic performance of each polymer, to reduce the cost of engineering polymers by dilution with lower cost materials, or to enhance the recycling of used plastics. These advantages of polymer blending for performance, economy, and ecology have accelerated research and development activities in the field of polymer blends and alloying in both academia and industry.[1–5]

Semicrystalline and multipurpose properties such as good mechanical and thermal properties, corrosion resistance, low cost, easy processing, easy design, and low density make polypropylene attractive for industrial applications. These properties depend on the compositions, crystalline structure, contents of amorphous and crystalline phases, crystal size, molding conditions, chemical compositions of the additives, filler content, and product technique. For industrial applications as an engineering polymer, however, its toughness due to its high transition temperature and, in particular, its notch toughness at low service temperatures are not sufficient and limit its applications. For this reason, price and performance characteristics and low-service-temperature characteristics can be considerably improved via blending with elastomers, particularly ethylene–propylene copolymer, ethylene–propylene–diene monomer (EPDM), and ethylene vinyl acetate (EVA). The toughness characteristics are improved by the blending of an elastomer with polymeric structures, and this causes a decrease in the elastic modulus and tensile and yield strengths and an increase in the elongation and impact strength properties. For this reason, multipurpose fillers and additives can be used to overcome this problem by ensuring better toughness and dimensional stability. Although improvements take place in these properties of polypropylene, no negative effect occurs in its processing with methods such as injection and extrusion molding. Polypropylene and elastomer compounds are the fastest growing general-purpose materials on the market. These compounds have a wide range of melt flow index (MFI) values and good adhesion to a variety of materials, and the price is low and permits the design of tailored materials.[6–24]

Polypropylene and elastomer [EPDM, EVA, and styrene–butadiene–styrene (SBS)] blend materials and composites are well suited for many indoor and outdoor applications because of their ozone and weathering resistance and good mechanical properties. This may be inherent to the hydrocarbon nature, almost saturated backbone, and mechanical properties of elastomers.[10–15]

In the literature, there are some published studies investigating the properties with respect to the effect of elastomer addition to polypropylene.[10–24] How-

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Resin	Trade name	Supplier	MFI (g/10 min)	Mooney viscosity	Density (d) (g/cm^3)	T_m (°C)	Shape
i-PP	Petoplen MH 418	Petkim	4–6[a]	—	0.925	166	Pellets
EVA 9	ML 30	Greenflex	2.5[b]	—	0.950	95	Pellets
EVA 18	MH 40	Greenflex	1.8[b]	—	0.950	87	Pellets
EVA 28	ML 60	Evatane	2.5[b]	—	0.950	75	Pellets
EPDM	IP 4770	Nordel	—	70[d]	—	—	Pellets
SBS	Dynasol Calprene	MTI	7.5[c]	—	—	—	Pellets
Stabilizer	Irganox B 225	Ciba	—	—	530–630 g/L	—	Powder

[a] 230°C, 2.16 kg/10 min.
[b] 190°C, 2.16 kg/10 min.
[c] 190°C, 5 kg/10 min.
[d] Typical Mooney viscosity, $ML_{1 + 4}$ at 125°C

ever, it is difficult to see the effects of both the elastomer type and content on the mechanical, microstructural, and thermal properties in these studies. Therefore, in this study, the effects of the elastomer type and content on the mechanical, thermal, and microstructural properties of isotactic polypropylene (i-PP) blends have been investigated. To this end, i-PP/elastomer blends containing 0, 3, 6, 9, 12, or 15 wt % elastomer were produced, and the mechanical and microstructural properties of the blends were determined.

EXPERIMENTAL

Materials

The polypropylene and different types of elastomers (EPDM, EVA, and SBS) used in this study were all commercial products, and the characteristic properties are listed in Table I. The i-PP used as a matrix phase in this study was Petoplen MH 418 (pellets) from Petkim (Petkim, İzmir, Turkey). Different elastomers such as EPDM (Nordel), three kinds of EVA with vinyl acetate (VA) concentrations of 9 wt % (EVA 9; Greenflex, Italy), 18 wt % (EVA 18; Greenflex, Italy), and 28 wt % (EVA 28; Evatane, Atofina, Germany), and SBS (Dynasol Calprene) were used. Irganox B 225, supplied by Ciba (Basel, Switzerland), was added as a synergistic processing and long-term thermal stabilizer to the polypropylene blends at a concentration of 0.3 wt % during the melt processing.

Compounding process and sample preparation

Extrusion

The materials were dried at 75°C for at least 1 h in a Binder ED 115 oven (Binder, Tuttlingen, Germany) before the compounding process. For all the types of elastomers, five different concentrations (3, 6, 9, 12, and 15 wt %), with an additive of 0.3 wt % Irganox B 225, were added to polypropylene. i-PP/elastomer blends were carried out with a Microsan MTV single-screw extruder (screw diameter = 30 mm, length-to-diameter ratio = 25; Microsan, İzmit, Turkey). The temperature profiles (210–225°C) were used from the feed zone to the die. The screw speed and pressure were 40 rpm and 6 bars, respectively. The extrudate was frozen inline in a water bath (~20°C), pelletized, and dried in an oven at 100°C for 2 h.

Injection molding

Tensile and impact test specimens (ISO 527.2 and 180) were prepared by injection molding with a 70-ton machine (Yonca, Istanbul, Turkey). Before the molding, the pellets were dried at 100°C for 1 h. A temperature range of 210–230°C, a mold temperature of 40°C, an injection time of 10 s, an injection pressure of 500 bars, and a dwell time of 10 s were used.

Measurement procedures

Mechanical testing

The tensile tests of the blends were measured on a Zwick Z010 tensile test machine (Zwick, Ulm-Einsingen, Germany) according to ISO 527.2 at a crosshead speed of 50 mm/min at room temperature. A computer was connected to the Zwick load cell, and a data acquisition program read the force recorded by the load cell. At least five specimens were tested for each blend, and the average value was calculated. The impact strengths of the blends were carried out on a Zwick impact test machine according to ISO 180 at room temperature with a notch radius of 1 mm, a notch angle of 45°, and a depth of 2.0 mm. At least seven specimens were tested for each blend, and the average value was calculated. Before the mechanical tests, all specimens were kept at room temperature for at least 72 h. The hardness tests of the blends were measured with a Zwick hardness tester according to ISO 868 at room temperature. At least four specimens
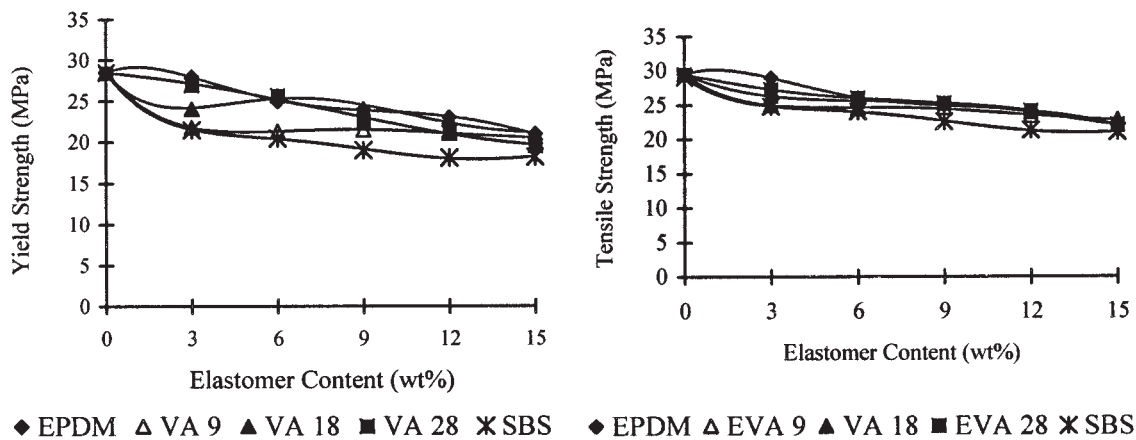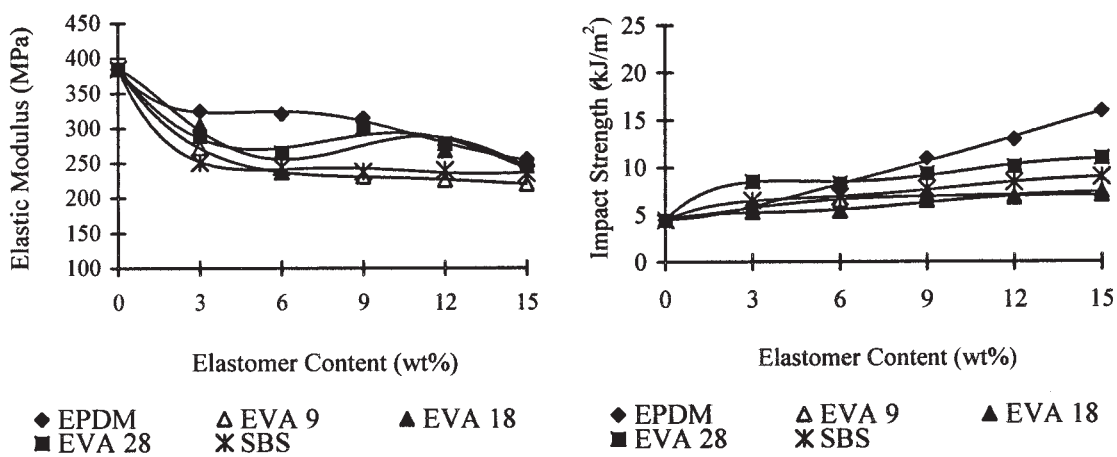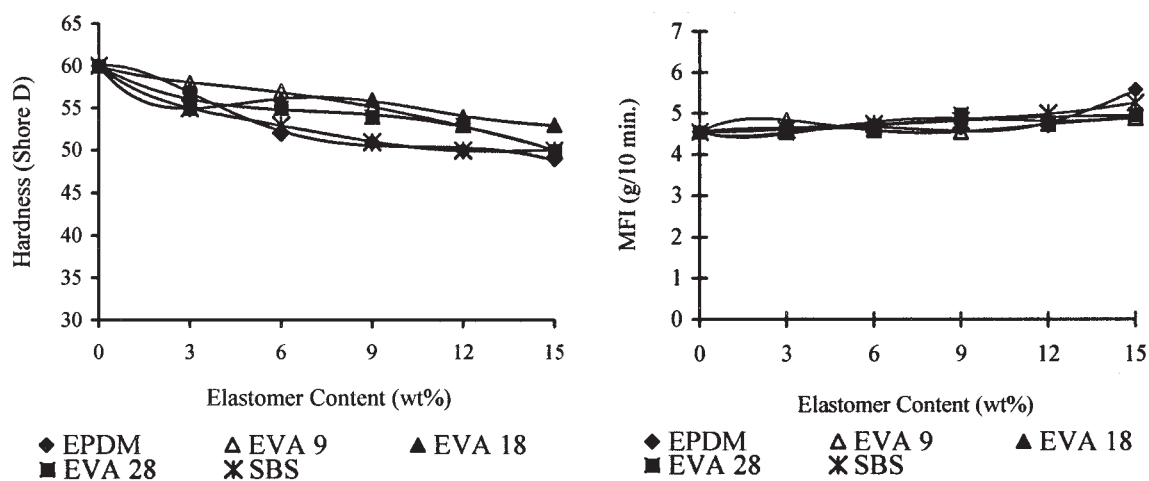
Figure 1 (a–e) Mechanical properties (yield strength, tensile strength, elastic modulus, impact strength, and hardness) and (f) thermal properties (MFI) of polypropylene/elastomer blends.
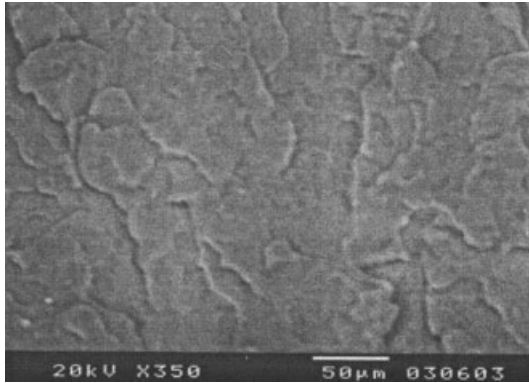
Figure 2 SEM micrograph revealing the appearance of the impact fracture surface of *i*-PP.

were tested for each blend, and the average hardness value was calculated.

Thermal testing

MFIs of *i*-PP/elastomer blends were determined on a Zwick 4100 MFI test instrument according to ISO 1133. The MFI values were calculated as averages over five specimens for each composition of the blend.

Microstructural study

The microstructures of *i*-PP/elastomer blends were studied on the fracture surfaces with the aid of a JEOL JSM 5410LV scanning electron microscope (JEOL, Tokyo, Japan). Before the scanning electron microscopy (SEM) investigation, the samples were coated with gold to a thickness of 25 Å for conductivity.

RESULTS AND DISCUSSION

Mechanical properties

The tensile and yield strengths, elastic modulus, impact strength, and hardness of the polypropylene/

elastomer blends are shown in Figure 1(a–e), respectively. The yield and tensile strengths [Fig. 1(a,b)] tend to decrease in direct proportion to the increase in the elastomer concentration. This is consistent with previous work.[11,15,20,21] The decrease in the yield and tensile strengths can be related to the structures of the elastomers. The elastomer structure increases the mobility of the polypropylene chains and causes the tensile properties to decrease.[11,13] The form of the material to be used as an elastomer material is the most important for the blending process. If the elastomer has much more flexibility, the yield and tensile strengths of the blend decrease.[11]

As can be seen in Figure 1(a,b), the elastomer type is also effective for the yield and tensile strength. SBS exhibits less tensile and yield strength than EPDM and EVA. Among the EVA types, EVA 28 provides higher yield and tensile strength values. This is also directly related to both the structure of the elastomer and the interface characteristics between *i*-PP and the elastomer.[10,11]

Figure 1(c) shows the effect of the elastomer content on the elastic modulus of polypropylene/elastomer blends. The elastic modulus decreases as the elastomer content increases, and this result is in good agreement with the literature.[13,17,20,21,23] A significant decrease can be observed with a 3 wt % concentration, in comparison with the decrease between 9 and 15 wt %. The lowest values are observed with the addition of 15 wt % elastomer. The reason for the decrease in the elastic modulus of the blends can be related to the structure of the elastomer.[11,15] The elastomer type has an effect on the elastic modulus. A significant decrease in the elastic modulus can be observed for EVA 9 and SBS. The reason for this decrease may be related to both the structure of the elastomer and weak adhesion between the elastomer and polypropylene matrix.[11]

Figure 1(d) shows the effect of the elastomer content on the impact strength values for the polypropylene blends. The impact strength values change signifi-
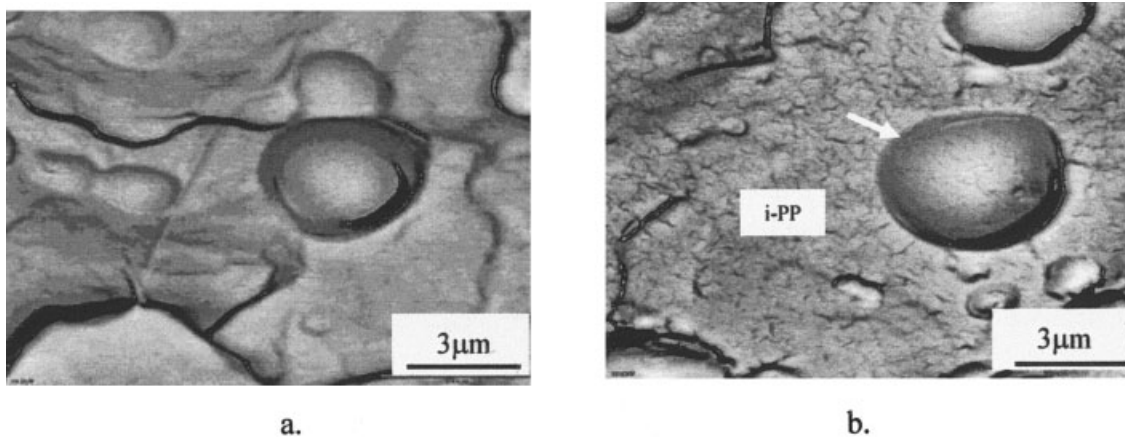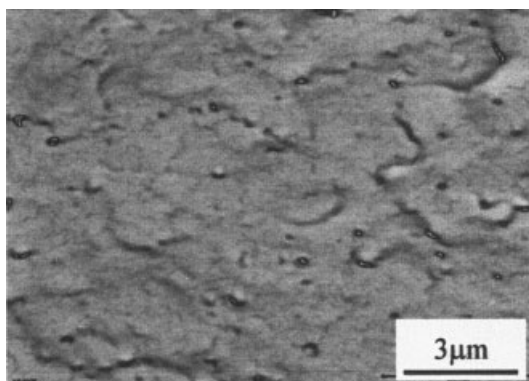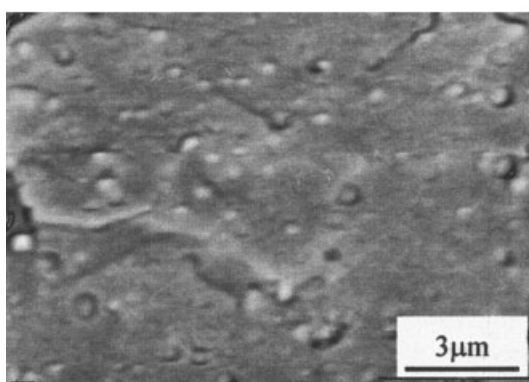

a.

b.

Figure 3 SEM micrographs revealing the appearance of the impact fracture surfaces of *i*-PP/EPDM blends: (a) 6 and (b) 9 wt % (the arrow shows EPDM).

a.

b.

Figure 4 SEM micrographs revealing the appearance of the impact fracture surfaces of *i*-PP/EVA 9 blends: (a) 9 and (b) 15 wt %.

cantly with the elastomer type and content. In general, the impact strength increases as the elastomer concentration increases, and this result is consistent with the literature.[11,12,15,16–21] However, the elastomer type has an effect on the increased impact strength. Among the elastomers, the most effective type is EPDM, and this is followed by EVA 28 and SBS. As the elastomer contents are considered, we can see that 3 wt % and higher values have an effect on higher impact strength values, especially for EPDM. It is well known that the structure without high rigidity exhibits higher impact strength. In general, the improvement in the impact properties may be due to the structure of the elastomer and improved interfacial adhesion of the elastomer to *i*-PP.[10,11] EVA 28 is a better impact modifier than EVA 9 and EVA 18. This result can be directly related to the greater flexibility of VA.[13]

Figure 1(e) shows the hardness values of the polypropylene/elastomer blends. The hardness values of the blends decrease with an increasing content of the elastomer. The decrease in the hardness with an increase in the elastomer may be related to the structure of the elastomer, which provides plastic deformation.[11,15,16–21]
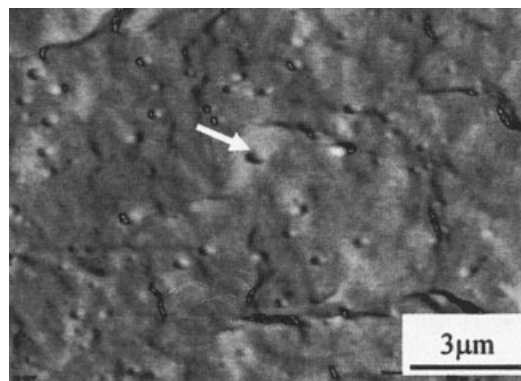

Figure 5 SEM micrograph revealing the appearance of the impact fracture surface of an *i*-PP/EVA 18 blend (9 wt %; the arrow shows EVA).

Thermal properties

Figure 1(f) shows the changes in the MFIs of *i*-PP/elastomer blends. The MFI values slightly increase in direct proportion to the increase in the amount of the elastomer. A maximum increase in MFI can be seen for the elastomer concentration of 15 wt %. The reason for the increase in MFI with the increase in the elastomer concentration may be related to its molecular mobility.[11]

Microstructural behavior

The SEM examination results of the fracture surfaces obtained from the Izod impact tests are shown in Figure 2–7. Figures 2 and 3(a,b) show the fracture surfaces of polypropylene (without elastomer) and polypropylene/EPDM blends, respectively.

Figure 3(a,b) shows good adhesion between *i*-PP and EPDM. EPDM contributes to the strong adhesion and homogeneous structure, resulting in good impact properties.[14,20,23] A nonhomogeneous distribution of elastomers in the matrix also causes properties to be weak. If the elastomers are distributed homogeneously in the matrix (this is quite hard most of the
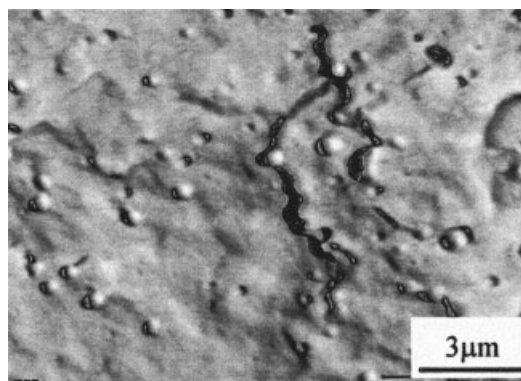

Figure 6 SEM micrograph revealing the appearance of the impact fracture surface of an *i*-PP/EVA 28 blend (15 wt %).
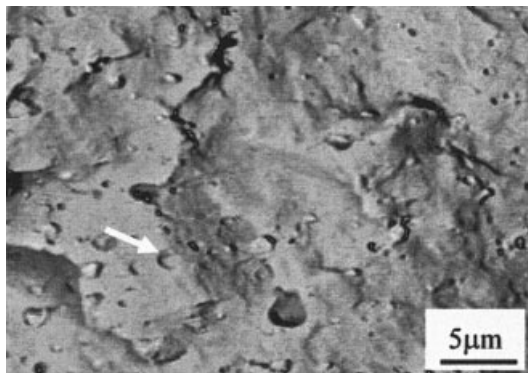
Figure 7 SEM micrograph revealing the appearance of the impact fracture surface of an *i*-PP/SBS blend (12 wt %; the arrow shows SBS).

time) and a strong interaction is sufficiently improved, then the blend is made to transfer higher loads. Therefore, in this study, the significant increases in the impact properties for EPDM can be related to both the structure of EPDM and homogeneous distribution with strong adhesion, and this result is in good agreement with the literature.[10,11,17]

Figures 4–6 show SEM micrographs of Izod impact fracture surfaces of specimens with various EVA contents. EVA shows a good distribution in the matrix of polypropylene. However, the increase in the amount of EVA makes the distribution within the matrix easier, and thus strong adhesion between *i*-PP and EVA exists. This also causes the impact strength of the blend to increase. Therefore, the impact strength increases as the EVA content increases. Among the EVA types, EVA 28 with a concentration of 15 wt % shows higher impact strength values. This increase in the impact strength can be related to both the flexible structure and homogeneous distribution with good adhesion (Fig. 6). As a result, the impact strength of *i*-PP/EVA increases as the content of EVA increases in the blend, and the most effective type for the impact strength is EVA 28. Figure 7 shows an SEM micrograph of the Izod impact fracture surface of a blend containing 12 wt % SBS. There is a good distribution in the matrix of polypropylene.

From the SEM examination results of the fracture surfaces, it can be concluded that all the elastomer types show a good distribution in the matrix of *i*-PP. However, considering only the distribution of the elastomers in the matrix, we find it difficult to explain the reason that EPDM produces higher toughness values than EVA 28 and SBS, so this can be explained by both adhesion between the elastomer and *i*-PP and the structure of the elastomer.[10] Therefore, it can be generalized that higher toughness values obtained for

EPDM are directly related to both good adhesion between EPDM and *i*-PP and the structure of EPDM, which is much more flexible than EVA and SBS.[11,17]

CONCLUSIONS

In this study, the effects of the elastomer type and concentration on the mechanical, microstructural, and thermal properties of polypropylene blends were investigated. The following results can be drawn from the experimental results:

1. The yield and tensile strengths, elastic modulus, and hardness decrease gradually as the elastomer content is increased.
2. The impact strength increases as the elastomer content increases.
3. EPDM is the most effective elastomer for higher toughness values and is followed by EVA 28 and SBS.
4. As the content of VA in EVA increases, the impact strength increases.
5. MFI increases slightly as the elastomer content increases.

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