Analysis of the Effects of the Polymerization Route of Ethylene–Propylene–Diene Rubbers (EPDM) on the Properties of Polypropylene–EPDM Blends

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ABSTRACT: A comparative study of two ethylene–propylene–diene rubbers (EPDM) polymerized by both conventional (Ziegler–Natta catalysts) and new techniques (metallocene catalysts) is presented. For this purpose, thermoplastic elastomers based on isotactic polypropylene (iPP) and EPDM blends at different percentages were prepared and their properties examined. In particular, the processing behavior and mechanical properties are reported. So, the flow properties analyzed by torque value, melt index, and rheological study reveal that the blends containing EPDM synthesized by metallocene catalyst present a smaller viscosity, thus offering better processing behavior. On the other hand, the mechanical properties show that metallocene EPDM rubbers give rise to more elastic materials with a higher deformation at break and resilience as well as a lower compression set. Moreover, the effectiveness of these innovative EPDM rubbers as impact modifiers for PP is demonstrated. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 85: 25–37, 2002

Key words: EPDM; metallocene; catalyst; elastomer thermoplastic

INTRODUCTION

In recent years the production of plastic–elastomeric rubber blends has dramatically increased, given their well-balanced physical and mechanical properties and their easy processability and relatively low costs. In addition, polyolefin blends attract additional interest because of the possibility of recycling their wastes, thus avoiding the complex and expensive processes of separation of the different components. These blends, commonly referred to as *thermoplastic elastomeric olefins* (TPOs), are a special class of thermoplastic elastomers (TPEs) that combine the good processing characteristics of thermoplastics at high temperatures[1–3] with the physical properties of conventional elastomers at service temperatures.[4,5] TPOs most utilized in industry are blends of isotactic polypropylene (iPP) and ethylene–propylene–diene rubber (EPDM). In fact, because of its high impact strength over a wide temperature range, EPDM is considered one of the most effective impact modifiers for PP.[6–11] The elastomeric phase is added to the PP with the main objective to improve its flexibility and properties at low temperatures.

In more recent years, the development of a new class of catalysts based on metallocene chemistry

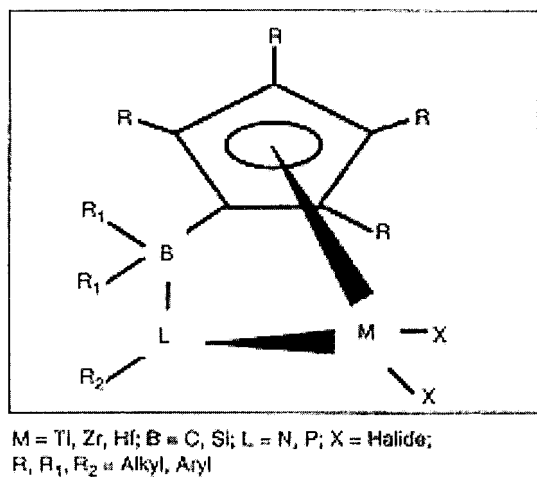M = Ti, Zr, Hf; B = C, Si; L = N, P; X = Halide;
R, R₁, R₂ = Alkyl, Aryl

Figure 1 Molecular structure of the constrained geometry catalyst.

has made a marked impact in the polyolefins industry.[12,13] These new catalysts, characterized for a specific constrained geometry catalyst system [single-site catalyst (SSC), the chemical structure of which is illustrated in Fig. 1], allow a more homogeneous distribution of the molecular structure and a better control of the concentration of monomer incorporated. Furthermore, they present a higher efficiency (higher relationship g product/g catalyst) compared to that of conventional heterogeneous Ziegler–Natta catalysts [multisite catalyst (MSC)]. Thus, the quantity of catalyst added to the final product is lower with metallocenes, and a washing step is not required after the polymerization has been completed, leading to polymers with lower residual catalyst than that of those obtained after the washing step when traditional catalysts are used. Moreover, the level of the metallic component has a direct effect on the color of the final material. Thus, the vanadium residue of the Ziegler–Natta catalyst often causes a yellowish tint to the polymer; however, using the constrained geometry catalyst, the quantity of metal residue (usually titanium) is lower and, as a result, the polymer has a much whiter and cleaner appearance.[14] Another advantage offered by this new process technology is given by the high reactivity of the catalysts, which allows the polymerization of many monomers that cannot be made using standard catalysts.

Initially, the activity of these catalysts was focused on semicrystalline thermoplastic materials, particularly polyethylene and polypropylene, but later their ability to produce copolymers and thermoplastic elastomers was demonstrated, allowing a significant achievement in the rubber industry. Thus, Du Pont Dow Elastomers (Wilmington, DE) has commercialized the first EPDM based on the proprietary Dow's INSITE[15] catalysts and process technology under the trade name Nordel IP rubbers. The INSITE process uses a combination of a constrained geometry catalyst system and innovative process technology to produce state-of-the-art polymers. The INSITE catalyst is different from other metallocene-type catalysts because of its molecular geometry, which forces a high exposure of the active metal site.

A comparative study of EPDM rubbers polymerized using traditional and new techniques was carried out by Reuter and Streit.[16] The authors revealed that a higher crosslinking density was obtained with the new polymers, which affects the suitability of these materials for use in seals.

Thus, the main goal of this study is the evaluation of the effectiveness of new EPDMs, based on the INSITE catalyst and process technology, as impact modifiers for polypropylene and to evaluate the possibility of replacing conventional EPDM rubbers for these applications. With this guiding purpose, thermoplastic elastomers based on PP–EPDM blends at different percentages, using two EPDMs rubbers synthesized by traditional and new techniques, were prepared and their properties analyzed.

EXPERIMENTAL

Materials and Preparation of Blends

Commercially available grades of polypropylene and both ethylene–propylene–diene terpolymer rubbers with 5-ethylidene–2-norborene (ENB) as termonomer were used in the present work. The materials specifications are listed in Table I.

Melt-blended specimens of these homopolymers with various compositions were processed in a Haake Rheomix 90 internal mixer (Haake, Germany), equipped with high shear roller-type rotors. The temperature of the mixing chamber was set at 170°C and the blending time was 10 min. The rotor rate was set at 60 rpm. Immediately after the completion of mixing, the materials were

Table I   Physical and Mechanical Characteristics of PP and EPDMs

Material	IPP	EPDM (Ziegler–Natta)	EPDM (Metallocenes)
Manufacturer	Montell (Italy)	Bayer (Germany)	Du Pont
Designation	C 30G	Buna EP T 6470P	Nordel IP NDR4725P
Density (g/cm^3)	0.905	0.86	0.88
Mooney viscosity ML (1+8) 125°C	—	55 ± 5	25 ± 4
Melt index (g/10 min)	6.0	—	—
Hardness (shore A)	—	68.7	
Ethylene content (%)	—	68 ± 4	70
ENB content (%)	—	4.5 ± 0.6	5 ± 0.4

compression molded at 180°C into 0.2-mm-thick plaques for testing over a period of 15 min. Different formulations varying the proportion of both polymeric phases, over a range of 0–100%, were then analyzed for several techniques.

Measurements

Torque versus time curves were recorded during the processing of the material in the Haake Rheomix 90 internal mixer. The melt flow index (MFI) was measured using a CFR 91 (Campana S.r.l., Italy) following the standard specifications (ASTM D-1238).

The rheological properties measurements were performed using a Rheometric Scientific ARES N2 dynamic mechanical spectrometer with parallel-plate geometry (Rheometrics Scientific, Inc., Piscataway, NJ). Tests were carried out in dynamic mode in a range of temperatures between 180 and 240°C. Tests were determined as a function of angular frequency in the range 0.015 to 20 Hz. The strain amplitude was maintained constant at 5%. The dynamic mechanical properties of the solid polymer were determined with the same instrument at a frequency of 5 Hz and the temperature programs were run from −100 to 50°C under a sinusoidal strain, controlled at a heating rate of 2°C/min in a flow of nitrogen. The oscillating dynamic strains amounted to 0.15%. Specimen dimensions were 50 × 10 × 2 mm.

Thermal analysis experiments were carried out by DSC Perkin–Elmer Pyris 1 (Perkin Elmer Cetus Instruments, Norwalk, CT) differential scanning calorimetry coupled with an intercooler in isothermal and dynamic modes. For isothermal testing, samples of about 8 mg were melted at 200°C for 10 min to eliminate any thermal history of the material; then they were cooled to the crystallization temperature T_c and maintained at that temperature to complete the crystallization of the matrix. Four crystallization temperatures were examined in a range between 125 and 132°C. Dynamic DSC scans were performed at a heating and cooling rate of 10°C/min in a temperature program from −50 to 200°C.

Tensile tests were performed at room temperature on a dynamometer model Lloyd LR 30K (Lloyd Instruments Ltd., UK). Tests were performed with a crosshead speed of 5 mm/min up to a deformation of 20% and 50 mm/min at break. The tensile properties were calculated according to ISO 37-1977. All tensile properties were the average of at least five measurements. Rebound resilience measurements were performed using a Schob pendulum (Zwick & Co., Germany) following the standard ISO 4662-1978 specifications. Compression set was measured according to ISO 815-1972 at 22 h at 70°C with 25% compression.

Impact experiments were carried out according to ASTM D-256 (v-notched) at two temperatures (−30°C and 25°C), with an impact speed of 3.48 ms^{-1} in an Izod pendulum Ceast model Resil 25 (Ceast, Italy), recording the maximum forces and stress during the impact test and the energy to fracture. The notches were prepared in a Ceast electrical notching apparatus at 20% of thickness and the angle of the "V" side grooves was 45°. Impact properties were the average of at least seven measurements.

RESULTS AND DISCUSSION

Rheological Study

The flow properties of the materials studied in the molten state were analyzed by melt index, torque curves, and parallel-plate rheometry. The variation of the melt index as a function of rubber
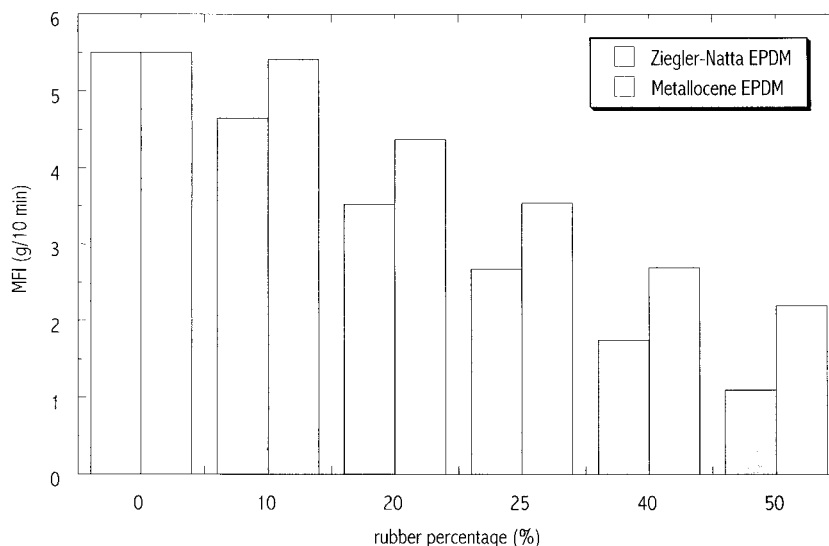
Figure 2 Melt flow index as a function of rubber content in the PP–EPDM blends.

percentage in the blend is reported in Figure 2. The results show that MFI gradually decreases as the rubber percentage in the blend is increased. Moreover, the thermoplastic elastomers prepared with EPDM rubber synthesized by innovative technology present not only a higher MFI compared with that of the conventional EPDM–PP blends but also, as a consequence, a higher flow speed in the molten state. It is well known that the MFI is a measure of the ability of the material to flow[17] and that an increase of MFI indicates that the new polymers have a better response during processing. It is also important to notice that the blends at low rubber percentages (up to 25%) do not present significant changes in the MFI value relative to that of homopolymer PP. Thus, it is reasonable to suppose that up to an elastomer level of 25%, the blends can be processed with the same processing methodology used for the pure PP without any additional cost being tacked onto the final product.

These results were further confirmed for the observations carried out by torque analysis. It is highly recognized that the torque value in the plateau region, obtained after stabilization, characterizes the viscous nature of non-Newtonian material in the molten state.[18] Figure 3 shows the torque values obtained during the processing of the materials in the internal mixer as a function of the blend composition. It can be seen that the incorporation of both EPDMs gives rise to an increase of the PP torque, moreso than in the presence of conventional EPDM rubbers (Ziegler–Natta catalyst). These results indicate that the blends prepared with conventional EPDM are marked by a greater difficulty of processing.

The rheological properties of the different materials in dynamic mode at different temperatures were also analyzed. The variation of the complex viscosity η^* as a function of the angular frequency for pure PP and the PP–EPDM blends (75/25) at 190°C is represented in Figure 4. From these results, it can be determined that the complex viscosity of all materials studied decreases with angular frequency and gradually increases in the presence of the elastomeric phase. Viscosity values of the materials studied at a frequency of 1 Hz (taking the PP viscosity as reference base), at two temperatures (190 and 230°C), are reported in Table II. It is worth noting that the viscosity values of the elastomer thermoplastics processed with conventional EPDM rubbers are higher that those prepared with metallocene catalysts. These results are in agreement with those previously obtained by melt index and torque analysis, demonstrating the better response during processing of the innovative EPDM–PP. These results can be explained by the fact that the new technology gives rise to polymers of a wide molecular weight distribution and low Mooney viscosity, and thus with better processability behavior.

In addition, a general processing behavior similar to that of PP has been observed in the blends with medium and low content of EPDM, particu-
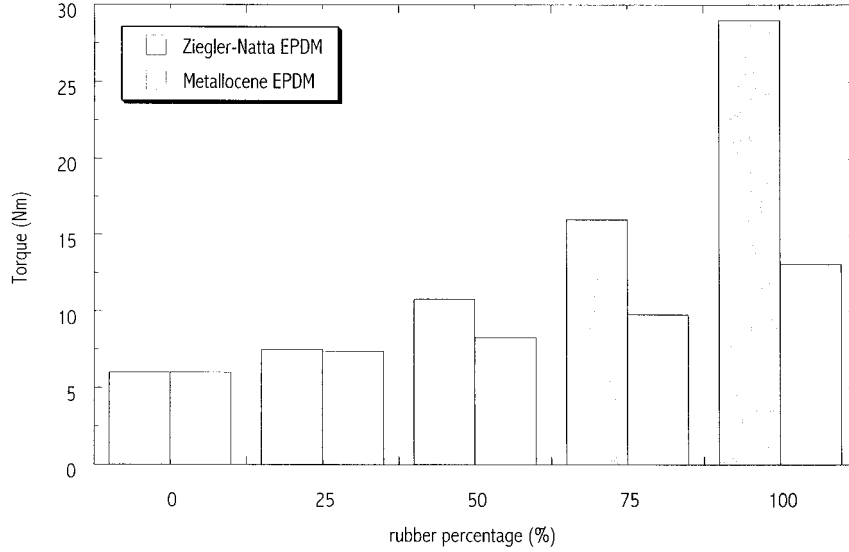
Figure 3 Torque values obtained during the processing of the PP–EPDM blends.

larly when the metallocene EPDM is added to the polymeric matrix. These results indicate that the same industrial thermoplastic molding processes used for virgin PP can be adopted with evident economic advantages. In particular, the blends up to 25% EPDM showed practically the same energy consumption during the mixing as that of virgin PP, indicating that no extra processing costs are expected for these materials. In fact, at this composition, the elastomer forms small particles dispersed in the PP matrix and the rheo-

logical properties of the blend are mainly controlled by the polymeric matrix.

The zero viscosity was also calculated by fitting the rheological data to the equation proposed by Ellis,[19] based on the Cross model[20]:

$$\eta = \frac{\eta_0}{(1 + (K\omega)^m)} \qquad (1)$$

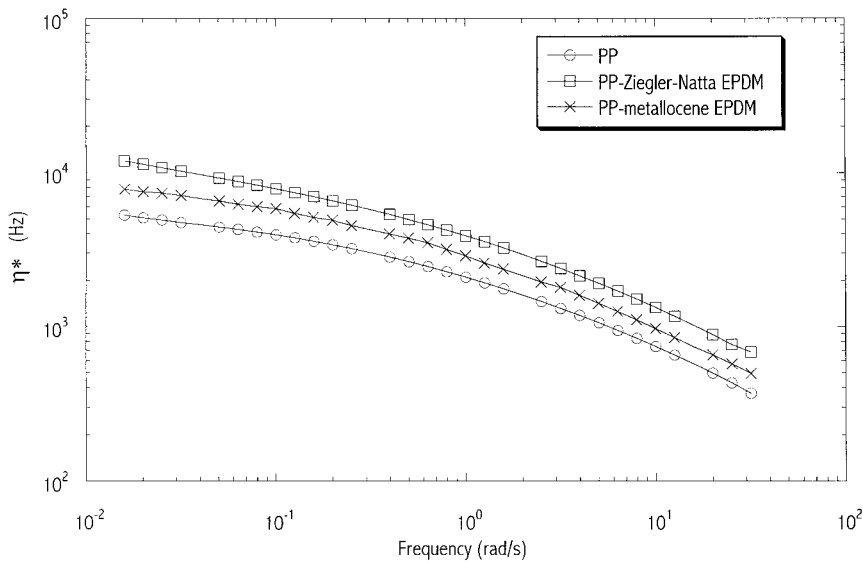where η_0 represents zero shear, a dimensionless constant. The results obtained are summarized in

Figure 4 Complex viscosity for PP and its EPDM blends (75/25).

Table II   Viscosity Properties of PP–EPDM Blends

	$\eta^*_{Blend}/\eta^*_{PP}$ (1 rad/s) at 190°C		Zero Viscosity η_0 (Pa s)$^{-1}$	
PP–EPDM (%)	Ziegler–Natta Catalysts	Metallocenes Catalysts	Ziegler–Natta Catalysts	Metallocenes Catalysts
75–25	1.9	1.4	18,000	10,500
50–50	3.9	1.9	43,000	27,800
25–75	6.3	2.7	87,000	52,000
00–100	12.3	6.8	105,000	73,000

Table II. As should be expected, the zero shear rate viscosity of the polymeric matrix increases with the incorporation of both EPDM rubbers, particularly in the presence of conventional rubbers.

On the other hand, the effect of the catalytic system on the crosslinking reaction of EPDM was evaluated using a curemeter Monsanto MDR 2000 (Monsanto, St. Louis, MO). In both cases, sulfur was used as crosslinking agent and the optimum conditions of the crosslinking reaction were determined (30 min at 160°C). The torque applied during the crosslinking reaction, measured as the difference between the maximum and minimum torque values, was registered and the data obtained for both EPDMs are presented, as a function of time, in Figure 5. From these curemeter curves, it can be easily deduced that the conventional rubbers need a higher energy

consumption during the crosslinking reaction than that of the new EPDMs, indicating a higher degree of difficulty in processing. These results are in concordance with those previously analyzed.

Dynamic Mechanical Analysis

The dynamic mechanical properties of PP–EPDM blends at different compositions were studied over a wide temperature range (−100 to 50°C), the results of which are reported in Table III. In the first place, it is appreciated that the blends show two glass-transition temperatures (T_g's), corresponding to those of their homopolymer constituents and revealing the very limited degree of miscibility of this type of systems. On the other hand, it can be observed that the T_g's of conventional rubbers are slightly higher than the T_g's of
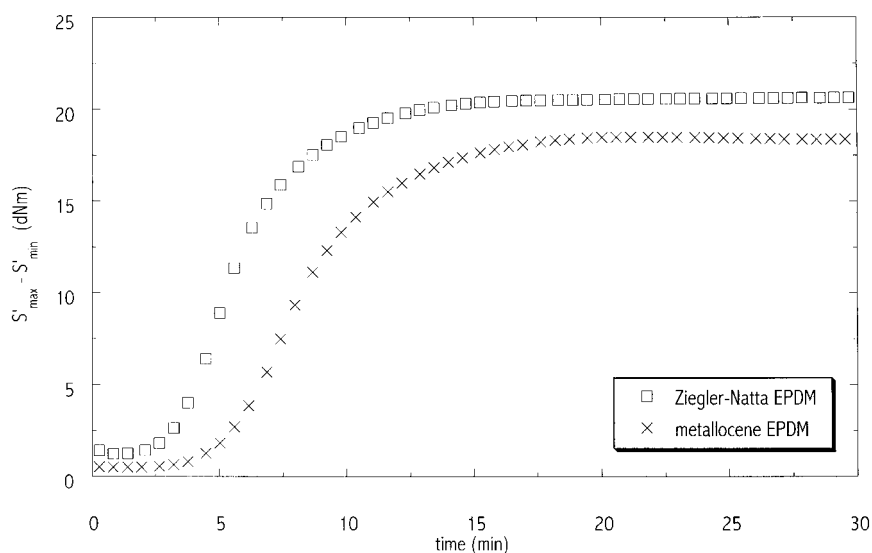

Figure 5   Curemeter curves for both EPDMs.

Table III   Dynamic-Mechanical Properties of PP–EPDM Blends

Property	PP–Ziegler–Natta EPDM				PP–Metallocene EPDM			
	75–25	50–50	25–75	00–100	75–25	50–50	25–75	00–100
T_g (EPDM)	−43.9	−40.1	−33.1	−30.0	−40.3	−34.6	−29.8	−27.5
Tan δ	0.039	0.087	0.268	0.504	0.035	0.098	0.298	0.462
G' (10^9/Pa)	0.51	0.39	0.09	0.053	0.48	0.35	0.04	0.018
T_g (PP)	8.1	5.9	4.0	—	8.0	6.3	4.7	—
Tan δ	0.079	0.079	0.087	—	0.075	0.073	0.079	—
G' (10^9/Pa)	0.27	0.19	0.02	—	0.31	0.28	0.032	—

rubbers catalyzed with metallocenes (−30 and −27.5°C, respectively). According to these results, and assuming that the T_g indicates the temperature limit at which a material acts as elastic element, it can be concluded the EPDMs based on the Ziegler–Natta catalyst have better low-temperature flexibility and a higher extent for use. In addition, it is important to notice that a displacement of the peak corresponding to EPDM to lower temperatures is observed as the PP concentration in the thermoplastic is gradually increased. Similar results have been reported in the literature,[6] where the authors discussed that the incorporation of the elastomeric phase gives rise to a higher thermal expansion coefficient in relation to that of the PP matrix. The cooling of the blend produces a negative hydrostatic pressure that acts on the elastomeric particles, thereby generating thermal tensions that can be responsible for the decrease of the EPDM glass-transition temperature.

Crystallization Study

The effects of the incorporation of both EPDMs on PP crystallization kinetics were also determined by differential scanning calorimetry (DSC), the results of which are reported in Table IV (A and B). Variable results were obtained as a function of the elastomer added, as can be observed in Figure 6, where the degree of crystallization curves as a function of time for PP and its blends [PP–EPDM (50/50)] are represented. It can be seen that the inclusion of conventional rubbers gives rise to an appreciable increase of the PP crystallization rate, showing a marked decrease of the half-time of crystallization. However, the metallocene rubbers show practically no change in the PP crystallization. These results were further confirmed by dynamic tests, as can be observed in Figure 7, where nonisothermal crystallization curves at 10°C/min are represented. Thus, the conventional EPDM gives rise to an appreciable increment of the crystallization temperature (T_c) of PP, whereas the incorporation of innovative rubbers has very little effect on the T_c of the polymeric matrix. On the basis of these results, it can be assumed that the Ziegler–Natta EPDM acts as an effective nucleating agent of PP; however, the metallocene rubbers do not significantly alter the crystallization behavior of the PP matrix. In addition, the blends prepared with both EPDMs present lower degrees of crystallinity compared to that of virgin PP.

Mechanical Properties

To evaluate the effect of EPDMs used for preparing the thermoplastic elastomers, the mechanical properties (tensile test, resilience, and compression set) were analyzed and the results obtained are reported in Table V (A and B). First, it can be observed that the mechanical properties are strongly dependent on the matrix composition. Thus, the tensile modulus and strength decrease significantly with the rubber content in the blend, whereas the incorporation of the elastomeric phase increases the elongation at break of the material. In general, it can be concluded that, as expected, there is a gradual nonlinear increase of the elastomeric behavior of the blend as the concentration of EPDM increases.

On the other hand, different results are obtained as a function of the catalyst used for the polymerization of the elastomer. In general, it was found that the blends containing EPDM rubbers based on metallocene catalysts and process technology are characterized by more elastic

Table IV   Crystallization Parameters of PP and Various PP–EPDM Blends

Material PP-EPDM	T_c (°C)	$\tau_{1/2}$ (s)	k (min^{-n})	n	T_m (°C)	AH (J/g)	Percentage Crystallization
A. PP and PP–conventional EPDM blends							
100–00						97.81	46.8
	125	81	3.25×10^{-1}	2.29	164.2		
	127	126	1.11×10^{-1}	2.40	164.8		
	130	275	1.45×10^{-2}	2.54	165.9		
	132	464	3.97×10^{-3}	2.59	166.8		
90–10						88.62	42.4
	125	36.6	2.25×10^{0}	2.38	164.9		
	127	51.6	1.03×10^{0}	2.62	165.3		
	130	102	1.61×10^{-1}	2.75	165.9		
	132	170	4.43×10^{-2}	2.64	166.6		
80–20						84.88	40.6
	125	37.9	2.15×10^{0}	2.46	164.9		
	127	48.0	1.25×10^{0}	2.64	164.6		
	130	98.0	1.95×10^{-1}	2.58	165.9		
	132	160	5.10×10^{-2}	2.65	166.0		
75–25						79.84	38.2
	125	60	6.93×10^{-1}	2.18	164.9		
	127	97	1.99×10^{-1}	2.70	165.0		
	130	178	4.14×10^{-2}	2.81	165.8		
	132	275	1.34×10^{-2}	2.69	166.8		
60–40						60.97	29.2
	125	68	5.23×10^{-1}	2.25	163.3		
	127	115	1.39×10^{-1}	2.47	164.3		
	130	194	3.78×10^{-2}	2.48	165.3		
	132	303	1.27×10^{-2}	2.47	165.9		
50–50						56.01	26.8
	125	77	3.64×10^{-1}	2.36	165.6		
	127	131	9.24×10^{-2}	2.70	166.1		
	130	213	2.63×10^{-2}	2.61	166.4		
	132	333	8.30×10^{-3}	2.83	166.5		
B. PP and PP–metallocenes EPDM blends							
100–00						97.81	46.8
	125	81	3.25×10^{-1}	2.29	164.2		
	127	126	1.11×10^{-1}	2.40	164.8		
	130	275	1.45×10^{-2}	2.54	165.9		
	132	464	3.97×10^{-3}	2.59	166.8		
90–10						90.90	43.5
	125	99.9	2.00×10^{-1}	2.43	163.3		
	127	148.0	8.15×10^{-2}	2.37	164.6		
	130	304.0	1.35×10^{-2}	2.43	165.3		
	132	499.9	3.46×10^{-3}	2.50	166.3		
80–20						79.70	38.1
	125	86	2.81×10^{-1}	2.50	163.6		
	127	128	1.10×10^{-1}	2.44	164.9		
	130	262	1.88×10^{-2}	2.45	165.6		
	132	456	3.28×10^{-3}	2.64	166.3		
75–25					74.6	35.7	
	125	85.8	2.84×10^{-1}	2.49	163.2		
	127	126.0	1.15×10^{-1}	2.42	164.9		
	130	265.9	1.75×10^{-2}	2.47	164.9		
	132	411.9	5.72×10^{-3}	2.49	166.2		

Table IV *Continued*

| Material PP-EPDM	T_c (°C)	$\tau_{1/2}$ (s)	k (min^{-n})	n	T_m (°C)	AH (J/g)	Percentage Crystallization
60–40						57.07	27.3
	125	92.0	2.52×10^{-1}	2.36	163.3		
	127	141.9	8.84×10^{-2}	2.39	164.3		
	130	296.0	1.61×10^{-2}	2.36	165.3		
	132	435.9	6.30×10^{-3}	2.37	165.9		
50–50						50.78	24.3
	125	104.0	1.79×10^{-1}	2.46	163.2		
	127	172.0	5.30×10^{-2}	2.44	164.3		
	130	358.0	8.11×10^{-3}	2.49	165.3		
	132	494.7	3.90×10^{-3}	2.48	166.2		

properties compared to those with conventional EPDM. In fact, the strength values at 100% elongation (Fig. 8) are slightly higher for the conventional EPDM–PP blends, whereas the elongation at break (Fig. 9) has characteristically higher values in the presence of innovative EPDMs.

These results were further confirmed for those obtained by compression set and resilience tests [Table V (A and B)]. Figure 10 shows the variation of the compression set as a function of both the rubber content in the blend and the type of catalyst used. Effectively, the blends catalyzed with metallocenes present better properties of both compression set and resilience. On the basis of these results, it can be concluded that the met-

allocene polymer has a better sealing property than that of the Ziegler–Natta elastomers.

Impact Properties

Izod impact properties were measured with notched specimens at two temperatures (25 and −30°C) and the results can be directly related to the toughness of the material. The availability of these properties at different temperatures is very useful for the analysis of the performance of the materials studied at service conditions. Experimental results have been used to compute the impact fracture energy given by the area under the force–strain curve measured during the test.
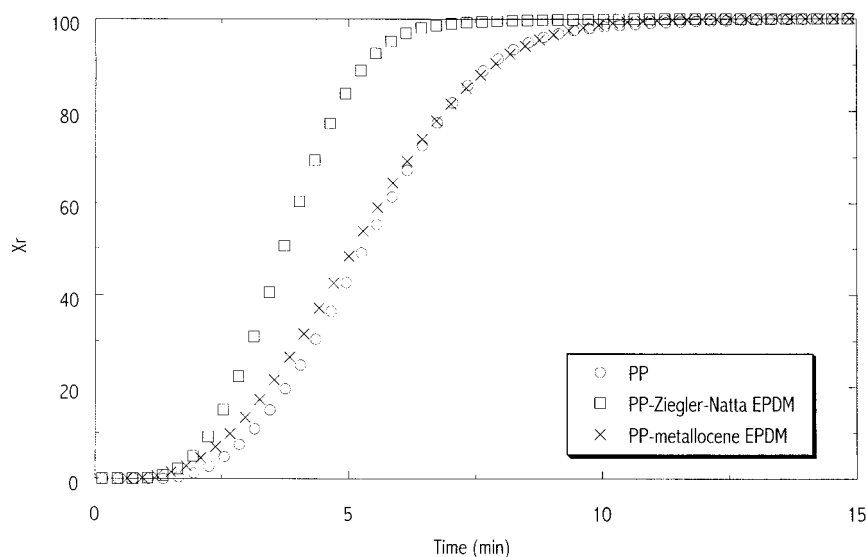
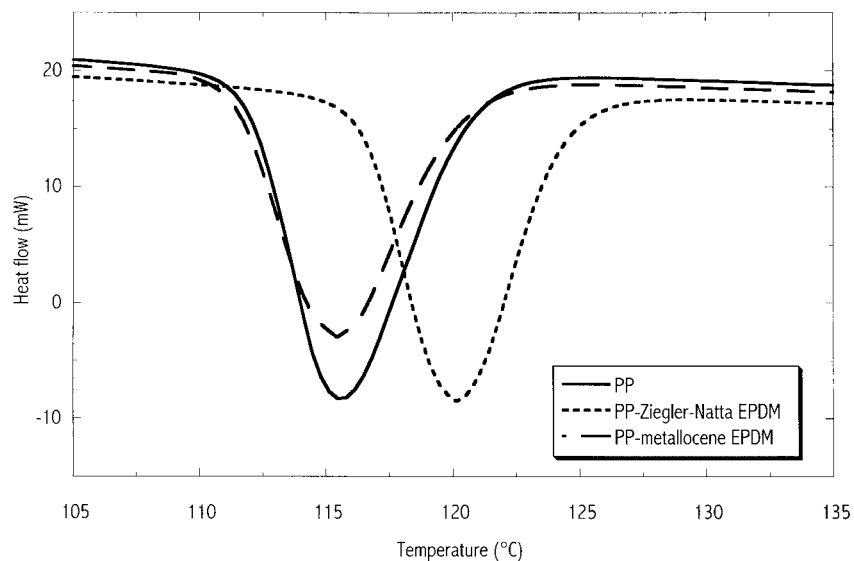**Figure 6** Degree of crystallization of PP and its EPDM blends (75/25) at 130°C.

Figure 7   Nonisothermal crystallization curves of PP and PP–EPDM blend (75/25) at 10°C/min.

Table V   Mechanical Properties of Various PP–EPDM Blends

| PP–EPDM (%)	Modulus (MPa)	Maximum Strength (MPa)	Elongation at Break (%)	Strength: Percentage Elongation (MPa)			Hardness (Shore A)	Resilience (%)	Compression Set (%)
				100	300	500			
A. PP–conventional EPDM blends									
90–10	1245	29.9	22	—	—	—	97.9	28	—
80–20	1024	22.9	85	—	—	—	97.5	28	—
75–25	897	20.8	97	—	—	—	97.4	29	—
60–40	571	15.6	365	14.2	14.9	—	97.0	29	—
50–50	350	12.7	542	9.4	9.8	12.5	95.6	30	78.5
40–60	237	8.6	657	5.3	6.6	7.9	93.7	35	63.4
25–75	38	12.6	1159	4.0	5.8	6.9	82.1	42	45.0
20–80	24.6	10.7	1193	3.2	3.7	4.7	77.4	46	41.7
10–90	8.7	11.9	1200	3.2	3.7	4.6	71.9	47	35.6
00–100	7	12.4	1217	2.5	3.0	4.2	68.5	49	20.1
B. PP–metallocences EPDM blends									
00–10	989	27.1	63.2	—	—	—	95.8	29	—
80–20	940	21.6	135	19.1	—	—	94.9	29	—
75–25	906	19.0	205	17.3	—	—	94.8	28	—
60–40	497	15.2	595	9.3	13.5	14.5	94.6	28	—
50–50	316	10.0	785	6.3	7.9	9.4	93.7	31	59.7
40–60	187	6.5	935	3.6	4.8	6.0	92.9	38	42.4
25–75	27	8.1	1440	3.2	4.5	5.5	79.8	46	28.1
20–80	18	10.4	1527	1.9	2.9	4.3	75.5	49	27.5
10–90	8	10.8	1599	1.8	2.4	3.2	69.8	50	21.4
00–100	5	15.1	1778	1.7	2.2	2.8	65.7	53	16.2
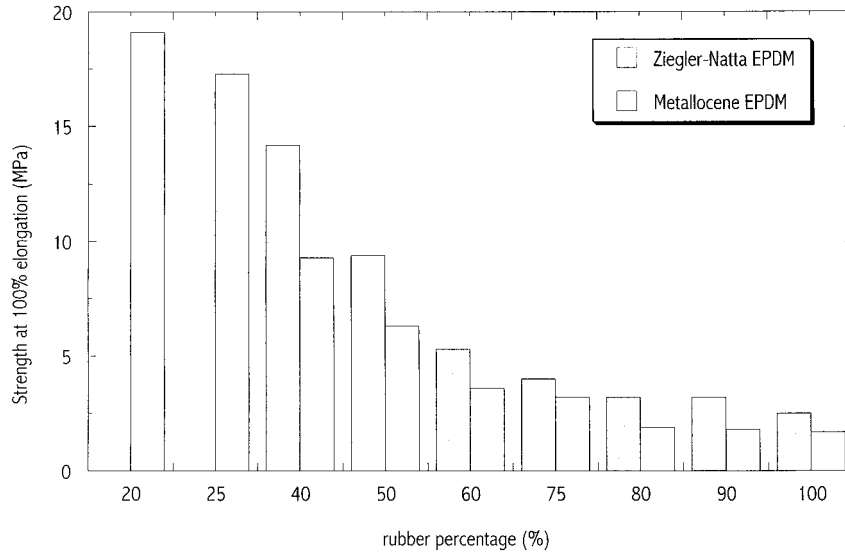
Figure 8 Stress at 100% elongation as a function of rubber content in the PP–EPDM blends.

The impact strength of materials studied is graphically represented in Figure 11. The inclusion of the elastomeric phase to PP gives rise to an appreciable increase of the matrix impact strength. Thus, it can be understood that EPDM rubber behaves as an effective impact modifier of PP, although its effectiveness at low temperatures is reduced. On the other hand, the PP impact strength gradually increases as the elastomer percentage in the blend is increased, par-

ticularly at room temperature. It is also interesting to note that no significant changes between the two EPDMs are observed, demonstrating the effectiveness of the new EPDMs synthesized by new technology as impact modifiers for PP.

The impact properties of polymeric materials are strongly dependent on the ability of the material to absorb and dissipate energy and they can be directly related to the toughness of the material. However, the impact test gives results with
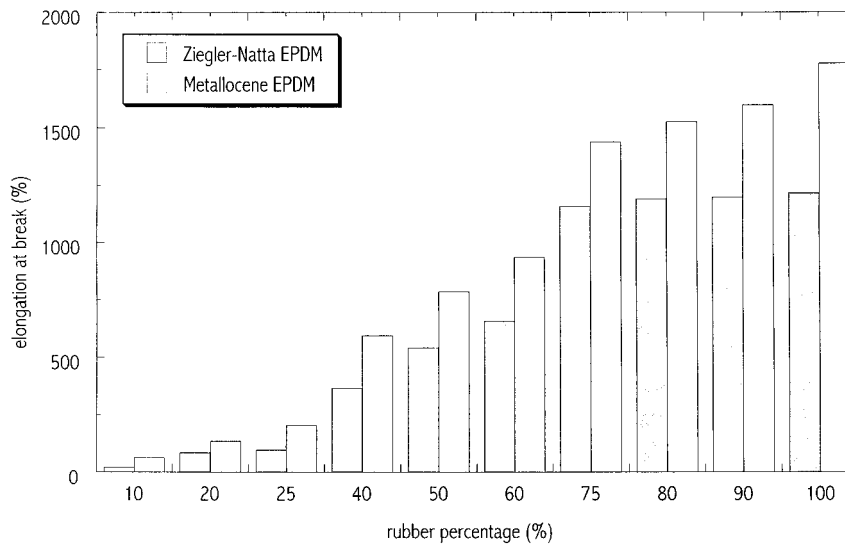
Figure 9 Variation of the elongation at break with the rubber percentage in the PP–EPDM blends.
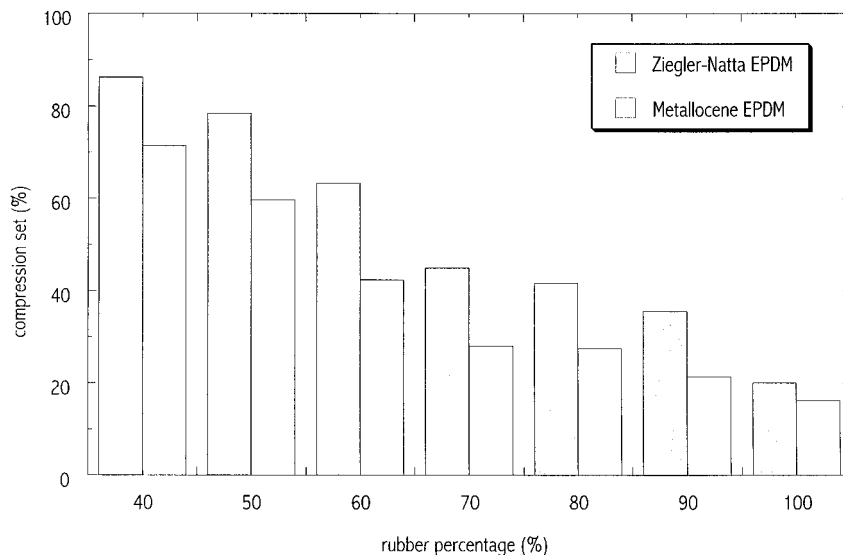
Figure 10 Compression set for both EPDMs.

poor reproducibility[21] and, furthermore, are often difficult to interpret and to compare with those obtained by other means of testing. In particular, in a blend, the explanation of the impact strength results is still more complex than that for a pure polymer because of the part played by both matrices and adhesion at the interface.

CONCLUSIONS

A comparative study between two EPDM rubbers polymerized with conventional Ziegler–Natta and innovative metallocene catalysts was performed. In this sense, different thermoplastic elastomers based on PP and EPDM blends were prepared. The rheological properties, measured by MFI, torque analysis, and rheological study, show that the blends prepared with the innovative EPDM present a better response during processing, showing a significant decrease of the viscosity compared with that of blends processed with conventional EPDM. Moreover, the new class of catalysts based on metallocene chemistry gives rise to materials with a wide molecular weight distribution and low Mooney viscosity, which facilitates
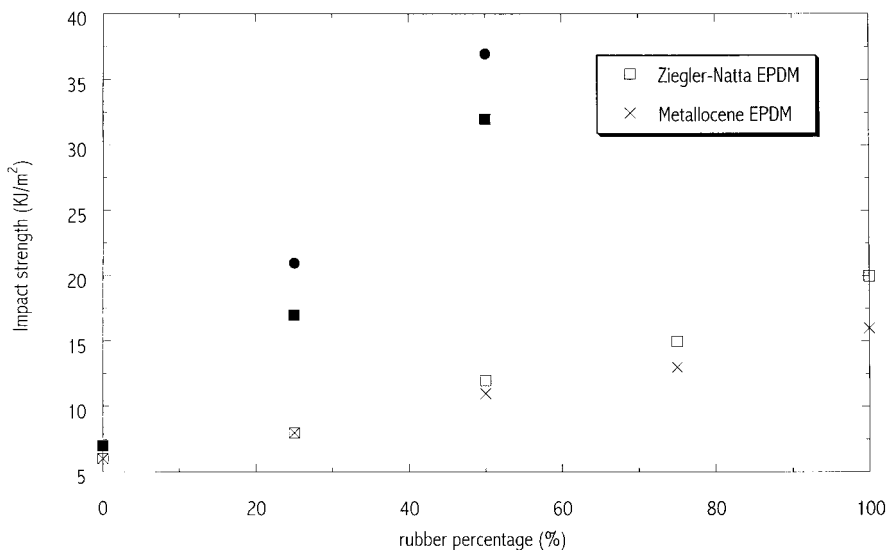

Figure 11 Impact strength of PP–EPDM blends as a function of rubber content and temperature.

the processing of these materials by common techniques, such as compression and injection molding. It is also interesting to note that the processability of the blends at low rubber contents (up to 25%) is similar to that of virgin PP, which suggests that this type of blends can be processed by typical methods of thermoplastics, without additional costs. In addition, the results of the mechanical characterization demonstrated that metallocene polymers are characteristically more elastic, showing a marked increase of both deformation and resilience as well as better compression set properties. Furthermore, the effectiveness of the new EPDMs catalyzed by metallocenes as impact modifier for PP was verified by impact test, from which similar results were obtained compared to those of blends prepared with conventional EPDM.

REFERENCES

1. Synrott, D. J.; Sheidan, D. F.; Kontos, E. G. in Thermoplastic Elastomers from Rubber–Plastic Blends; De, S. K.; Bhowmick, A. K., Eds.; Ellis Horwood: New York, 1990.
2. Matthew, N. M.; Tinker, A. J. J Nat Rubber Res 1986, 1, 240.
3. Batiuk, M.; Harman, R. M.; Healy, J. C. (to B.F. Goodrich Co.) U.S. Pat. 3,919,358, 1975.
4. Hoppner, D.; Wendorff, J. H. Colloid Polym Sci 1990, 268, 500.
5. Karger-Kocsis, J.; Csiku, I. Polym Eng Sci 1987, 27, 241.
6. Da Silva, A. N.; Tavares, M. B.; Politano, D. P.; Coutinho, M. B.; Rocha, M. C. J Appl Polym Sci 1997, 66, 2005.
7. Walker, B. M. Handbook of Thermoplastic Elastomers; Van Nostrand Reinhold: New York, 1979.
8. Whelan, A.; Lee, H. S. Development in Rubber Technology: Thermoplastic Rubbers; Applied Science: London, 1982.
9. Thomas, S.; George, A. Eur Polym J 1992, 28, 1451.
10. Campbell, D. S.; Elliot, D. J.; Wheelans, M. A. Nat Rubber Technol 1978, 9, 21.
11. Anon. Br Plast Rubber 1978, 32.
12. Laird, J. L.; Edmondson, M. S.; Riedel, J. A. Rubber World 1997, 42.
13. Ewen, J. A. Sci Am May 1997.
14. Laird, J. L.; Reidel, J. A. 148th Technical Meeting of the Rubber Division, ACS, October 1995.
15. Sylvest, R. T.; Lancestor, G.; Betso, S. R. Kaut Gummi Kunstst 1997, 50, 186.
16. Reuter, M.; Streit, G. Gummi Fasern Kunstst 1997, 3, 201; Int Polym Sci Technol 1997, 24, 5.
17. Manley, T. R. in Developments in Rubber and Rubber Composites; Evans, C. W., Ed.; Applied Science: London, 1983.
18. Sperling, L. H.; Jordhamo, G. M.; Manson, J. A. Polym Eng Sci 1986, 26, 517.
19. Carriere, C. J.; Silvis, H. C. J Appl Polym Sci 1997, 66, 1175.
20. Cross, M. M. J Colloid Sci 1965, 20, 417.
21. Nielsen, L. E. Mechanical Properties of Polymers and Composites; Marcel Dekker: New York, 1974; p 308.