

Polypropylene–rubber blends: 2. The effect of the rubber content on the deformation and impact behaviour

A. van der Wal¹, R. Nijhof, R.J. Gaymans*

Laboratory of Polymer Technology, Department of Chemical Technology, Twente University, P.O. Box 217, NL-7500 AE, Enschede, The Netherlands

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Abstract

The effect of the rubber content on the deformation and impact behaviour of polypropylene–EPDM rubber blends is studied. The blends are made on a twin screw extruder. The rubber content ranged from 0 to 40 vol.%. The tensile modulus and the yield stress decrease linearly with increasing rubber content. The crystallinity of the PP phase as measured with differential scanning calorimetry did not change with rubber content. The fracture behaviour was studied with a notched Izod impact test and with an instrumented single edged notched tensile test at 1 mm/s and 1 m/s. The blends were studied in temperature range from -80 to 120°C . The brittle–ductile transition temperature (T_{bd}) decreases with increasing rubber content from 85°C for pure polypropylene to -50°C for a 40 vol.% blend, a shift of 135°C . The T_{bd} with the notched Izod test and the SEN at 1 m/s are very comparable. The T_{bd} for the SEN at 1 mm/s if compared to the 1 m/s are at a 30°C lower temperature. The brittle–ductile transition at the low test speed is gradual, while at high test speeds the transition is abrupt, discontinuous. A good criterion for the onset of ductility is the crack propagation displacement. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Polypropylene–EPDM blends; Strain rate; Rubber content

1. Introduction

Polypropylene (PP) is a semi-crystalline polymer with very interesting mechanical and thermal properties. For application as engineering plastic its toughness and in particular its notched toughness is not sufficient. Under impact conditions unnotched polypropylene exhibits a brittle–ductile transition temperature between 0 and 20°C [1], while the corresponding transition temperature of the notched polypropylene is about 100°C [2]. The notched impact toughness can, like with other polymers, be considerably improved by having a dispersed rubber phase present. The deformation of semi-crystalline polymers is mainly by shear banding. The micro deformation of the rubber modified semi-crystalline polymers is a multi-stage mechanism; elastic loading of the blend, cavitation of the rubber particles and shear deformation of the matrix [3,4]. The rubber has two functions: it has to relieve the volume strain by cavitation and it has to act as a stress concentrator. Relieving of the volume strain is directly related to the rubber content [5] and also the stress concentration is directly related to the rubber content [6].

The rubber in PP can cavitate and the PP deforms after cavitation by shear banding [7,8]. As a result of internal voiding of the rubber particles, the triaxial stress behind the crack tip is partially relieved. This change in the stress state facilitates yielding of the matrix.

Rubber modified PP can be made in a reactor process or by blending rubber in PP on an extruder. Ethylene–propylene copolymers are commonly employed as impact modifiers for polypropylene. The morphology of the polypropylene matrix is affected by the dispersed rubber phase. Ethylene–propylene copolymers, in particular, act as crystallization nuclei and thus reduce the spherulite size of the polypropylene matrix [9,10]. The crystallinity of the polypropylene matrix is not at all, or at most only slightly, affected by the dispersed ethylene–propylene copolymer [11–13].

The deformation and impact behaviour of polypropylene–rubber blends has been studied extensively [14–39]. The modulus [17,23,35] as well as the yield stress [17,38] decrease with rubber content. This decrease exceeds expectations based on the Rule of Mixtures. The effect of the rubber content on the modulus [17,23,29] and the yield stress [17,38] for polypropylene–rubber blends has been described by general compound models. The models for the modulus basically differ from those describing the yield stress. The particle size has no, or only a slight effect on the

* Corresponding author.

¹ Present address: Resina Chemie, 9607 PS Foxhol, The Netherlands.

Table 1
Starting materials (all data from data sheet [42,43])

Polypropylene		EPDM	
Grade	Vestolen P7000	Grade	Keltan 820
MFI ^a	2.4 dg/min	Mooney ^b	74
Density	901 kg/m ³	Density	860 kg/m ³
Modulus	1500 MPa	Ethylene content	55 wt.%
Yield stress	34 MPa	Termonomer	DCPD

^a 230°C, 21.6 N.

^b ML (1 + 4), 125°C.

modulus and the yield stress of polypropylene–rubber blends [39].

The notched fracture behaviour of rubber toughened polypropylene depends on test conditions such as temperature and test speed. With increasing temperature and decreasing test speed [40], the fracture type changes from brittle to ductile. Polypropylene and rubber modified polypropylene show a sharp brittle–ductile transition. The effect of the test speed on the fracture behaviour is rather complicated since with increasing test speed the plastic deformation process in front of the crack tip changes from isothermal to adiabatic. Consequently at the crack tip the temperature rises with increasing test speed [40].

The fracture behaviour of rubber toughened polypropylene depends also on the blend morphology: particle size and rubber content. The effect of the particle size on the brittle–ductile transition temperature (T_{bd}) depends on the test speed [39]. At low test speed, the T_{bd} decreases slightly with increasing particle size, whereas at high test speed (impact conditions) the T_{bd} increases strongly with increasing particle size [39].

Several criteria have been proposed for the brittle–ductile transition of polymers. A criterion based on the precrack plastic-zone volume was proposed for polycarbonate [41]. The precrack plastic zone is the plastic zone formed in front of the notch tip during crack initiation. It was suggested that the fracture type changes from brittle to ductile if this plastic zone volume exceeds a critical value. The size of the precrack plastic zone seems to be related to the displacement at the moment of crack initiation.

We studied the effect of the rubber content on the fracture behaviour of polypropylene–EPDM blends at low and high test speed. Special attention is paid to the brittle–ductile transition and the related change in the deformation process.

2. Experimental

2.1. Materials

The materials used in this study are a polypropylene homopolymer (Vestolen P7000, Vestolen GmbH) and an EPDM rubber (Keltan 820, DSM). The materials were kindly supplied by their manufacturers and were used as

delivered. The materials specifications are listed in Table 1 [42,43].

2.2. Specimen preparation

Blends were made on a corotating twin screw extruder. During a first extrusion step, a 60/40 vol.% PP–EPDM master blend was prepared. During a second extrusion step, this master blend was diluted to afford a blend series with varying rubber content. Both extrusion steps were carried out on a Berstorff ZE 25 co-rotating twin screw extruder ($L/D = 33$, $D = 25$ mm). The second extrusion step was carried out over the last quarter of the screws to minimize the change in blend morphology. The extrusion conditions for the first extrusion step were 190°C and 100 rpm, while the conditions for the second extrusion step were 230°C and 100 rpm.

Rectangular bars (ISO 180/1, $74 \times 10 \times 4$ mm³), and dumb-bell-shaped specimens (ISO R527-1, $10 \times 3 \times 115$ mm³) were injection moulded on a 221-55-250 Arburg Allrounder injection moulding machine. The barrel and the nozzle temperature were 200 and 230°C respectively, and the screw speed was 100 rpm. The mould temperature was 40°C. A single-edge 45° V-shaped notch (tip radius 0.25 mm, depth 2 mm) was milled in the bars.

2.3. Particle size analysis

Samples for particle size analysis were taken from the core of the bars. The details concerning the particle size analysis are found elsewhere [44]. The number (D_n) and weight (D_w) average particle size was determined. These averages are defined as:

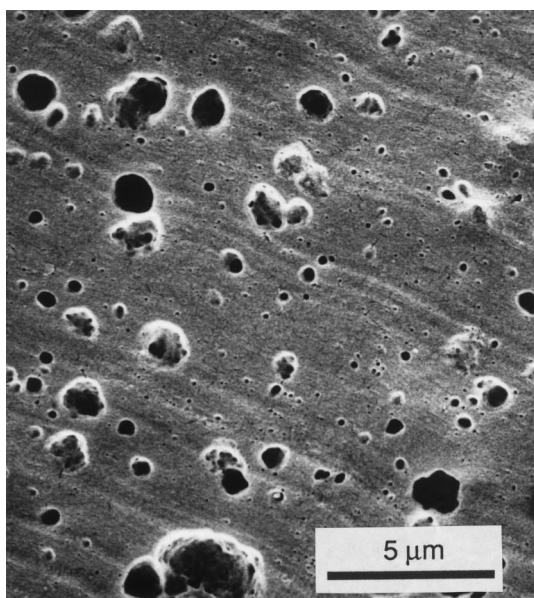
$$D_n = \frac{\sum n_i d_i}{\sum n_i}, \quad D_w = \frac{\sum n_i d_i^2}{\sum n_i d_i} \quad (1)$$

2.4. Properties

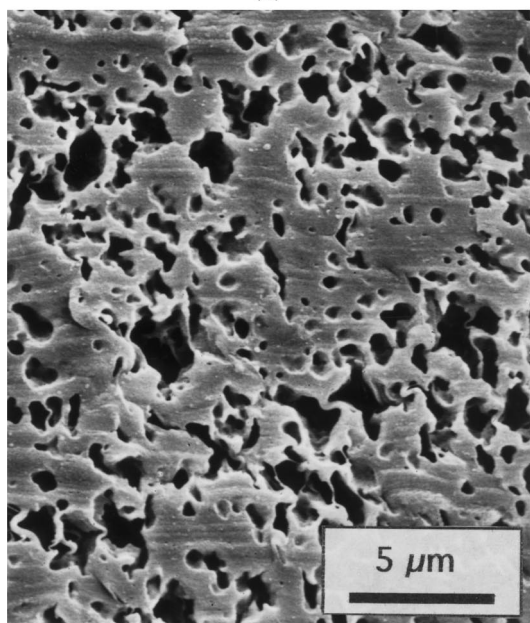
Differential scanning calorimetry (DSC) was carried out on a Perkin–Elmer DSC 7. Samples (about 10 mg) were taken from the core of the dumb-bell-shaped specimens. Scans were recorded at a heating rate of 20°C/min. The melting temperature was defined as the peak temperature, and the melting enthalpy was calculated from the peak area. Measurements were performed twice.

Dynamical mechanical analysis (DMA) was performed on the gauge section of the dumb-bell-shaped specimen ($50 \times 10 \times 3$ mm³) by a Myrenne ATM3 torsion pendulum. The spectra of the blends and pure polypropylene were recorded at a heating rate of 1.8°C/min, the EPDM spectra were obtained at 0.5°C/min.

The tensile tests were carried out on the dumb-bell-shaped specimens (ISO R527-1, $10 \times 3 \times 115$ mm³) with an Instron tensile machine (5 mm/min) (strain rate 8.3×10^{-4} s⁻¹). The impact strength was determined by the Izod impact test on notched bars (ISO 180/1, $74 \times 10 \times 4$ mm³).



(a)



(b)

Fig. 1. SEM micrographs illustrating the blend morphology: (a) 10 vol.%; and (b) 40 vol.% blend.

Table 2
Particle sizes for the PP–EPDM blends

Rubber content (wt.%)	D_n^a (μm)	D_w^b (μm)
5	0.25	0.50
10	0.28	0.67
20	0.34	0.69
30	0.42	0.86
40	0.67	0.92

^a D_n = number average particle size.

^b D_w = weight average particle size.

Table 3
Glass transition temperatures at 1 Hz for the PP–EPDM blends and the starting materials

Rubber content (wt.%)	T_g : PP (°C)	T_g : EPDM (°C)
0	6	–
10	4	–55
20	4	–54
30	3	–53
40	2	–53
100	–	–54

The single edged notched (SEN) tensile test with the Izod notched bars were carried out on a Schenck servo-hydraulic tensile machine (fitted with an oven) at 1 mm/s and 1 m/s. The stress was calculated by dividing the force by the initial cross-sectional area behind the notch ($\sim 32 \text{ mm}^2$). The sample length between the clamps was 45 mm. All measurements were done five times. Determining the strain rates of the necking materials studied with a notched tensile test is difficult. An estimation of the strain rate can be obtained in the region where the deformation is still elastic. As the cross section ahead of a notch is 20% smaller than in the rest of the sample the strain rates there are 20% higher. With a clamp distance of 45 mm the strain rate estimated in the elastic region is at 1 mm/s (0.027 s^{-1}) and at 1 m/s (27 s^{-1}).

3. Results and discussion

3.1. Materials

Blends with a fine dispersion were made of PP (with an MFI of 2.4) and an EPDM (with a Moony viscosity of 74) on a twin screw extruder. Extrusions at very high shear rates and temperatures $\geq 230^\circ\text{C}$ were avoided as at these conditions degradation of polypropylene takes place. First a 40% blend was made which was diluted in a second blending step. For testing, the samples were injection moulded.

The morphology of the blends is studied with SEM (Fig. 1). The surfaces are parallel to the flow direction during injection moulding. Even at 40% rubber, the rubber is well dispersed. The rubber particles are fairly randomly distributed but rows of particles can be seen. Elongation of particles is not observed. The particle sizes of the blends are listed in Table 2. Both the number (D_n) and the weight

Table 4
Melting enthalpy at the first heating stage for the polypropylene and the PP–EPDM blends

Rubber content (vol.%)	$\Delta H_{m,1}$ (J/g)	$\Delta H_{m,1}$ (J/g PP)
0.0	96	96
10.0	86	96
20.0	76	95
30.0	64	91
40.0	57	95

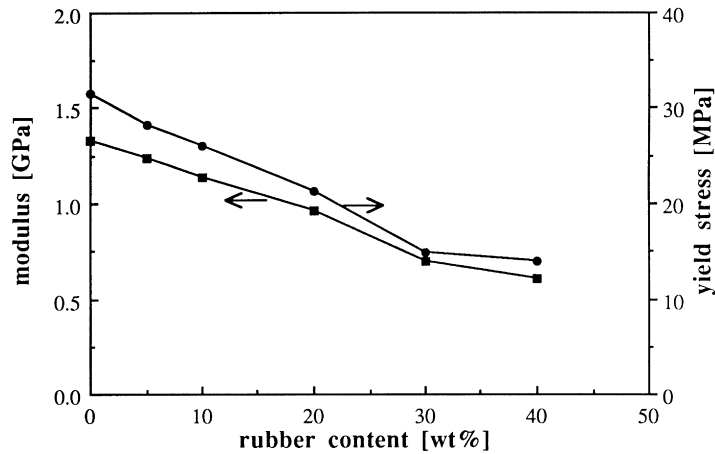


Fig. 2. Tensile modulus and tensile yield stress as a function of the rubber content for the PP-EPDM blends at 5 mm/min: ■: modulus; ●: yield stress.

average (D_w) particle size increases with increasing rubber content. A similar trend has been observed for other blend systems [45].

The glass transition temperature (T_g) values of the blends and the starting materials were determined by Dynamical Mechanical Analysis at 1 Hz (Table 3). The T_g is defined as the peak in the loss modulus curve (G''). If T_g is defined as the peak in the $\tan \delta$ curve, it would be about 5°C higher. The T_g of the EPDM in the blend is almost equal to the T_g of pure EPDM. The T_g of the polypropylene phase decreases with increasing rubber content, this is attributed to a plastizing effect [29,46]. Thus with increasing rubber content some plastization of the PP phase seems to take place.

The melting enthalpy in a DSC experiment at varying rubber content is listed in Table 4. The melting enthalpy of the blends decreases with decreasing polypropylene content. The melting enthalpy of the matrices remains almost constant irrespective of the rubber content, except

for the 30 vol.% blend which exhibit a slightly lower matrix melting enthalpy. At a heat of fusion of 207 J/g for 100% crystalline polypropylene [47], the crystallinity of PP in the blends is about 45 wt.%.

3.2. Tensile properties

The tensile modulus and the tensile yield stress (strain rate $8.3 \times 10^{-4} \text{ s}^{-1}$) as a function of rubber content are shown in Fig. 2. The modulus and the yield stress decrease almost linearly with increasing rubber content. The effect of the rubber content on the modulus and the yield stress is almost as strong, although these effects are described by entirely different models [6]. The decrease in the modulus and the yield stress is approximately 55% for the 40 vol.% blend compared to pure polypropylene. The decrease in modulus is stronger than the rule of mixture expected for a blend with well dispersed particles.

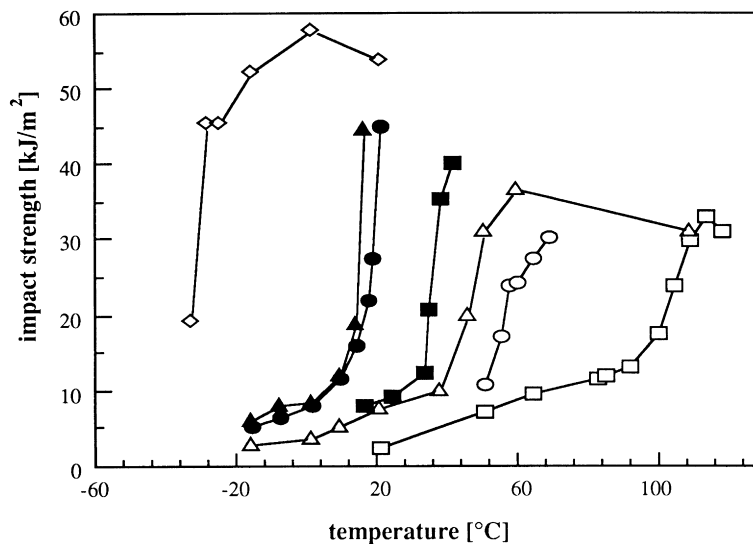


Fig. 3. The notched Izod impact strength as a function of the temperature for pure polypropylene and the PP-EPDM blends. Rubber content (vol.%): □, 0%; ○, 5%; △, 10%; ■, 15%; ●, 20%; ▲, 25%; and ◇, 30%.

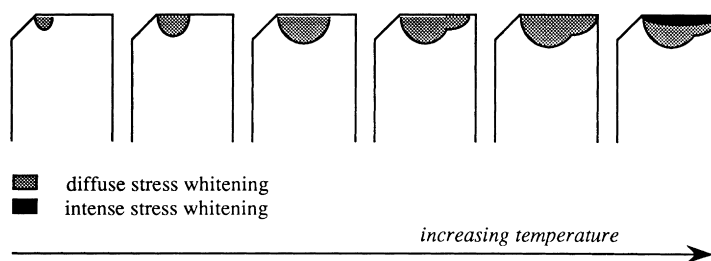


Fig. 4. Schematical representation of the development of the stress whitening zone after a notched Izod impact test with respect to temperature in the polypropylene-EPDM blends.

3.3. Notched Izod

The notched Izod impact strength of blends as a function of the temperature is shown in Fig. 3. The impact strength increases with the temperature and shows a sharp (stepwise) increase with respect to temperature, followed by a decrease. The impact strength at low temperatures is relatively low (2–10 kJ/m²) and the fracture is brittle. At high temperatures the impact strength is very high (30–60 kJ/m²) and the fracture is ductile. The transition from brittle to ductile is sharp (step wise). The brittle-ductile transition shifts with rubber content to lower temperatures. In the ductile region the impact strength decreases with temperature. The decrease in impact strength at higher temperatures is accompanied by the incomplete fracture of the specimens. In notched tensile experiments where the samples were fully fractured a similar decrease was observed. Thus the decrease in impact strength with temperature is mainly a temperature effect. The decrease in impact strength with temperature might be due to the decrease in yield stress with temperature [2].

All the blends showed significant stress whitening after

fracture. The evolution of the stress whitening zone (SWZ) with respect to the temperature is shown in Fig. 4. The effect of the temperature and the rubber content on the evolution of the SWZ is equivalent. At low temperatures the first SWZ changes from restricted to a zone around the notch, to be extended over the width of the specimen. At a temperature where there is a sharp increase in the impact strength, a second, more intensely SWZ was formed along the fracture surface. The intense SWZ is surrounded by a more diffuse SWZ as was observed previously [11,44]. The diffuse stress whitening is due to voided rubber particles [7]. The intense stress whitening is due to a strongly deformed voided particles as a result of matrix yielding [7].

3.3.1. Brittle-ductile transition in Izod

The brittle-ductile transition temperature (T_{bd}) from the Izod test is taken as the intersection of the tangent line of the sharp increase with the tangent line of the linear part in the impact strength temperature curve prior to the sharp increase. In the brittle region the fracture is fast but there can be plastic deformation ahead of the notch before the fracture is initiated. In the brittle region the stored energy

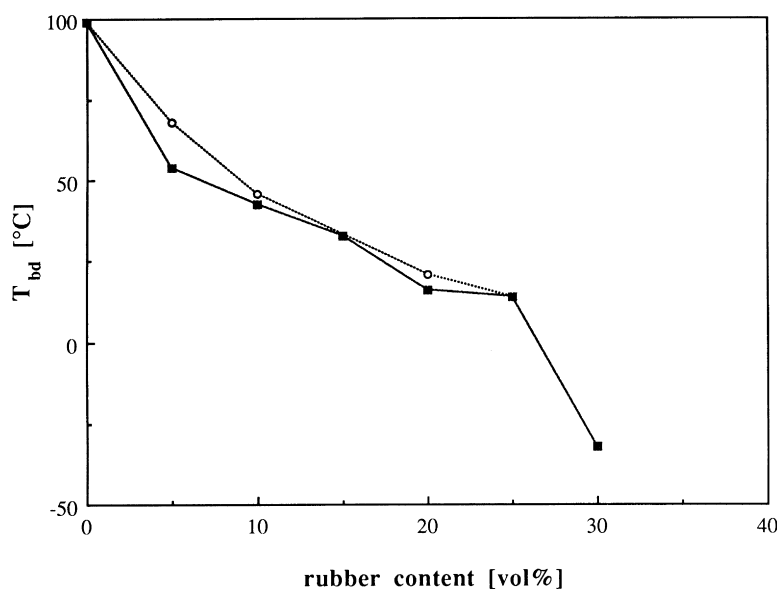


Fig. 5. The Izod brittle-ductile transition temperature (T_{bd}) versus the rubber content for the PP-EPDM blends as measured by notched Izod: ■, actual blends; ○, corrected for particle size variation ($D_w = 0.9 \mu\text{m}$).

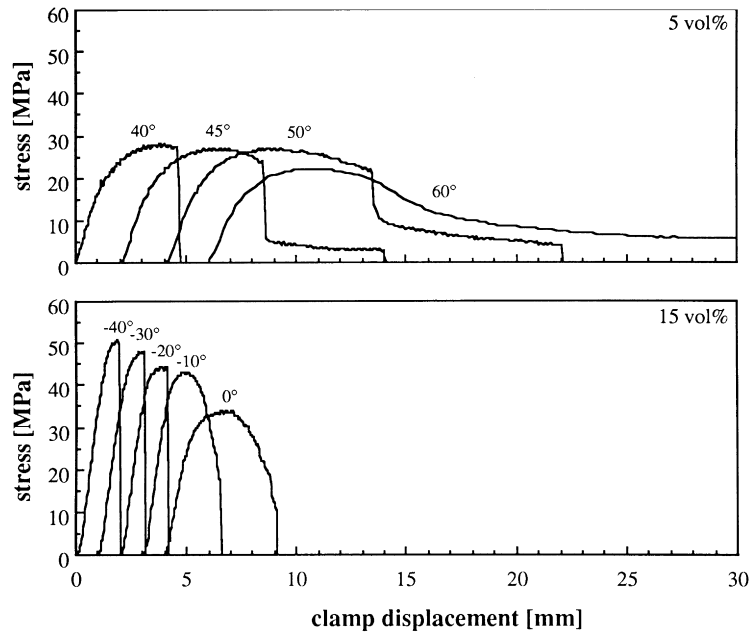


Fig. 6. Stress–strain (displacement) curve obtained by an SEN tensile test (1 mm/s) for the PP–EPDM blends at various temperature for a 5 vol.% and a 15 vol.% blend.

in the sample at the moment of crack initiation is sufficient to propagate the crack through the entire specimen. In the ductile region, the whole fracture surface is intensely stress whitened, the crack speed is much lower and for crack propagation additional energy is needed. The T_{bd} strongly decreases with rubber content (Fig. 5). The neat PP has a T_{bd} at 100°C. The 30% rubber blend has its T_{bd} at –30°C, this is 130°C lower than for PP and well below the T_g of PP. In the studied blends the particle size increases with rubber content (Table 2). It is known that with increasing particle size the T_{bd} shifts to higher temperatures [39], and with the particle size correlation a correction is made for the studied blends.

The corrected T_{bd} was calculated for a particle size, $D_w = 0.9 \mu\text{m}$ (Fig. 5). This means that the corrected T_{bd} of the blends with the low rubber content are now at a higher temperature, but the effect of the correction is small.

3.4. SEN tensile test: 1 mm/s

The notched fracture behaviour at low test speed was studied by a single-edged notched (SEN) tensile test at 1 mm/s. The stress–displacement curves obtained at different temperatures for the 5 and 15 vol.% blend are given in Fig. 6. The stress–displacement curves in the temperature

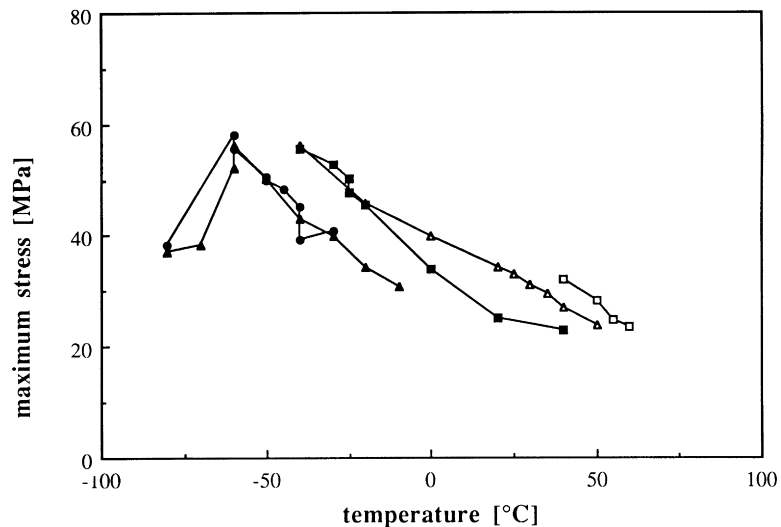


Fig. 7. Maximum stress in a SEN tensile test (1 mm/s) as a function of the temperature for pure polypropylene and the PP–EPDM blends. Rubber content (vol.%): □, 0; △, 10; ■, 20; ●, 30; and ▲, 35. The stress is the net cross-sectional stress, neglecting stress concentration at the notch tip.

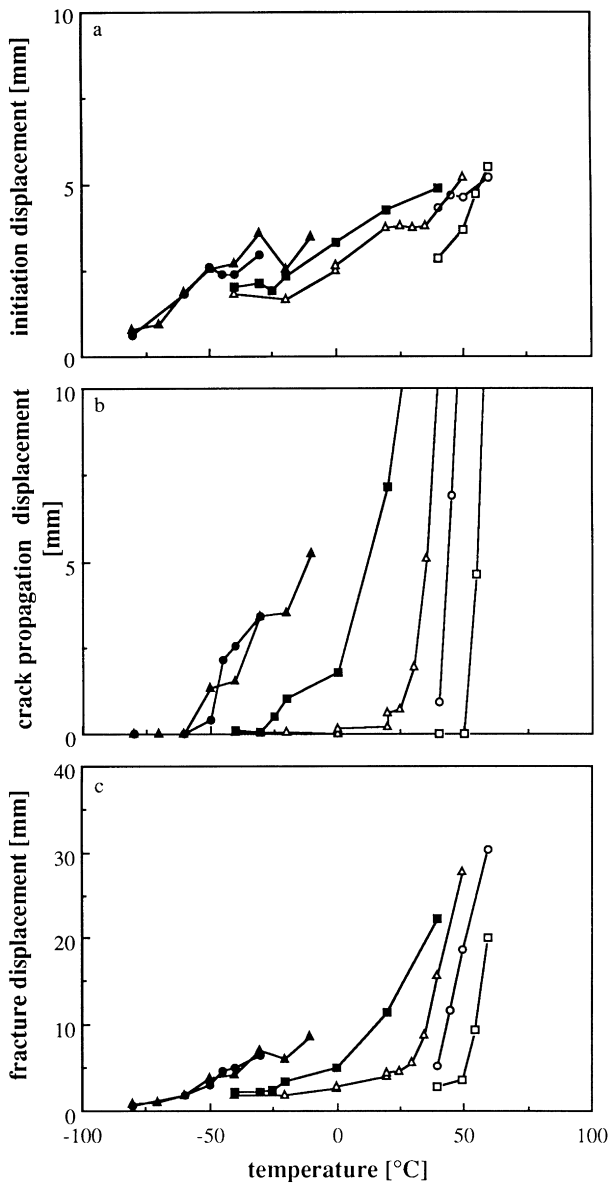


Fig. 8. Displacement using SEN tensile tests at 1 mm/s for pure polypropylene and the PP-EPDM blends. Rubber content (vol.%): \square , 0; \circ , 5; \triangle , 10; \blacksquare , 20; \bullet , 30; and \blacktriangle , 35.

range of the brittle–ductile transition of the 5 vol.% blend is similar to the curves for PP [2] and are typical of blends with a rubber content ≤ 10 vol.%. The curves of the 15 vol.% are typical of blends with a rubber content ≥ 15 vol.%.

The fracture process may be divided into an initiation stage and a crack propagation stage. During the initiation stage deformation is taking place before a crack is initiated. Crack propagation starts after the crack has been initiated. In the stress–displacement curve this is near the stress maximum. The sharp drop in stress at low temperatures means that after a crack has been initiated no additional energy is needed for fracture propagation. This type of fracture is referred to as brittle fracture. At higher temperatures

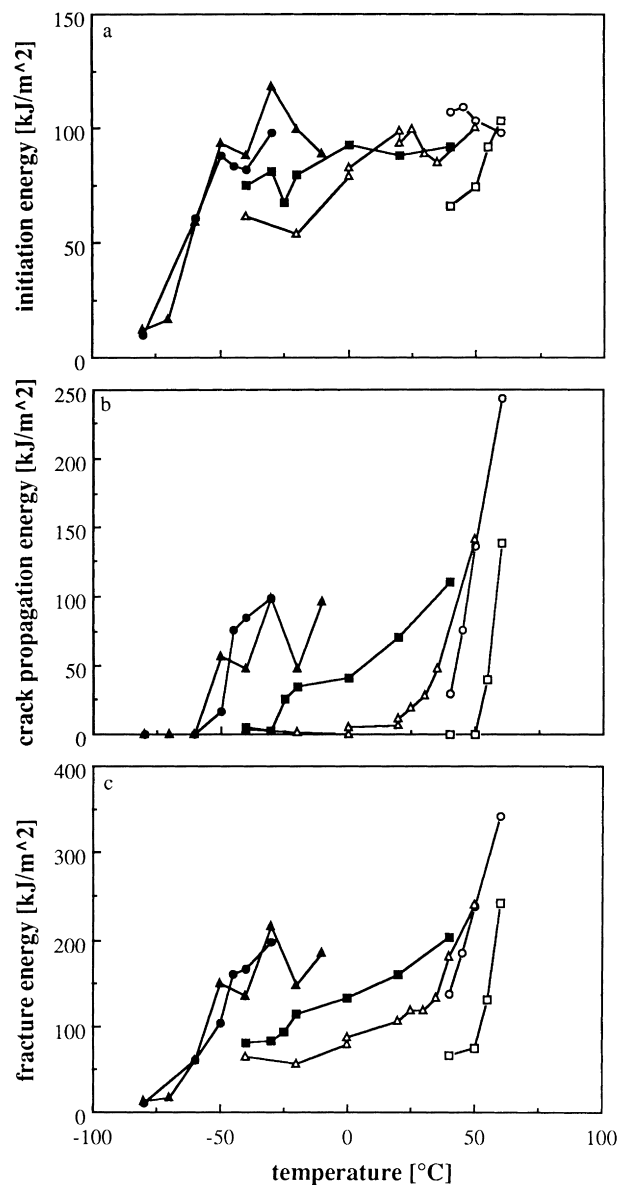


Fig. 9. Supplied energy during SEN tensile tests at 1 mm/s for pure polypropylene and the PP-EPDM blends. Rubber content (vol.%): \square , 0; \circ , 5; \triangle , 10; \blacksquare , 20; \bullet , 30; and \blacktriangle , 35.

additional energy is needed for fracture. This type of fracture is referred to as ductile.

For the 5% blend the brittle fracture is below 40°C and the fully ductile fracture is at $\geq 60^\circ\text{C}$. In the temperature region between 40 and 60°C the fracture after crack initiation starts as brittle but becomes ductile. In the case of ductile fracture (above 60°C), the 5 vol.% blend exhibits pronounced yielding with necking before crack initiation. The crack initiation is after the stress maximum (Fig. 6). In the 15 vol.% blend this yielding ahead of the notch is considerably less pronounced and the crack initiation is at the stress maximum (Fig. 6). Here too the ductile fracture just past the brittle ductile transition at -25 and 0°C is not fully ductile, but

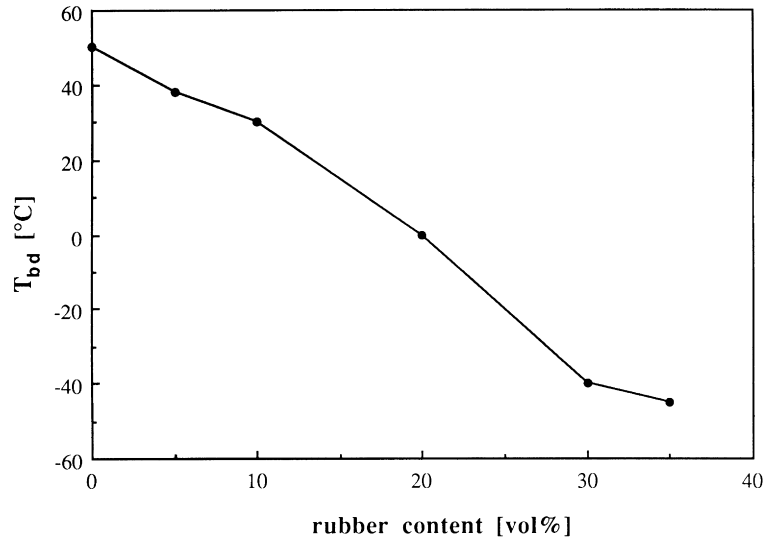


Fig. 10. Brittle–ductile transition temperature (T_{bd}) versus the rubber content at 1 mm/s for the PP–EPDM blends.

now there is a brittle part at the end of the fracture path. The brittle–ductile transition at low speeds is a gradual process.

The ‘Maximum stress’ is defined as the maximum stress in the stress–displacement curves obtained by SEN tensile tests. The stress is the net cross-sectional stress, neglecting the stress concentration at the crack tip. The maximum stress as a function of the temperature is shown in Fig. 7. For the sake of clarity, only some of the tested blends are shown.

In the very low temperature region the maximum stress increases with temperature, followed by a gradual decrease at higher temperatures. The increase in maximum stress in the very low temperature region with temperature is a result of an increase in fracture strain of the very brittle materials. Past the maximum matrix yielding at least in the notch is possible. The decrease in maximum stress at higher

temperatures is due to a lowering of the yield stress [2]. The increase in maximum stress at very low temperatures is accompanied by the onset of stress whitening. This stress whitening is due to internal voiding of the rubber particles [1,5]. Thus the voiding of the rubber particles plays a role in the increase in the maximum stress. At temperature below the T_g of the rubber, voiding of the rubber does not take place. If the rubber has cavitated then the volume strain is lowered and shear yielding is possible. At higher temperatures the maximum stress values decreases with increasing rubber content. The higher the rubber content the lower the PP content is and thus the lower yield stress is.

The displacements of initiation, crack propagation and fracture are given as a function of temperature (Fig. 8). The energies of initiation, crack propagation and fracture are given in Fig. 9. For the sake of clarity, only some of the

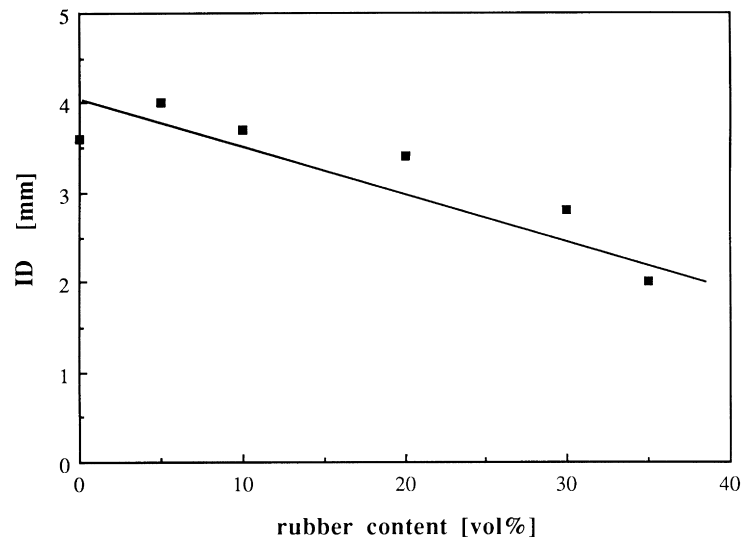


Fig. 11. Initiation displacement at the brittle–ductile transition (ID_{bd}) versus the rubber content at 1 mm/s for the PP–EPDM blends.

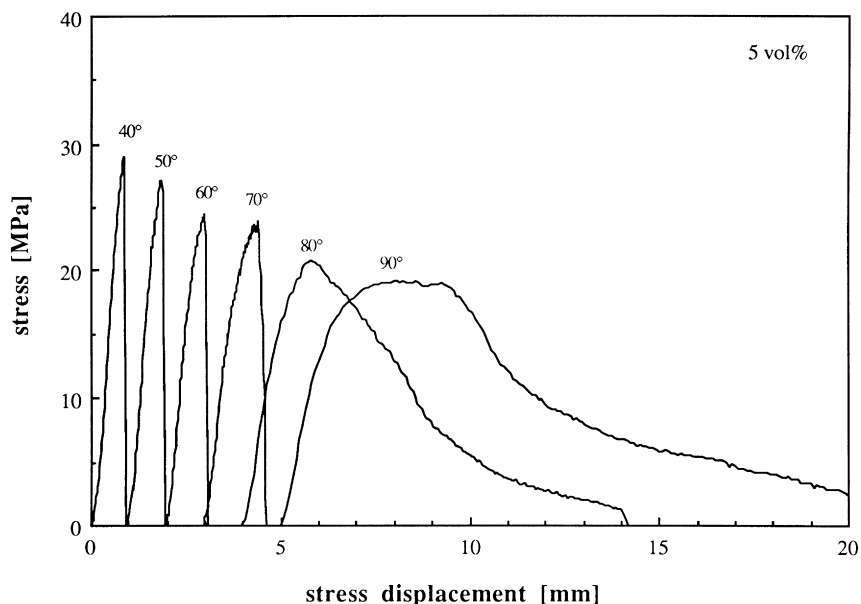


Fig. 12. Stress–displacement curves obtained by an SEN tensile test (1 m/s) for the 5 vol.% blend at various temperatures.

tested blends are shown. Crack propagation was assumed to start at the transition in the stress–displacement curve near the stress maximum. However, in the case of highly ductile fracture no clear transition was observed and determination of the beginning of crack propagation was somewhat arbitrary.

The initiation displacement (ID) increases with increasing temperature and with rubber content (Fig. 8(a)). At higher temperatures and with more rubber present the plastic deformation before crack initiation is larger. At an ID of approximately 4 mm, necking over the entire cross section of the specimen is observed. The initiation energy (IE) increases strongly in the brittle region (-70 and -50°C) and then gradually increase $> -50^{\circ}\text{C}$ (Fig. 9(a)). In the

ductile region both the temperature and the rubber content seems to have little effect on the IE. At temperatures $> -50^{\circ}\text{C}$ the maximum stress decreases and the initiation displacement increases with temperature and rubber content. For the Initiation Energy these two effects seem to level each other out.

The displacement to fracture after a crack has been initiated is called the crack propagation displacement (CPD) and the additional energy necessary to fracture the sample after a crack has been initiated is called the crack propagation energy (CPE). The CPD (Fig. 8(b)) and the CPE (Fig. 9(b)) increase strongly with temperature. At low temperatures, in the brittle region, the CPD and the CPE are zero. At a certain temperature, the CPD and the

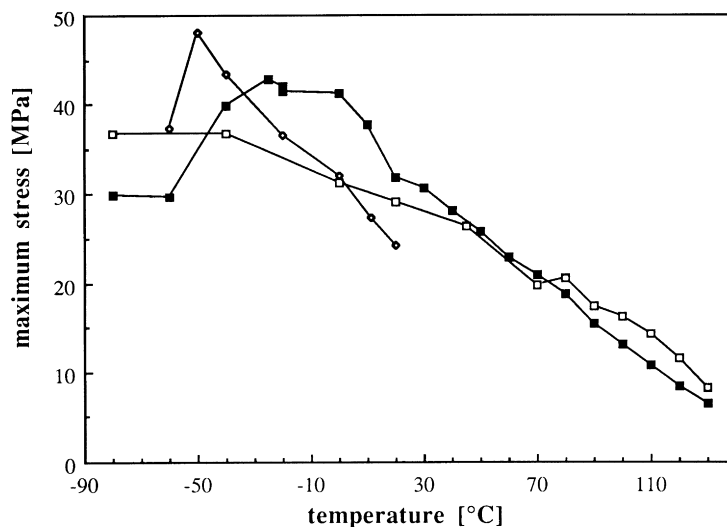


Fig. 13. Maximum stress during an SEN tensile test (1 m/s) as a function of temperature for pure polypropylene and the PP–EPDM blends. Rubber content (vol.%): \square , 0; \blacksquare , 20; and \diamond , 40. The stress is the net cross-sectional stress, neglecting the stress concentration at the notch tip.

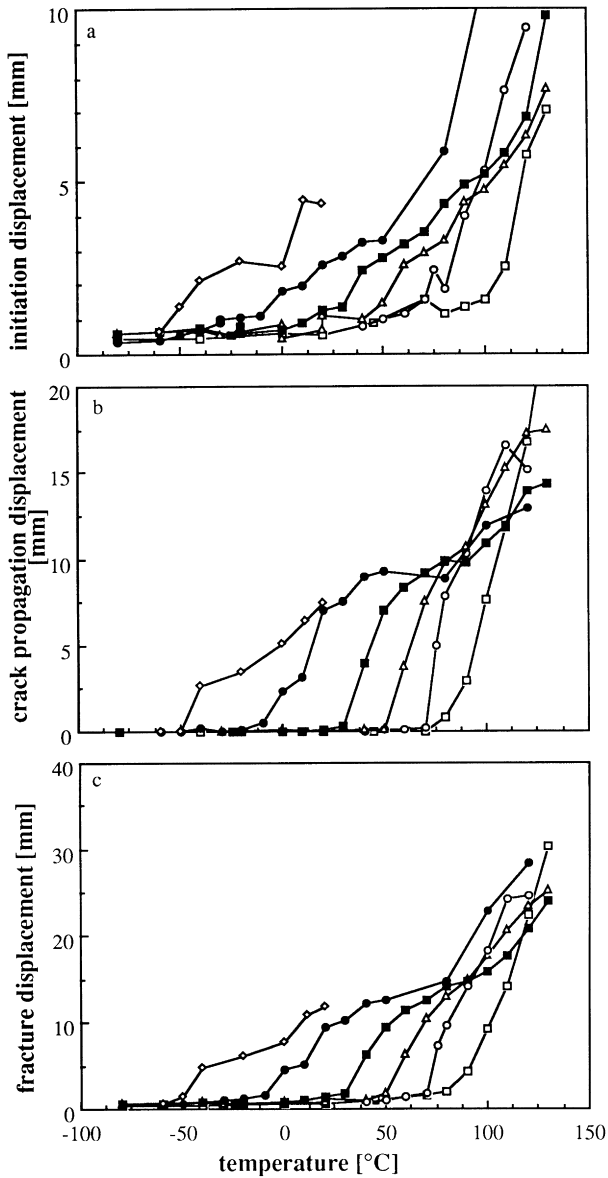


Fig. 14. Displacement during SEN tensile tests at 1 m/s as a function of the temperature for pure polypropylene and the PP-EPDM blends. Rubber content (vol.%): □, 0; ○, 5; △, 10; ■, 20; ●, 30; and ◇, 40.

CPE start to rise and the fracture type changes from brittle to ductile. The rise is at the onset gradual. With increasing rubber content the CPD and the CPE curves shift to lower temperatures. Over the studied temperature range the CPD and the CPE increase with rubber content. A question is to what extent the deformation in the initiation stage affects the crack propagation process.

The total fracture displacement and fracture energies are given in Figs. 8(c) and 9(c). It can be seen that the major contribution for the total strains and total energies in the ductile region come from the CPD and the CPE. The transition from brittle to ductile in the total fracture curves are not so sharp and it is better to use the CPD or the CPE.

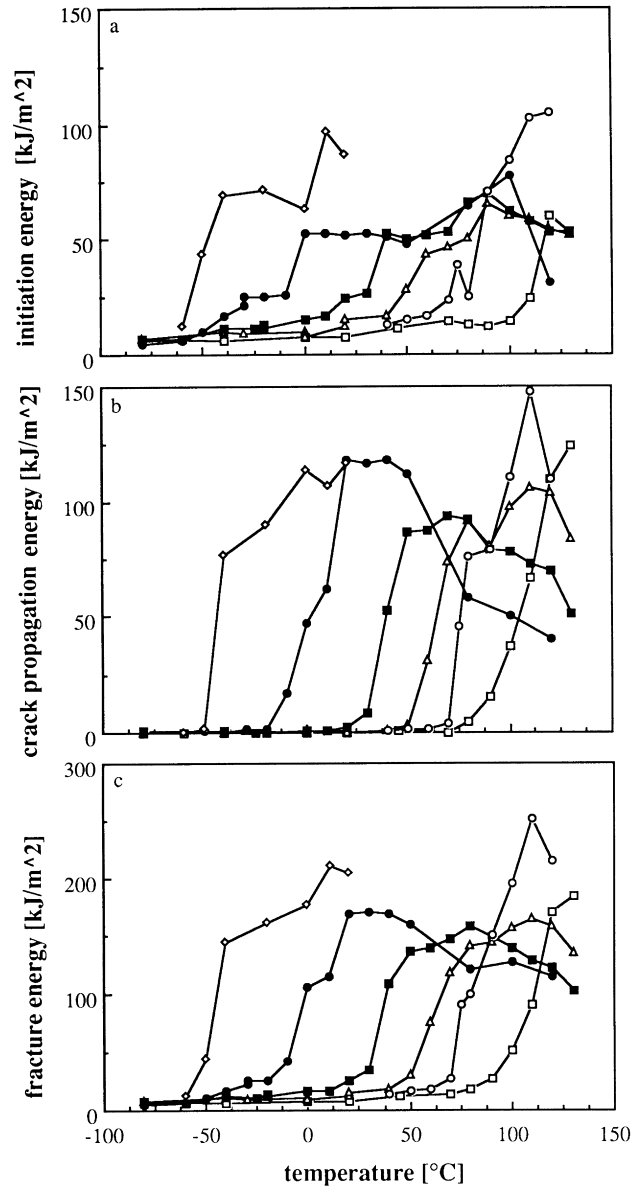


Fig. 15. Supplied energy during SEN tensile tests at 1 m/s for pure polypropylene and the PP-EPDM blends. Rubber content (vol.%): □, 0; ○, 5; △, 10; ■, 20; ●, 30; and ◇, 40.

3.4.1. Brittle–ductile transition at 1 mm/s

The onset of this rise in CPD and CPE we call the brittle–ductile transition. At this low test speed the transition from brittle to fully ductile is as we have seen in Figs. 6 and 8(b) over a temperature range. Similar gradual brittle–ductile transitions low speeds have been observed on polyamide–rubber blends [48] and on polycarbonate [49]. The transitions can equally well be determined from the CPD as from the CPE. With rubber content the curves and the brittle–ductile transition shift to lower temperatures (Figs. 8(b) and 9(b)). The brittle–ductile transition temperature (T_{bd}) as a function of rubber content is given in Fig. 10.

The T_{bd} decreases with increasing rubber content. For the higher rubber contents the T_{bd} is well below the glass

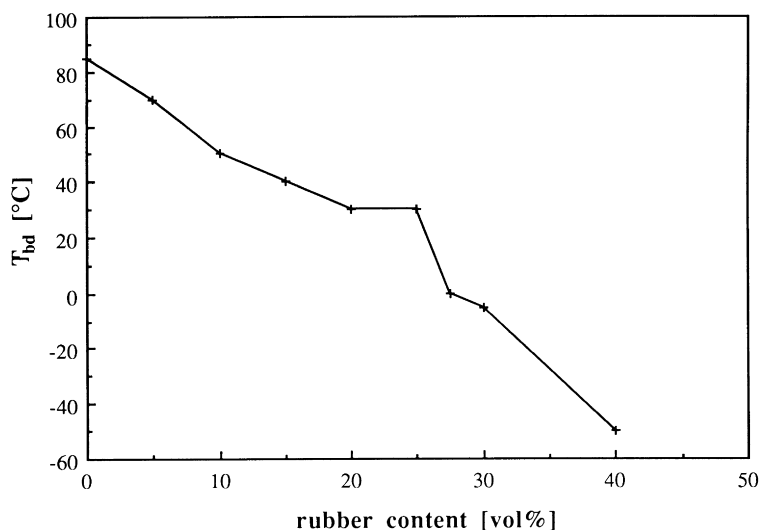


Fig. 16. The brittle–ductile transition temperature (T_{bd}) at 1 m/s versus the rubber content for the PP–EPDM blends.

transition temperature of the PP (-5°C) and is for the 35% blend at -45°C which is near the T_g of the EPDM. As the particle size at low test speeds has little effect on the T_{bd} no correction has to be made for the difference in particle size in the blends (Table 1).

It is interesting to look at the initiation displacement at the brittle–ductile transition (ID_{bd}) as a function of the rubber content (Fig. 11). The ID_{bd} is high for PP and decreases with rubber content. Just before crack initiation with a small amount of rubber (at high temperatures) necking over the entire cross section ahead of the notch is taking place. At ID_{bd} , at a high rubber content (at low temperatures) the plastic deformation is limited to the notch region. The T_{db} is not simply related to a value of the ID_{bd} .

3.5. SEN tensile test: 1 m/s

The fracture behaviour at high test speed was studied with SEN tensile tests at 1 m/s. The stress–displacement curves as function of temperature for the 5 vol.% blend are shown in Fig. 12. These curves are typical for all tested materials, including pure polypropylene. At this speed the transition from brittle to fully ductile deformation is sharp. In the case of ductile fracture, yielding during initiation at 1 m/s is considerably lower than at 1 mm/s (compare Fig. 12 with Fig. 6).

The maximum stress for some blends as a function of the temperature is shown in Fig. 13. The stress is the net cross-sectional stress, neglecting the stress concentration at the notch tip. The maximum stress in the blends increase sharply at -60°C followed by a decrease at a higher temperature. This sharp increase at -60°C coincides with the onset of voiding of the rubber particles, as we have seen in the SEN test at low speed. Due to the cavitation of the rubber the stress state ahead of the notch changes and plastic deformation can now take place more easily. The maximum

stress of pure polypropylene remains constant between -80 and -40°C , and decreases above -40°C with increasing temperature. From the observed maximum in the stress–temperature curves, plastic deformation, first only in the notch area and later over the whole cross section is possible. Surprising is that the maximum stress of the 20% blends is between -40 and 40°C higher than those of PP. The values for the 40% blend is between -60 and -20°C higher than the 20% blend and at higher temperatures is much lower as expected.

The displacement by initiation, crack propagation and fracture is shown in Fig. 14. The energy by initiation, crack propagation and fracture is shown in Fig. 15. For the sake of clarity, not all the tested blends are shown.

The ID (Fig. 14(a)) increases with increasing temperature and increasing rubber content. The ID and the IE (Fig. 15(a)) seem to increase in steps with temperature. At very high temperatures the IE curves passes through a maximum.

The onset of CPD (Fig. 14(b)) and the CPE (Fig. 15(b)) of the blends with temperature is sharp. After the step wise increase in CPD the further increase is gradual. At 80 – 90°C the blends show a second sharp increase. This second sharp increase may be due to neck formation during initiation. Pure polypropylene has a less sharp onset in CPD and CPE. The CPE of the blends passes through a maximum. A similar maximum is also present in polypropylene [12]. As a first approximation, the CPE is assumed to be proportional to the product of maximum stress and CPD. With increasing temperature, the maximum stress decreases and the CPD increases. Prior to the CPE maximum, the increase in the CPD relatively exceeds the decrease in maximum stress, the CPE increases. Beyond the CPE maximum the increase in the CPD is relatively lower than the decrease in maximum stress, the CPE decreases. At a temperature above the brittle–ductile temperature for all the polymers (e.g. 130°C) the CPE decreases with increasing rubber content. As most of

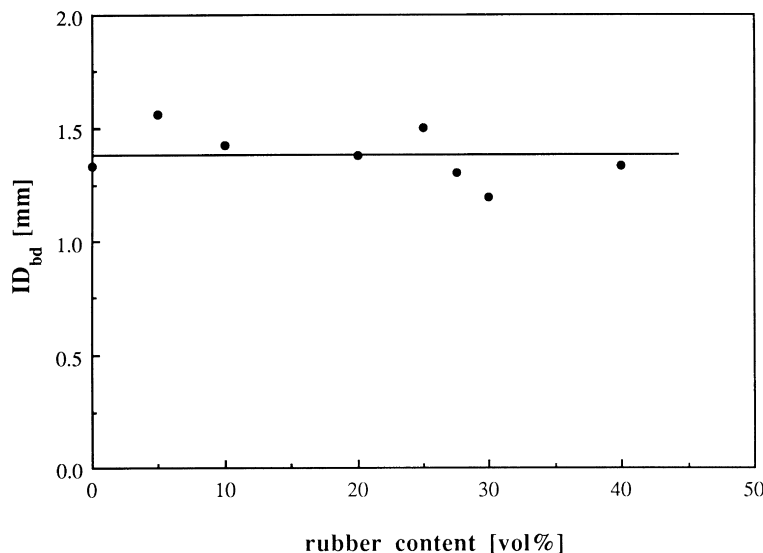


Fig. 17. Initiation displacement at the brittle/tough transition (ID_{bd}) versus the rubber content at 1 m/s for the PP-EPDM blends.

the plastic deformation comes from the deformation of the matrix and as in the blends less matrix is present the deformation energy is lower.

The total fracture energy curves seem to have S-shapes (Fig. 15(c)). With rubber content these curves are shifted to lower temperatures. The total fracture energy comes largely from the CPE. If the deformation had been in bending like with the Izod test the relative contribution of the initiation is even smaller (Fig. 8). In bending most plastic deformation is limited to the notch region while in notch tensile the whole cross section ahead of a notch can yield before a crack is initiated.

3.5.1. Brittle-ductile transition at 1 m/s

As T_{bd} is taken the onset of CPD was determined by extrapolating the CPD temperature curve to zero. The T_{bd} as a function of the rubber content is shown in Fig. 16. The T_{bd} of this PP is at 85°C and decreases strongly with rubber content. For the 40% blend a T_{bd} of -50°C was observed, this is well below the T_g of PP (-5°C) and very near the T_g of the EPDM. The shift in T_{bd} from PP to the 40% blend with the Izod test is 135°C. At high test speed the T_{bd} is affected by the particle size [39]. The correction for the particle size in the Izod test to 0.9 μm is a 20°C shift of the blends with the low rubber contents. The PP is not affected and the 30 and 40% blends are already 0.9 μm and do not change either. From this it is expected that Fig. 16 is only a minimal change if corrected for the particle size difference.

The initiation displacement at the brittle ductile transition (ID_{bd}) is independent of the rubber content (Fig. 17). This suggests that the ID_{bd} is a possible criterion for the brittle-ductile transition as has been suggested before [13]. The ID_{bd} are lower than the values observed at 1 mm/s test speed and particularly lower than the value if the cross section ahead of the notch is necking. So it is not clear whether the initiation displacement is a criterion for the

ID_{bd} . Well above the T_{bd} the ID is much higher and necking is taking place here.

4. Conclusions

The yield stress and the modulus decreases linearly with decreasing rubber content. The rubber content has a very strong effect on the deformation behaviour of notched PP-EPDM blend.

At low test speed the brittle-ductile transition is a gradual transition. In the initiation phase considerable plastic deformation is taking place ahead of the notch. The deformation zone can be as large as the whole cross section ahead of the notch. This plastic deformation at the T_{bd} with PP and the blends with a small amount of rubber (<10%) is accompanied by a necking mechanism. In the blends with the higher rubber content the plastic deformation at the T_{bd} is limited to the notch region. In these blends the necking is now taking place well above the T_{bd} .

At 1 m/s test speed the brittle-ductile transition is a sharp transition. The initiation displacement at the brittle-ductile transition is small and independent of rubber content. In the initiation phase the deformation is limited to the notch region. The strain rates in the notched Izod impact test and in the SEN tensile test at 1 m/s are comparable. The T_{bd} as determined by a SEN tensile test at 1 m/s (SEN T_{bd}) is compared to the value obtained from a notched Izod impact test (Izod T_{bd}) in Fig. 18. For pure polypropylene the SEN T_{bd} is lower than the Izod T_{bd} . The fact that the Izod T_{bd} is higher can be expected, since the stress during a notched Izod impact test is more triaxial than the stress during an SEN tensile test. This is attributed to the bending moment at the notched Izod impact test. Surprisingly, the Izod T_{bd} of the blends is considerably lower than the SEN T_{bd} .

The SEN T_{bd} at low test speeds as compared to the SEN at

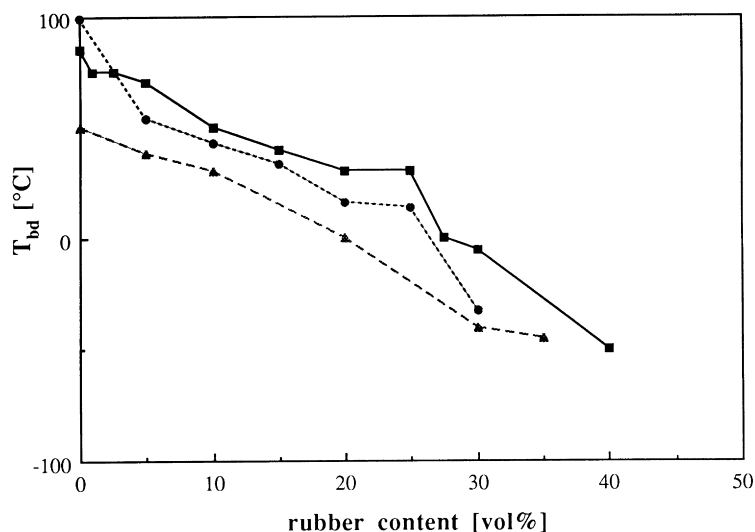


Fig. 18. The brittle–ductile transition temperature (T_{bd}) as a function of the rubber content for the PP–EPDM blends at various fracture tests: ●, notched Izod; ■, SEN tensile 1 m/s; and ▲, SEN tensile 1 mm/s.

high test speeds is over the whole rubber content range 20–30°C lower (Fig. 18). This 20–30°C shifts hide the fact that the transition at low speeds proceeds differently and that at high test speeds significant temperature rise is taking place. At high test speeds the deformation becomes adiabatic and the heat generated by the plastic deformation warms up the plastic region considerably [50].

Polypropylene can effectively be toughened by blending with EPDM. The brittle–ductile transition temperature (T_{bd}) decreases with 40% rubber to -50°C which is comparable to PP with a decrease of 135°C . The T_{db} on notched samples of the higher concentration blends are much lower than the T_{bd} of PP unnotched. Thus the function of the rubber is much more than to reduce the notch sensitivity of the sample. The function of the rubber is thus next to relieving the volume strain and thereby the multi-axial stress state, also to act as a stress concentrator which enhances the shear banding.

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