Compatibilization of PP/PE Blends and Scraps with Royalene: Mechanical Properties, SAXS, and WAXS

T. Nedkov, F. Lednický, M. Mihailova

Institute of Macromolecular Chemistry, Academy of Sciences of the Czech Republic, v. v. i. 162 06 Prague 6, Czech Republic

ABSTRACT: PP/PE 93/7 model virgin blends and recycled scraps were compatibilized with Royalene (EPDM/PE 65/35 blend) and mechanically tested. No differences in impact and tensile properties between them were found. However, the tensile-impact strength increased almost twice with 10%-compatibilized sample in comparison with uncompatibilized ones. The yield stress of blends containing 10% Royalene decreased to 75–80% of the original value. This effect is in agreement with microhardness measurements; the increase in the compatibilizer content causes softening of the blend. The elongation at break and elongation at yield do not depend on the compatibilizer concentration. The compatibilizer does not influence the degree of crystallinity (WAXS data) of the blends either. Vickers microhardness is in good agreement with Tabor's relationship. The differences between long periods of HDPE in Royalene and LDPE in PP/PE blends (SAXS) proved PE/EPDM interaction. The interaction plays a key role in the toughening of PP/PE blends. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 226–233, 2008

Key words: polyolefins; blends; compatibilization; recycling; morphology

INTRODUCTION

Toughness, tensile strength, and hardness rank among most important mechanical properties of polymer materials. The ratio between the price of materials and their mechanical characteristics determines the use of virgin and recycled plastics in everyday life. X-ray analysis gives information on structure and often helps to explain mechanical behavior of plastics. In the present article, mechanical and X-ray methods are combined to examine PP-rich PP/PE blends and scraps. Morphology of these systems was described elsewhere.[1]

The improvement of mechanical properties of PP-rich PP/PE blends using EPDM rubber has been the subject of many studies. With increasing EPDM content, impact strength increases,[2–6] while the yield stress decreases,[2,3,5] the blends become softer, and their microhardness decreases.[7] Used originally for the purpose of metallurgy, microhardness was also applied to polymers and associated with the yield stress.[8–11]

A peculiarity in WAXS diffraction pattern of PP/PE blend is the coincidence of the strongest PE peak 110 with the composed reflection 111, 131, 141 of PP. The change in intensity and shape of PP and PE reflections can provide information about the crystalline regions of both polymers in the blend. SAXS curves of PP/PE allow the determination of the long period of crystalline lamellae.

The aim of this study is to present additional information on mechanical properties and X-ray analysis of the same blend systems as in the previously published article,[1] in which the complicated phase structure was interpreted (flake structure as arrangement of the crystalline regions in the dispersed phase). It is intended to show that despite the complicated morphology the properties of the blends are commensurable with the other systems containing EPDM rubbers, and that virgin and recycled systems of the same composition practically do not differ from each other in mechanical properties, and that even for these systems microhardness can represent their mechanical behavior. All these specifics are important both for the industrial significance (re-usage of the PP/PE blends of the industrial waste, measurement of actual mechanical behavior of the real products that have shapes not allowing application of classical measuring techniques) as well as for the basic research (phase structure after repeated

processing, possibility of microhardness measurements of heterogeneous polymer blends).

EXPERIMENTAL

Materials and blend preparation

Compression-moulded blends of model (virgin) material components and industrial recyclates were examined. The virgin materials were low-density polyethylene (LDPE) PFS 4020 and polypropylene (PP) R 401 were received from Terplast (Poland). The compatibilizer Royalene IM 7565 (Uniroyal Chemical Co., Naugatuck, CT) is a blend of Royalene 580HT and HDPE, weight ratio 65 : 35. Royalene 580HT is an ethylene-propylene copolymer with the ratio (E/P) 52/48.

Industrial recycled materials are 93/7 mixtures of the same types of PP and PE (received from Terplast, Poland), to which is added: 0.033% Irganox 1010, 0.067% Irgafos 168, and 0.1% calcium stearate.

The model and recyclate samples are prepared from the virgin and industrial material in the ratio PP/PE 93/7, to which 0, 1, 2.5, 5, 10 wt % Royalene compatibilizer was added. Selection of the compatibilizer concentrations (same as in the previous article[1]) was adapted to compare the results directly with those published,[4] dealing with another PP/PE/EPDM blends, EPDM concentration having 2.5, 5, and 10%. For mixing, a Brabender plasticorder with W50EH chamber was used. Samples were prepared as square plates with 1.3–1.5 mm thickness.

Details of the used materials and sample preparation are described elsewhere.[1]

Mechanical experiments

Impact strength

The tensile impact strength was measured at 23°C using a Zwick 5102.10/00 tester (pendulum length 225 cm), equipped with a special fixture for test specimen according to ISO 8256. The samples were machined in a dumb-bell shape. A preliminary non-breaking test was performed to evaluate the kinetic energy of holder and specimen. The reading value of pendulum was subtracted from the value obtained in the real break experiment. The specimens were cut from pressed plates. The presented values were averaged as the arithmetical mean of 10 measurements of test pieces.

Tensile properties

Tensile experiments were performed at room temperature with Instron 6025 according to the ISO 527 standard. The crosshead speed was 50 mm/min.

The reported values were averaged from five measurements.

Microhardness

Microhardness according to Vickers was measured by indentation of a defined pyramid into a sample. Microhardness was calculated by the formula (1)

$$MH_v = 2P \sin (\theta_{pyr}/2)/d_{pyr}^2 = 1.854P/d_{pyr}^2 \qquad (1)$$

(P is the applied load in N, d_{pyr} is the size of the diagonal at the base of the imprinted pyramid in mm) and the values of MH_v are obtained in MPa. The diagonal at the base of pyramid was measured with a microhardness tester mhp-160 combined with a microscope NU-2, Zeiss. The pyramid diagonal was measured with accuracy 0.4 μm. The coefficient 1.854 is a geometrical factor, which is associated with the tip angle $\theta = 136°$ of the indentor. Loads of 0.20, 0.49, and 0.78 N were used. Diagonal values were averaged from 10 measurements. The size of the indentation pyramid was 1 mm at the base and 30 s loading time was chosen. To eliminate elastic effects during deformation, microhardness was determined as the slope from the dependence of P on d_{pyr}^2.

The errors in the graphs of mechanical characteristics represent standard deviations.

X-ray experiments

SAXS

CuKα radiation, with wavelength λ = 1.54 Å is used. Kratky camera has a 60-μm entrance slit and 42-cm flight path. Ni-filtered radiation was registered with a position-sensitive detector, the resolution of which is ca 0.1 mm. The intensities were taken in the range of the scattering vector $q = (4\pi\sin \theta)/\lambda$ from 0.005 to 0.2 Å^{-1}.

WAXS

Wide-angle diffractograms were taken on a HZG4A power diffractometer. CuKα radiation with wavelength λ = 1.54 Å was used. The radiation was monochromatized with a Ni filter and pulse-height high-voltage analyser and registered with a scintillation counter. Diffractograms were taken in the range of scattering angles 2θ = 5°–40°.

RESULTS AND DISCUSSION

Mechanical

The tensile impact strength increases with increasing compatibilizer content for both model and recycled
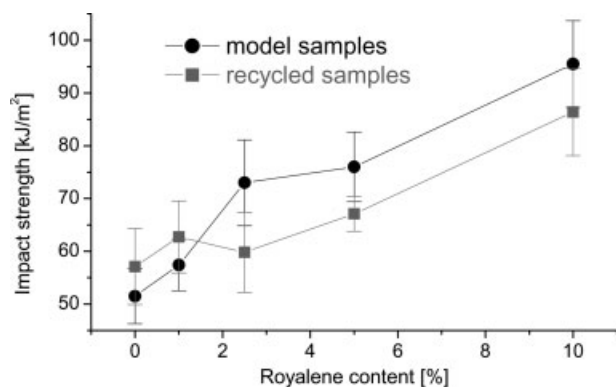
Figure 1 The dependence of tensile impact strength on the Royalene content for model and recycled samples.

blends (Fig. 1). Up to 2.5% of Royalene, the increase in tensile impact strength is within the limits of experimental error. A notable increase above 5% and about double increase at 10% Royalene content was observed. These results are similar to those of Kruliš et al.,[2] who investigated the blends PP/(EPDM/PE) with 75% of PP, with the EPDM/PE ratios 0, 5, 10, 15, 20, and 25%. Omitting concentrations of EPDM less than 5% EPDM, they found that the blend with 10% rubber content exhibits twice as high impact strength than that of uncompatibilized blend. When a higher amount of EPDM was added, the impact strength slightly decreased. The authors pointed out that at the PE/EPDM ratio 60/40, both the polymers form interpenetrating phases, which improve toughness of the blend. Blom et al.[3] examined PP/HDPE/EPDM blend 90/10/5. They also obtained a double increase in impact tensile strength in comparison with neat PP. Fortelný et al.[4] obtained a double increase in the impact strength of the PP/LDPE 80/20 on the addition of 10% EPDM as well. Choudhary et al.[5] report that the Izod impact strength increases five to eight times for a ternary blend (PP/HDPE)/EPDM, where the PP/HDPE ratio is kept constant (90/10) and the rubber content ranges between 0 and 25%. The Izod impact strength has a maximum at 20% of EPDM. These results, in particular the impact strength increasing five to eight times, are different from all those results referred to above but similar to the results of Zheng et al.[6] The authors of the two last-mentioned article investigated the notched impact properties of the blend, unlike the previously considered science teams, who used samples without notches. Notched samples are less sensitive to impurities and nonhomogeneities, as the notch restricts the volume under stress and consequently, reduces the number of weaker points caused by any contamination. Although the absolute value of notched impact strength is lower than the value of impact strength, its relative increase is higher with compatibilization.

From our results and all the aforementioned studies, we can deduce that the compatibilizer Royalene substitutes neat EPDM in the ternary blend with the similar success. Royalene improves the impact strength of PP/PE blends about twice and compatibilizes the recycled blends as well.

The yield stress (Fig. 2) monotonically decreases on increasing the compatibilizer content. The decrease at the 10% content of Royalene is about 28% with virgin blend and about 30% with the recycled blend.
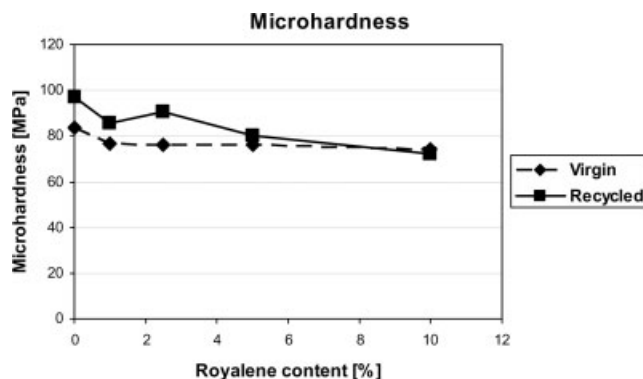
The decrease in the yield stress with increasing Royalene content is monotonic. Up to 2.5% of Royalene, the decrease is slight, almost in the limits of experimental error. As Royalene is softer than PP and PE, the irreversible plastic deformations, the beginning of which is characterized by the yield point, start to occur earlier with increasing rubber content in the blend. Blom et al.[3] found that 5% of EPDM causes only a slight drop in the yield stress ($\sim$ 10%). Kruliš et al.[2] showed that addition of EPDM rubber to PP monotonically decreased the yield stress of the mixture, while addition of EPDM to a PP/HDPE blend has almost no effect on the yield point. Choundhary et al.[5] found that the yield stress decreases monotonically with increasing the rubber content up to 25% of rubber, which is the highest EPDM concentration in their study. The whole drop of the yield stress for this EPDM concentration ranges amounts to about 30%.

It can be stated that despite the fact that Royalene decreases the yield stress of PP/PE blends, it can replace other EPDM compatibilizers as far as the yield stress is concerned.

The yield stress results (Fig. 2) can be compared with microhardness measurements (Fig. 3). Microhardness decreases with the Royalene content. The relative decrease in microhardness at the 10% Royalene compatibilizer content is 22% for the virgin material, and 26% for the recycled blend. The decrease
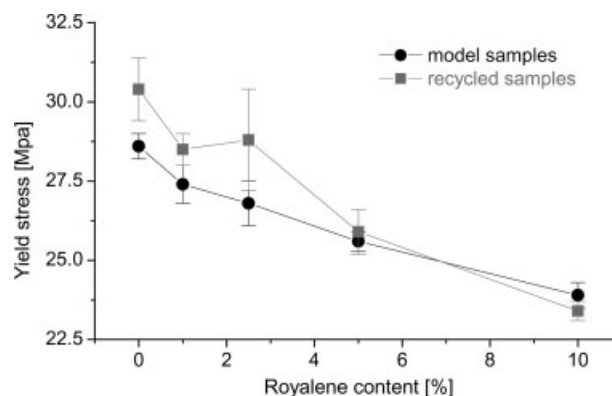
Figure 2 The dependence of yield stress on the Royalene content for model and recycled blend.

Figure 3 Dependence of microhardness on the Royalene content.

is in full accord with about the same decrease in the yield stress.

Microhardness determined in a "classical" way, i.e., from the individual pyramid diagonal and the corresponding load depends on the applied load and, therefore, it provides no reliable microhardness values with polymers, in general. The physical reason is based on the fact that polymers are not fully plastic materials. After load removal, recovery of the polymer always takes place. $P - d_{pyr}$ dependence is influenced by elastic contributions to the resulting dimensions of the pyramid diagonal. Closer to the true microhardness values are those obtained with higher loads P, but a better approximation is the determination of microhardness from the $P - d_{pyr}^2$ dependence. That approach is justified when the dependence is linear and the elastic contribution to the deformation does not depend on the applied load. The latter condition is obviously fulfilled as the fully elastic deformation is limited by the existence of yield stress point. The linearity condition was checked in our measurements using three loadings (0.20, 0.49, and 0.78 N). In most cases of virgin blends, linearity was fair with the exception of the material with 10% of Royalene. For recycled blends, a higher scatter was found. However, even taking the scatter into microhardness calculation, the results did not differ by more than 10%.

Correction for the instant elastic response using several loads is legitimate under the above conditions as it eliminates the dependence of measured values on the applied load. It was used by Flores et al.,[12–14] Balta Calleja et al.[15] and other authors investigating various polymers. Dependence of the microhardness results on the indentor penetration in the samples was documented by Zhorin et al.[7] also for neat PE and PP. Similar results for various polymers were published.[16] Effects of load and loading time on the hardness of an acrylic polymer were studied in a wide range of loads and loading times.[17] Calculation of the Vickers microhardness

from the highest applied load was used for epoxy composites.[18]

Correlation of the microhardness MH_v for a pyramid indentor with the yield stress σ_y follows Tabor's relationship

$$MH_v = 3\sigma_y \tag{2}$$

This is the basic relation between indentation hardness and bulk properties. It stems from the classical theory of plasticity, being originally formulated for metals, and is rigorously applicable only to an ideally plastic solid showing no recovery ([19], c.f.[11]). The relationship is also applied to polymers.[9]

Comparison of microhardness with tensile yield stress (c.f.[8]) for our samples shows a very good agreement with Tabor's relationship (Fig. 4): The ratio of microhardness to yield stress exhibits only small differences for all Royalene concentrations in both virgin and recycled blends. It does not depend on the Royalene content differing from the value 3 by less than 7%. However, there is a small systematic shift depending on the polymer: recycled materials exhibit higher MH_V/σ_y values than those with the virgin materials.

Validity of Tabor's relationship was reported for polyethylene blends.[20] On the other hand, deviation of measured values from 3 was reported for polymers, depending on the material structure and properties. Flores et al.[8] showed that for modified chain-extended PE this ratio can vary between 2 and 4. In their study, the value of 2 is appropriate for chain-folded PE, while the chain-extended PE has a value of about 3. Though the differences with our materials are much smaller, we believe that the reason for the difference is worth being studied.

In stress–strain experiments, the ultimate tensile strength and elongation were also measured (Figs. 5
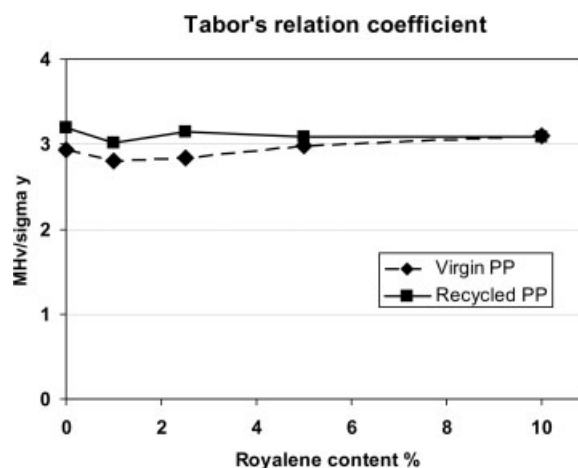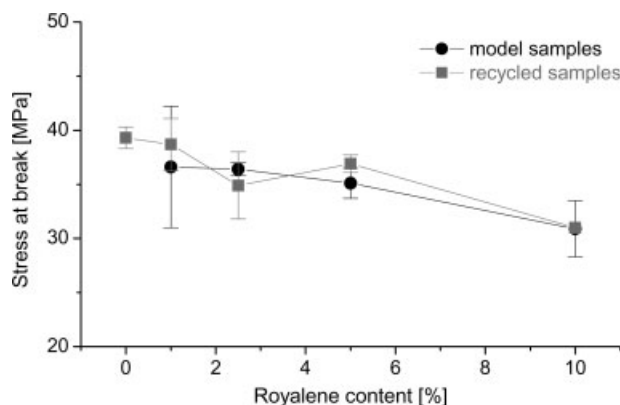

Figure 4 Tabor's correlation between microhardness and yield stress depending on the Royalene content.
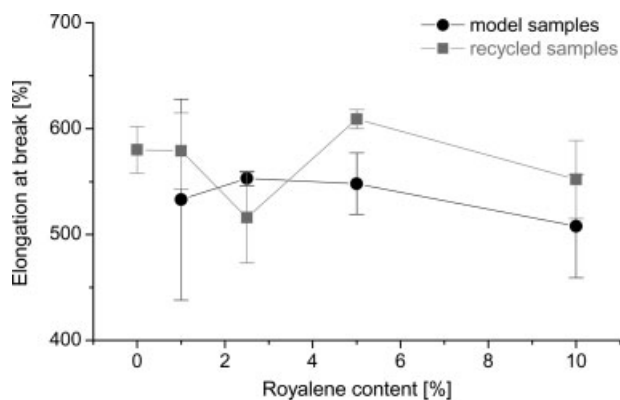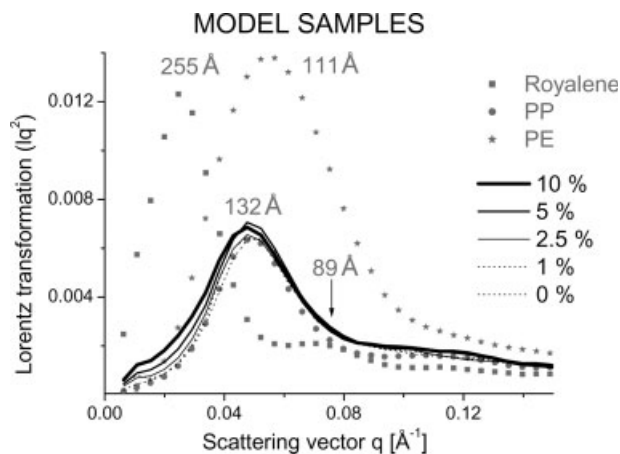
Figure 5 Dependence of the elongation at break on the Royalene content for model and recycled samples.

Figure 7 Lorentz-corrected SAXS curves for model samples.

and 6). Elongation at break (Fig. 5) does not depend on the Royalene content, with the exception of the point that corresponds to 2.5% Royalene. For recycled blend there is a slight drop at this point. No further conclusion can be drawn for the model blends, as the sample with 1% Royalene shows quite a large experimental error. The elongation at break is not influenced by EPDM for blends of PP/HDPE/EPDM of the composition between 90/10/0 and 85.5/9.5/5 (Ref. 3). For this system, Blom et al.[3] point out a slight drop in elongation at break at 1 % of compatibilizer. Choudhary et al.[5] report the minimum of elongation at break at about 5% of EPDM for the PP/HDPE/EPDM blends, of constant ratio 90/10 and variation of EPDM from 0 to 25%. As they did not measure the intermediate concentrations of compatibilizer, it is impossible to elucidate the minimum value more precisely. None of the above articles explains that. Further investigations are needed seeking the answer. There are not many reports in the literature about tensile characteristics of the investigated blends, as EPDM is not intended to improve the blend properties. It can be concluded

that not even Royalene does so. The stress at break (Fig. 6) tends to decrease with increasing Royalene content. However, the decrease is very small, almost in the limits of experimental error. These results are similar to those of Blom et al.[3]

Small angle X-ray scattering (SAXS)

SAXS experiments of model and recycled samples are given in Figures 7 and 8. There is practically no difference between the model and the recycled samples according to SAXS experiments. SAXS conclusions are drawn from Lorentz-corrected curves (Figs. 7 and 8). Long periods of PP and PE lamellae are 132 and 111 Å, respectively. They coincide with the most frequently published values. The SAXS curve of Royalene shows its ordered structure. Royalene is characterized by two maxima, at 255 and 89 Å. No SAXS data for Royalene were found in literature. As Royalene contains about 33% of HDPE, it could be expected that Royalene exhibits some features of PE. Actually, the PE/EPDM compatibilizer

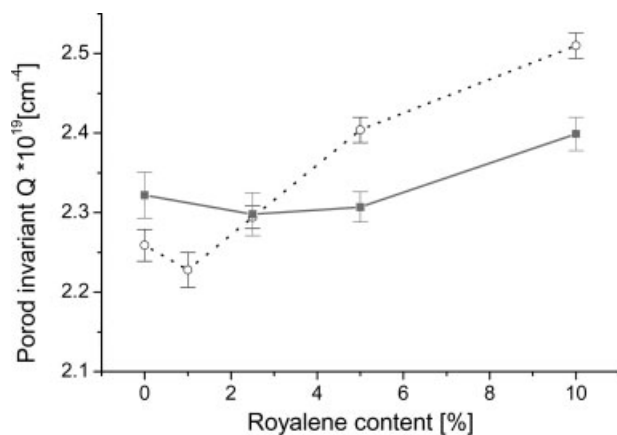**Figure 6** Dependence of the stress at break on the Royalene content for model and recycled samples.
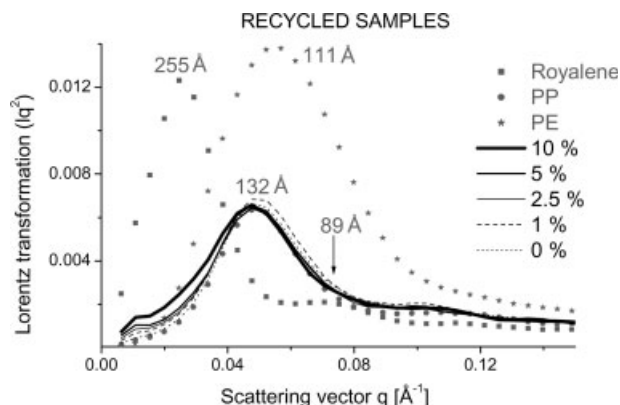
Figure 8 Lorentz-corrected SAXS curves for recycled samples.

Figure 9 Porod invariant in dependence on the Royalene content of model blends (open circles) and recycled blends (hatched squares).

has completely different ordered structures in comparison with PE, as it follows from the positions of the observed two maxima. This finding is in a good agreement with the determined phase structure of Royalene.[1] In SAXS curves of blends, PP as the major phase predominates. They almost repeat the SAXS result obtained for neat PP. Nevertheless, at very small angles, a small influence of the Royalene can be reported. This is because of the increasing Royalene concentration and appearance of maximum at 255 Å.

Lorentz correction enhances the maxima but strongly suppresses the intensity at lower angles. However, even if the correction is applied, still the influence of Royalene on the blend curves can be detected.

The Porod invariant Q was calculated in order to evaluate the effect of Royalene content on the scattering power of the samples.

It is defined as follows:[21]

$$Q = \frac{1}{2\pi^2} \int_0^\infty q^2 \frac{d\sum}{d\Omega}(q)dq \qquad (3)$$

where $d\sum/d\Omega(q)$ is the smeared intensity curve converted to absolute scale.

The Q values versus the compatibilizer content are shown in Figure 9.

The scattering power of the blends slightly increases at the 10% Royalene content. This effect is more significant for the model samples than for the recycled ones. A special procedure was applied to determine the experimental error of the smeared intensity. The determination of Porod invariant is performed by calculation of the integral from zero to infinity. The tail of this curve (wide-angle region) fluctuates and can obscure the effect of Royalene on the examined area at very small angles. Because of

that, the tail is replaced by two curves – taking correspondingly the minimal and maximal values of intensity during fluctuation. In this way, the maximal and minimal tail curves are created, in addition to the already existing main average tail curve. These curves are not extended to the full range of scattering angles, as the scattering power at a farther part of SAXS curve is not significant. The maximal and minimal fluctuation tail curves allow obtaining the maximal and minimal values of Porod invariant, which give the possibility to determine its experimental error.

It can be seen from Figure 9 that the increase in Porod invariant exceeds the limits of experimental error. This indicates an increase in the scattering power of the sample with Royalene content at lowest angles, which points to changes in the phase structure of the blends. The connection of the Porod invariant to mechanical properties of the blends can be seen in the changes in their flake structure (c.f.[1]). We believe that Porod invariant can be an effective help in the interpretation of supermolecular structures disclosed by other morphological techniques.

Wide-angle X-ray scattering (WAXS)

The WAXS results are presented in Figures 10 and 11 for the model and recycled blends, respectively.

The WAXS diffraction patterns for PP and PE are known.[22] PP in our samples mainly consists of the α-phase, with reflections 110, 040, 130, and the composed reflection 111, 131, 141. Neat PP also contains an insignificant amount of β-phase, which appears in the weak peak at the strongest β-phase reflection 300. PE is characterized by reflections 110, 200, and 020, the last being the weakest. The HDPE component of Royalene crystallizes, which is confirmed by the identical peak positions of Royalene and PE dif-
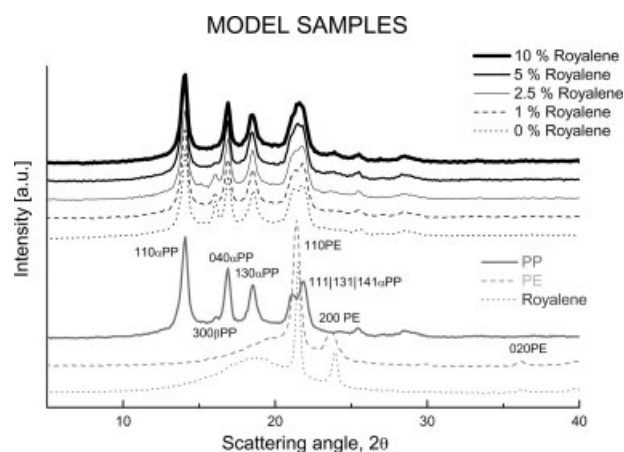
Figure 10 WAXS curves for model blends with 0–10% Royalene and for neat polymers.

fractograms. As can be expected, a significant part of Royalene remains amorphous. The result is the wide amorphous halo with maximum at about $2\theta = 18°$. The position of the Royalene halo is slightly shifted to lower angles with respect to that of PE. It is clear from the graphs that compound reflection of crystalline PP 111, 131, 141 changes its shape. This reflection, the strongest PE 110 reflection, and the reflection of Royalene at about the same scattering angles $21°$–$22°$ form a single peak. This phenomenon is also reported by Martins et al.[23] However, in contrast to Royalene, the EPDM rubber used by the authors is an amorphous polymer, so the broadening and an increase in the compound PP peak is caused by the crystalline nature of HDPE only.

The X-ray degrees of crystallinity α_c, calculated by the formula[24]

$$\alpha_c = I_c/(I_c + I_{am}) \qquad (4)$$

where I_c and I_{am} are integral intensity of crystalline and amorphous regions, respectively, are presented in Table I.

The error due to the calculation of degree of crystallinity does not exceed 3%. The amorphous halo of PP was constructed after Weidinger and Hermans.[25] The amorphous halo of PE was not taken into consideration, as its contribution to the amorphous halo of the blend is estimated to be close to the calculation error. It can be concluded that the degree of crystallinity for all blends is about 46% and does not change with increasing Royalene content. These results are similar to those of Hlavatá and Horák.[26] They found that the degree of crystallinity of PP/HIPS blends with SBS compatibilizer slightly decreases and the decrease is about 3%.

Almost the same value of X-ray crystallinity seems to contradict the decrease in microhardness with increasing Royalene content. Similar situation was

TABLE I
X-Ray Degree of Crystallinity for Model and Recycled Blends

Royalene content (%)	Model blend (%)	Recycled blend (%)
0	44	48
1	46	46
2.5	46	47
5	47	45
10	46	46

observed with modified ultra-high molecular-weight polyethylene.[27] Very slight changes in crystallinity were accompanied by marked changes in microhardness. The explanation takes into account changes in the quality of the amorphous component. In our samples, the softer Royalene component is obviously responsible for the notable changes in microhardness but almost negligible changes in crystallinity.

CONCLUSIONS

1. The model (virgin) and industrial recycled PP/PE blends of composition 93/7 show practically identical mechanical properties. This conclusion allows a new utilization of investigated industrial recycled polyolefins.
2. The impact strength of blends increases with increasing Royalene content. With 10 wt % of Royalene, the impact strength is about twice as high as with the noncompatibilized blend.
3. Addition of 10% of soft Royalene decreases yield stress to 3/4 of the value obtained for the uncompatibilized PP/PE blend. Vickers microhardness reflects well the yield stress, with the values of Tabor's relationship equal to 3.
4. The elongation at break and elongation at yield do not change by compatibilization while the stress at break slightly decreases.
5. X-ray diffraction studies reflect differences in supermolecular structure, which are responsible for the differences in mechanical properties. WAXS diagrams show that the HDPE part of Royalene crystallizes in the same unit cell as neat PE does. However, the Royalene periodicity (255 and 89 Å) in SAXS curves differs from that of neat PE (111 Å). This determines a different long period, which is probably due to PE/EPDM interactions.
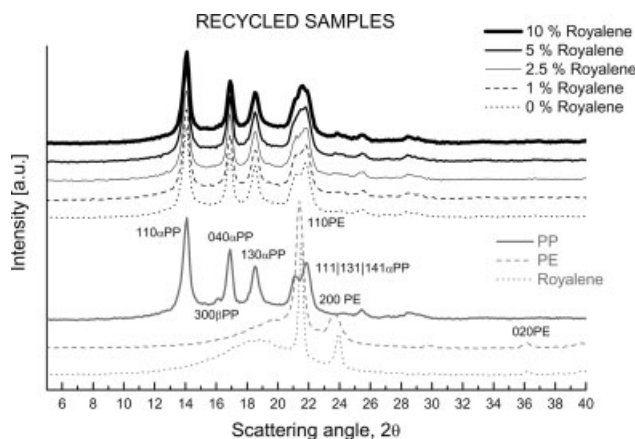
Figure 11 WAXS curves for recycled blends with 0–10% Royalene and for neat polymers.

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