Wollastonite-Reinforced Polypropylene Composites Modified With Novel Metallocene EPR Copolymers. II. Mechanical Properties and Adhesion

Iztok Švab,[1,2] Vojko Musil,[1] Anđela Pustak,[3] Ivan Šmit[3]
[1]*University of Maribor, FEB Maribor, Institute of Technology, Razlagova 14, 2000 Maribor, Slovenia*

[2]*ISOKON, Production and Processing of Thermoplastics, Ltd, Mestni trg 5a, 3210 Slovenske Konjice, Slovenia*

[3]*Ruđer Bošković Institute, Bijenička 54, 10002 Zagreb, Croatia*

Mechanical properties and adhesion phenomena of isotactic polypropylene/wollastonite/metallocene propylene-ethylene copolymers (iPP/W/EPR) composites were studied as a function of metallocene propylene-based copolymers content from 0 to 20 vol%. The composites with different surface treated wollastonites and two types of EPR have shown similar behavior of most mechanical properties except elongation at break and impact strength respective of the difference in some characteristics of used EPR elastomers. The increase and the difference in elongation at break could be explained by renewed spherulitic morphology of the iPP matrix. Stronger interactions between EPR-1 and two used types of wollastonites than between EPR-2 and corresponding wollastonites concluded from the surface properties led to the difference in impact strength behavior. The determined mechanical properties confirm the assumption coming out of structural investigations that metallocene EPR elastomers are rather efficient impact modifiers than encapsulation compatibilizers for the iPP/wollastonite composites. POLYM. COMPOS., 30:1091–1097, 2009. © 2008 Society of Plastics Engineers

INTRODUCTION

Primary production and use of polymer-matrix composites have been grown intensively due to favorable cost/performance ratio. Further growth of particulate-filled polymers was directed by transformed goal due to increasing demands to fulfill a functional role of fillers in engineering plastics [1–3]. The fillers affect ultimate mechanical properties (i) directly, as rigid inorganic particles with determined properties (shape, size, and modulus) and (ii) indirectly, by affecting the crystallization processes in polymer matrix and ultimate supermolecular structure of semicrystalline polymer. Deterioration of impact properties by introduction of filler into polymer matrix could be abolished by addition of appropriate elastomer. To obtain a good stiffness/toughness balance, an elastomer should be efficient as an impact modifier as well as an adhesion promoter between polymer and mineral filler.

Among thermoplastics, isotactic polypropylene (iPP) offers a wide spectrum of favorable properties and cost-effectiveness that could be improved by modifying with inorganic fillers and/or elastomers. Mineral wollastonite, $CaSiO_3$ (calcium metasilicate) is, among commonly used fillers (talc, calcium carbonate, glass fibers, mica, glass beads), suitable reinforcing filler for the iPP due to its acicular crystal habit (high aspect ratio), relatively high hardness, and to its nucleation ability [1–3]. Thereby, the wollastonite increases tensile and flexural modulus as well as scratch and mar resistance [4–8]. Furthermore, the incorporation of wollastonite into iPP matrix changes the microdeformation mechanism of reinforced iPP [8]. Surface treatment of wollastonite enhances the interactivity with iPP matrix that may increase tensile strength, mar and scratch resistance, and improves impact strength of composites [9–12]. The efficiency of elastomeric copolymers as impact modifiers and/or compatibilizing agents between filler and matrix for the iPP/wollastonite composites have been rarely investigated [13–16]. Therefore, ethylene-propylene metallocene copolymers (EPR) with propylene being the major component (actually tough EP copolymers containing at least 70 mol% of propylene and less than 20 wt% of ethylene are uniquely soft and elastic)

Correspondence to: Ivan Šmit; e-mail: ismit@irb.hr

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were introduced in the iPP/wollastonite composites as impact modifiers and potential compatibilizers.

The aim of our investigation was to determine structure-mechanical properties relationship of the iPP/wollastonite/EPR composites with incorporation of two types of wollastonites and two metallocene EPR elastomers of different viscosity. In this article the effect of wollastonites and EPRs on the mechanical properties of the iPP/wollastonite/EPR composites are studied in correlation to their final structure.

EXPERIMENTAL

Materials

The materials used in this study were isotactic polypropylene (iPP), two types of wollastonite and two types of propylene-based metallocene copolymers with propylene being the major component. The iPP used for sample preparation was Moplen HP501L, Basell (melt flow rate [MFR] = 6 g/10 min, $\rho = 0.90$ g/cm^3, $M_n = 120{,}000$ g/mol). Applied mineral fillers were proprietary (combination of silanes) surface treated wollastonite (W1) Tremin 939 300 ZST, Quarzwerke (ρ = 2.85 g/cm^3, specific surface 1.2 m^2/g, d(50%) = 9 μm) and wollastonite surface treated with aminosilane (W2) Tremin 939 300 AST, Quarzwerke (ρ = 2.85 g/cm^3, specific surface 1.2 m^2/g, d(50%) = 9 μm). Two metallocene propylene-ethylene copolymers of ExxonMobil with different viscosity Vistamaxx-VM-1100 (MFI(EPR-1) = 4.5 g/10 min; ρ = 0.863 g/cm3; M_n = 92,900; M_w/M_n = 3.4) and Vistamaxx-VM-1120 (MFI(EPR-2) = 20 g/10 min; ρ = 0.863 g/cm3; M_n = 48,100; M_w/M_n = 2.66) were used as elastomers.

Sample Preparation

Binary iPP/wollastonite and ternary iPP/wollastonite/elastomer composites were prepared in an oil-heated Brabender 50WH kneading chamber. The iPP/wollastonite ratio was kept constant at 92/8 vol% and the elastomer of 2.5, 5, 10, and 20 vol% was added. The components were put into a chamber preheated up to 200°C with a rotor speed of 50 min^{-1} and were kneaded for 7 min. Upon that, the melt was rapidly transferred to a preheated laboratory press and compression molded into 1- (for tensile test) and 4-mm (for measurement of notched impact strength) thick plates. The pressing temperature was 220°C, pressure was 100 bar and the pressing time 14 min for 1-mm and 11.5 min for 4-mm thick plates. Afterwards, the plates were cooled down to room temperature in the air. The specimens for the tensile tests and notched impact strength were cut from the plates according to appurtenant standards.

Tensile Tests

Tensile properties (Young's modulus, yield stress, elongation at yield stress, tensile strength at break, elongation at break) were measured according to ISO 527 standard using Zwick 147670 Z100/SN5A apparatus at 23°C and strain rate of 2 mm/min. For each sample, five measurements were carried out and the average values were calculated within standard deviation of 5%.

Notched Impact Strength

Notched impact strength was measured by Zwick apparatus at 25°C according to Charpy test (DIN 53453). For each sample, 12 measurements were carried out and the average values were calculated within standard deviation of 5% as well.

Contact Angle Measurements

Surface free energies of materials were determined throughout the measurements of contact angles. Contact angles on the film of the iPP and EPRs as well as on plate-like compacts of the wollastonite fillers were measured on a DataPhysics OCA 20 Instrument. Sessile drops (2 μL) of test liquids: water (distilled twice λ = 1.33 μL cm^{-1}), formamide (p.a. 99.5%, Fluka) and diiodomethane (p.a. 99%, Aldrich), at 23°C were used for the advancing contact angle measurements. The contact angle was measured from 10 to 30 s after drop deposition. Average values of at least five drops on different places of the same sample were taken and standard deviation was always lower than 2°. Surface free energies of the iPP, wollastonite fillers, and EPRs were calculated by harmonic mean equation [17]:

$$\gamma_{\mathrm{lv}}(1+\cos\ \theta)=\frac{4\gamma_{\mathrm{s}}^{\mathrm{d}}\gamma_{\mathrm{lv}}^{\mathrm{d}}}{\gamma_{\mathrm{s}}^{\mathrm{d}}+\gamma_{\mathrm{lv}}^{\mathrm{d}}}+\frac{4\gamma_{\mathrm{s}}^{\mathrm{p}}\gamma_{\mathrm{lv}}^{\mathrm{p}}}{\gamma_{\mathrm{s}}^{\mathrm{p}}+\gamma_{\mathrm{lv}}^{\mathrm{p}}} \tag{1}$$

where γ^{d} was the dispersive and γ^{p} the polar component of the surface free energy (surface tension), γ_{lv} and γ_{s} were the surface tension of liquid and surface free energy of solid, respectively, whereas θ was the contact angle.

This evaluation method was integrated in the software (SCA 20) and automatically computer processed. The method requires the use of at least two test liquids with known surface tensions and their polar and dispersive components. Each additional liquid increases the accuracy of estimation. The surface tensions of the test liquids [18] used for contact angle measurements are presented in Table 1.

RESULTS AND DISCUSSION

Tensile Test

Stress–Strain Curves. Stress-strain curves of neat iPP and composites with different surface treated wollastonites and EPR elastomers are presented in Figs. 1 and 2. The introduction of wollastonite into neat iPP changes the character of fracture from relatively ductile (neat iPP) to

TABLE 1. Surface tensions (γ_{lv}) and their dispersive (γ_{lv}^{d}) and polar (γ_{lv}^{p}) components of test liquids.

Liquid[18]	Surface tension (mJ/m^2) γ_{lv}^{d}	γ_{lv}^{p}	γ_{lv}
Water	21.8	51.0	72.8
Formamide	39.0	19.0	58.0
Diiodomethane	50.8	0.0	50.8

a brittle fracture of binary iPP/wollastonite composites. The decrease in ductility is a result of relatively high stiffness of wollastonite filler superimposed by reinforcing effect due to its high aspect ratio. Low amount of EPRs (up to 5 vol%) added to binary composites reduces the stiffness and yield maximum and slightly increases elongation in comparison to the iPP/wollastonite composite. Further addition of EPR elastomers to the iPP/wollastonite composites additionally decreases yield maximum with less expressed necking and significantly increases the elongation. Although the influence of both EPR-1 and EPR-2 elastomers on stress–strain curves behavior of ternary composites seems to be quite similar, there are some differences in ultimate elongations. This similarity contrasts from observed distinguished influence of poly(styrene-*b*-ethylene-*co*-butylene-*b*-styrene) triblock copolymer (SEBS) and the SEBS grafted with maleic anhydride (SEBS-*g*-MA) on stress-strain behavior of the iPP/wollastonite/SEBS(SEBS-*g*-MA) composites [16, 19].

Young's Modulus. The influence of wollastonite and EPR elastomers on Young's modulus of the iPP composites is shown in Fig. 3. Expectable, the incorporation of 8 vol% of stiff needle like wollastonite particles into the iPP matrix has the reinforcing effect. Furthermore, the stiffness of binary composites may additionally increase by increasing the overall degree of crystallinity because of β-iPP phase nucleation ability of wollastonite [20]. Higher overall crystallinity of the iPP matrix might also increase Young's modulus due to stiffer crystalline than amorphous phase [21, 22].

Young's modulus of all ternary iPP/wollastonite/EPR composites decreases upon the addition of EPRs in similar way. Young's modulus noticeable decreases at lower amount of added EPRs (2.5 vol%), while higher amounts of added EPRs (above 2.5 vol%) to the iPP/wollastonite composites decrease Young's modulus gradually, almost linearly. Similar Young's modulus behavior of composites with both EPRs is also in contrast to observed differentiated effect of SEBS and SEBS-*g*-MA elastomers on Young's modulus behavior of these composites [16]. Young's modulus behavior of the iPP/wollastonite/EPR composites is very close to this one for the iPP/wollastonite/SEBS composites with nonpolar character of SEBS [16].

Yield Stress. Yield stress is mainly affected by the appearance of interfacial interactions between components in composite (in contrast to modulus usually unaffected by the interactions). Therefore, the evaluation of yield stress might be an indicator of strength of interactions in the composite [23]. The influence of wollastonites and

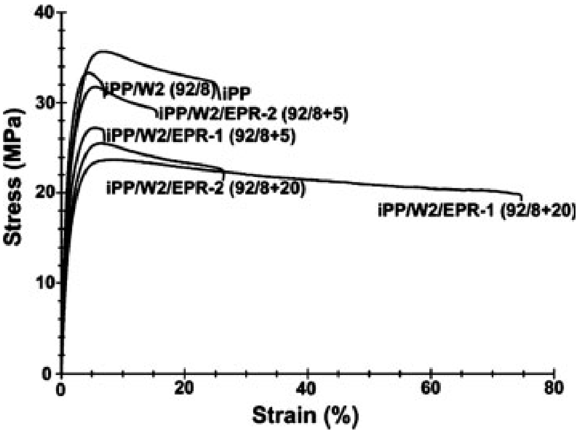

FIG. 2. Stress–strain curves of the iPP/W2/EPR composites.

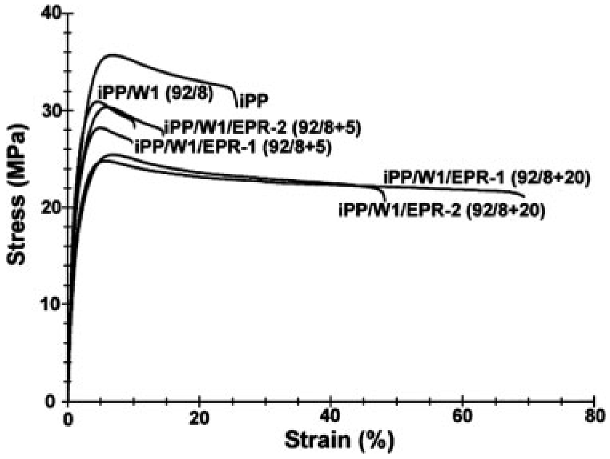

FIG. 1. Stress–strain curves of the iPP/W1/EPR composites.

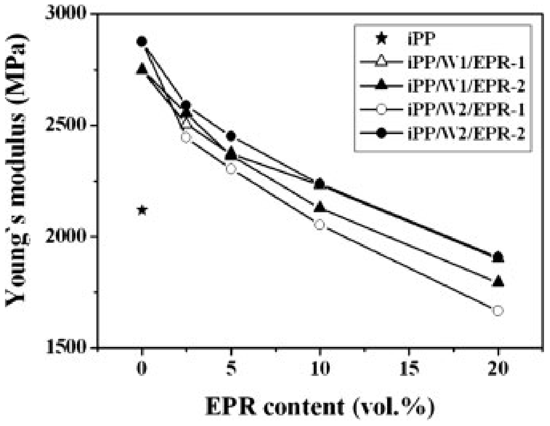

FIG. 3. Young's modulus of the iPP composites in dependence on EPR content.

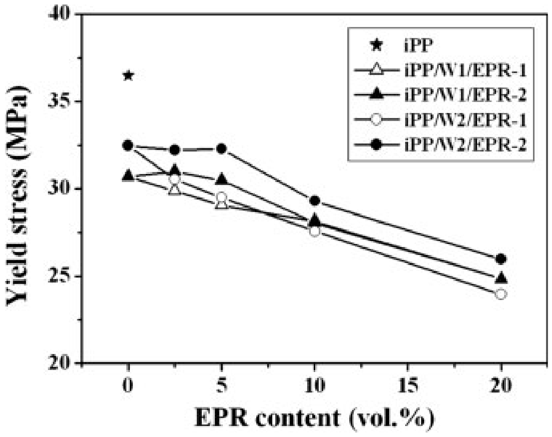

FIG. 4. Yield stress of the iPP composites in dependence on EPR content.

EPRs on yield stress of iPP composites is shown in Fig. 4.

The yield stress of neat iPP is higher in comparison to binary and ternary iPP composites according to observation of the increase of stiffness and the decrease of the yield stress with incorporation of wollastonite filler into iPP matrix [10]. The addition of both EPRs to the iPP/wollastonite composites decreases the yield stress gradually (see Fig. 4). The slope of close yield stress values for the iPP/wollastonite/EPR composites (see Fig. 4) is slightly lower that this one for the iPP/wollastonite/SRBC composites [16], i.e., composites with EPRs exhibit somewhat higher yield stress values at 20 vol% of added elastomer. It could be ascribed to somewhat stronger interactions between iPP-EPR chains than between iPP-SEBS chains. This fact is in accordance to presumable partial cocrystallization of both (iPP and EPRs) crystalline phases [24, 25].

Elongation at Yield. The extent of yield strain of semicrystalline polymers like iPP primarily depends on the strengthening of tie molecules in amorphous interlayers as well as intercrystalline and interspherulitic links. Rigid wollastonite particles incorporated into neat iPP somewhat decrease the elongation at yield probably due to the stiff effect of wollastonite particles as shown in Fig. 5. The addition of both EPR elastomers to the iPP/wollastonite composites increases the elongation at yield negligibly. Such yield strain behavior of presented composites with close elongation values almost unaffected by increased EPRs content (>2.5 vol%) is very close to this one for the iPP/wollastonite/SEBS composites [16], where SEBS particles are mainly separately dispersed in the iPP matrix.

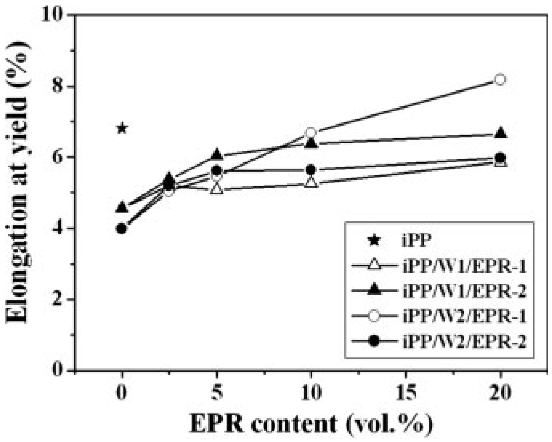

FIG. 5. Elongation at yield of the iPP composites in dependence on EPR content.

Tensile Strength at Break. Generally, the behavior of tensile strength at break (see Fig. 6) is similar to the yield stress behavior of the iPP/wollastonite/EPR composites (see Fig. 4). However, there are some differences in the values of tensile strength at break. Incorporation of 8 vol% of wollastonite into iPP matrix negligibly affects the tensile strength at break (see Fig. 6) in comparison to a significant decrease of yield stress (see Fig. 4). Furthermore, the differences between tensile strength at break values with addition of EPR elastomers are more pronounced than between yield stress values. It could be a result of differences in strengthening of the iPP/wollastonite composites toughened with different types and amounts of EPR elastomers in necking region after yield during tensile stretching. Slight differences in interactivity between iPP and two EPRs may also contribute to the pronounced differences between tensile strength at break values.

Elongation at Break. The behavior of elongation at break is usually inversely to tensile strength at break. It means that with the decrease in tensile strength at break one might expect an increase in elongation, and vice versa [3]. The introduction of wollastonite into neat iPP

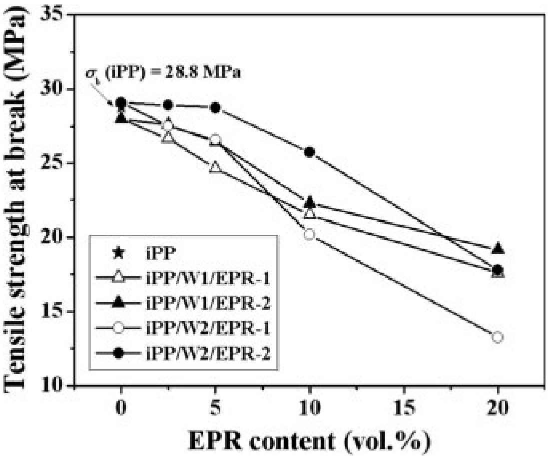

FIG. 6. Tensile strength at break of the iPP composites in dependence on EPR content.

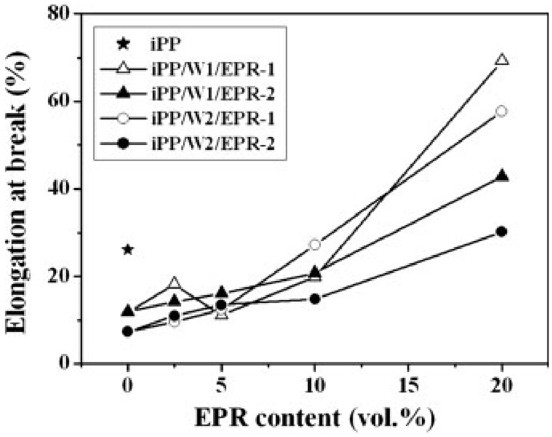

FIG. 7. Elongation at break of the iPP composites in dependence on EPR content.

decreases the elongation at break (see Fig. 7) similarly as to the yield strain (see Fig. 5). The elongation at break of ternary iPP/wollastonite/EPR composites gradually increases with elastomer content up to 10 vol%. This behavior of elongation at break is quite similar to this one for the iPP/wollastonite/SRBC composites [16]. It seems that lower content of elastomers (till 10 vol%) in ternary iPP composites slightly affect elongation at break. Higher increase and more pronounced differences between values of elongation at break of different used elastomers are observed at higher elastomer content (20 vol%) (Figs. 1, 2, and 7). The increased ductility could be explained by enhanced toughening or plastification with higher elastomer content. Observed renewed spherulites in the composites with 20 vol% of elastomers in comparison to binary iPP/wollastonite composites may additionally contribute to the enhanced ductility of these composites (Fig. 1 in Ref. 24).

The composites with EPR-1 exhibit somewhat higher elongation at break than the composites with EPR-2. The same β-iPP phase content and similar increase of crystallization temperature of the iPP/wollastonite/EPR composites [24] indicates that the β-iPP phase could not the reason for this difference in ductility [26]. However, the values of elongation at break at 20 vol% of added EPR elastomers are still lower than these ones for the composites with SEBS-*g*-MA which strongly encapsulates and disorients the wollastonite particles [16].

Impact Properties

Notched Impact Strength. Notched impact strength, as a measure of impact resistance of engineering materials defined with energy absorbed by the material under load, is important property in the field where the material has to withstand higher impact loadings. Since the numerous factors (filler aspect ratio, particle size and rigidity, interactions, elasticity of elastomers, structural changes in polymer matrix, etc.) affect the impact strength of multicomponent composites, it is difficult to resolve and to determine the affectivity of particular factor. The fillers, as rigid components, usually decrease the impact strength, whereas the elastomers usually increase it. Therefore, a proper combination of those components should be selected in order to achieve the balance of the mechanical behavior of such engineering polymeric materials. The incorporation of 8 vol% of wollastonite into iPP reduces the impact strength to some extent as shown in Fig. 8. The addition of EPRs to the iPP/wollastonite composites steadily and almost linearly increases the notched impact strength due to toughening effect of elastomers. The values of impact strength for four subsystems are very close. However, the composites with EPR-1 (at contents higher than 5 vol%) exhibit slightly higher impact strength than these ones with EPR-2 copolymer. These differences could be caused by somewhat stronger interactions between EPR-1/wollastonite than between respective pair EPR-2/wollastonite as were proven by adhesion parameters (see Adhesion phenomena). Stronger adhesion of wollastonite with EPR-1 elastomer could be a reason for location of some elastomer particles at the wollastonite particle surface diminishing thus the deteriorative effect of wollastonite on the impact properties. On the other side, EPR-2 elastomer particles are mainly separately distributed in the iPP matrix since better miscibility of iPP with EPR-2 than with EPR-1. As a result EPR-2 exhibits lower effect on the impact properties in ternary composites.

Adhesion Phenomena in Ternary Composites

The morphology of composites very much depends on adhesion phenomena in engineering composites. In this manner, the study of adhesion phenomena is suitable to composite engineering [27–29]. The morphology and con-

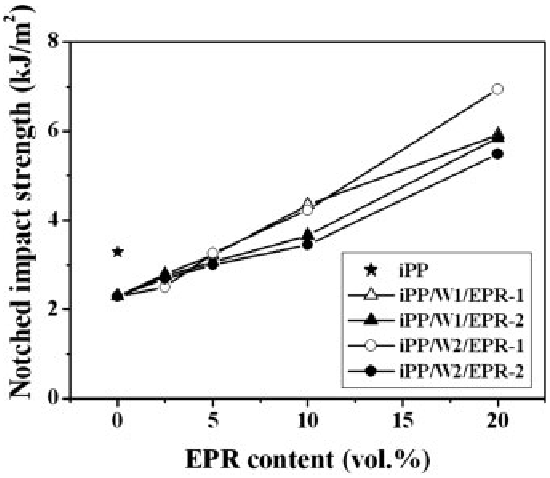

FIG. 8. Notched impact strength of the iPP composites in dependence on EPR content.

TABLE 2. The surface free energies (γ_s) and their dispersive (γ_s^d) and polar (γ_s^p) components of the iPP, wollastonite fillers, and elastomers.

Sample	Surface free energy (mJ/m^2)		
	γ_s^d	γ_s^p	γ_s
iPP	31.5	1.3	32.8
W1	36.9	29.0	65.9
W2	37.4	33.5	70.9
EPR-1	26.7	4.7	31.4
EPR-2	25.3	1.4	26.7

sequently the properties of composites could be predicted in some extent, using the calculation of surface free energies of pure materials and adhesion parameters. For this purpose, the surface free energies of pure materials (given in Table 2) are calculated from the results of contact angle measurements by using harmonic mean equation.

The results in Table 2 exhibit quite low values of polar component of the surface free energy (γ_s^p) of the iPP and elastomers as a consequence of their nonpolar nature. However, there is some difference between EPR copolymers. EPR-1 has higher polar component value in comparison to the EPR-2, which is surprisingly fact because of the same chemical constitution of used copolymers. The wollastonites used might be expected to exhibit higher surface free energies with regard to the iPP and elastomers. Additionally, adhesion parameters such as work of adhesion, (W_{mf}), interfacial free energy (γ_{mf}) and spreading coefficient (coefficient of wetting) (S_{mf}) were used to predict the adhesion strength of possible pairs polymer/filler, polymer/elastomer and elastomer/filler in ternary composites and, consequently, to affect the morphology of corresponding ternary composites.

The adhesion parameters were calculated with the following equations [30]:

$$W_{mf} = \gamma_f + \gamma_m - \gamma_{mf} \tag{2}$$

$$\gamma_{mf} = \gamma_f + \gamma_m - 2(\gamma_f^d \times \gamma_m^d)^{1/2} - 2(\gamma_f^p \times \gamma_m^p)^{1/2} \tag{3}$$

$$S_{mf} = \gamma_f - \gamma_m - \gamma_{mf} \tag{4}$$

where subscripts m and f mean matrix and filler, respectively.

The results of the studies on the effective adhesion for a given system indicate some conditions as optimal: thermodynamic work of adhesion as a maximal, spreading coefficient as a positive value and interfacial free energy as a minimal (tends to null) [29]. The calculated adhesion parameters for possible pairs in the iPP/wollastonite/EPR composites are given in Table 3. Higher work of adhesion and spreading coefficient, as well as lower interfacial free energy of EPR-1/W1 and EPR-1/W2 in comparison to EPR-2/W1 and EPR-2/W2 pairs were obtained. According to the conditions for effective adhesion, stronger adhesion could be expected in investigated ternary iPP composites with EPR-1 and both wollastonites. Furthermore, somewhat higher work of adhesion and interfacial free energy of the iPP/EPR-1 than iPP/EPR-2 were obtained. Although some authors [31–33] were found good correlation between the degree of miscibility and work of adhesion in the case of polymer blends, interfacial free energy is more relevant for defining the miscibility of the polymers. Interfacial free energy is the inversely proportioned to the strength of intermolecular interactions in polymer blends [34–36]. In this case it could be expected higher extent of miscibility of iPP and EPR-2 rather than between iPP and EPR-1. This assumption, reflected by impact strength behavior, is in a good accordance also with the results of phase morphology investigations which have shown that more EPR-1 than EPR-2 particles were accommodated at the surfaces of wollastonite particles [24].

Determined difference in adhesion properties led to the difference in phase morphology between composites with EPR-1 and EPR-2 copolymers [24] that reflects in their mechanical properties.

TABLE 3. Adhesion parameters for possible pairs in iPP/wollastonite/elastomer composites.

System	Possible pairs	Adhesion parameters (mJ/m^2)		
		Work of adhesion, W_{mf}	Interfacial free energy, γ_{mf}	Spreading coefficient, S_{mf}
iPP/W1/EPR-1	iPP/W1	80.3	18.4	14.7
	iPP/EPR-1	63.0	1.3	n.a.
	EPR-1/W1	86.1	11.2	23.3
iPP/W1/EPR-2	iPP/W1	80.3	18.4	14.7
	iPP/EPR-2	59.1	0.4	n.a.
	EPR-2/W1	73.8	18.8	20.5
iPP/W2/EPR1	iPP/W2	81.7	22.0	16.0
	iPP/EPR1	63.0	1.3	n.a.
	EPR1/W2	88.3	14.0	25.4
iPP/W2/EPR2	iPP/W2	81.7	22.0	16.0
	iPP/EPR2	59.1	0.4	n.a.
	EPR2/W2	75.1	22.4	21.8

n.a, not available.

CONCLUSION

The mechanical properties of ternary iPP/wollastonite/EPR composites, where metallocene EPR copolymers were used as impact modifiers and compatibilizers, were influenced by observed morphology caused by partial miscibility of the iPP and EPR chains as well as by different level of adhesion between components. The composites with two different surface treated wollastonites and two EPRs used exhibit similar behavior of most mechanical properties irespective of the difference in properties of the components, especially used EPR elastomers (viscosity, adhesion). Observed difference in elongation at break between the composites with 20 vol% of EPR-1 and EPR-2 could be resulted from the difference in spherulitic morphology renewed by introducing higher content of EPR elastomers. Stronger interactions between EPR-1 and other two components (iPP and wollastonite) than between iPP and EPR-2 and/or wollastonite components, respectively, concluded on the basis of adhesion parameters, may cause the difference in phase morphology and, consequently, in impact strength behavior. The mechanical properties confirm the assumption coming out of structural investigations that metallocene EPR elastomers are rather efficient impact modifiers than encapsulation compatibilizers for the iPP/wollastonite composites [24].

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