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Tensile and impact properties of three-component PP/wood/elastomer composites

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Abstract. Polypropylene (PP) was reinforced with wood flour and impact modified with elastomers to increase stiffness and impact resistance simultaneously. Elastomer content changed in four (0, 5, 10 and 20 wt%), while that of wood content in seven steps, the latter from 0 to 60 wt% in 10 wt% steps. Structure and adhesion were controlled by the addition of functionalized (maleated) polymers. Composites were homogenized in a twin-screw extruder and then injection molded to tensile bars. Fracture resistance was characterized by standard and instrumented impact tests. The results showed that the components are dispersed independently of each other even when a functionalized elastomer is used for impact modification, at least under the conditions of this study. Impact resistance does not change much as a function of wood content in PP/wood composites, but decreases drastically from the very high level of the PP/elastomer blend to almost the same value obtained without impact modifier in the three-component materials. Increasing stiffness and fiber related local deformation processes led to small fracture toughness at large wood content. Micromechanical deformation processes depend mainly on the strength of PP/wood interaction; debonding and pull-out take place at poor adhesion, while fiber fracture dominates when adhesion is strong. Composites with sufficiently large impact resistance cannot be prepared in the usual range of wood contents (50–60 wt%).

Keywords: *damage mechanism, PP/wood composites, impact modification, interfacial adhesion, composite structure*

1. Introduction

Wood flour and natural fibers are used in increasing quantities mainly for the reinforcement of commodity polymers [1–3]. Such reinforcements have many advantages over particulate fillers or glass fibers; they increase stiffness considerably, they are obtained from renewable resources, are available in abundant quantities, cheap, and light at the same time [2, 4, 5]. Major application areas of these materials are the building and the automotive industries. In structural applications often large stiffness and impact resistance are required simultaneously, which are achieved traditionally by the combination of several functional additives. Composites used as bumper materials, for example, usually contain an elastomer to improve impact resistance and a filler or fiber to increase stiffness [6–8]. Research has started as early as the 80's on these materials [6, 9–12] and they have been commercially available for several decades.

Structure can be quite complicated in such multicomponent materials. Two boundary structures may form in them: the two components, i.e. the elastomer and the filler, can be distributed separately

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from each other in the polymer matrix [13–15], or the elastomer can encapsulate the reinforcement to create embedded structure [6–8, 16]. The actual structure is determined by the adhesion and shear forces prevailing in the melt during homogenization, the first favoring embedding because of thermodynamic reasons, while the second separate dispersion through the shearing apart of the elastomer layer from the wood fiber [17]. Usually intermediate structures form in composites produced under practical conditions, a part of the filler is embedded into the elastomer phase, but individual elastomer droplets and filler particles can be also located in the matrix. Structure can be tailored by the control of interfacial adhesion through the use of appropriate coupling agents [18–21]. Functionalized polymers are used to control structure in polypropylene. The introduction of maleated PP (MAPP) leads almost exclusively to the separate dispersion of the components. The reaction of the maleic anhydride group with the wood fibers creates strong covalent bonds, on the one hand, and the interdiffusion of MAPP forms entanglements with the matrix polymer, on the other, resulting in strong adhesion and good stress transfer as well [22–24]. Adhesion force changes from about 100 mJ/m^2 to nearly 1000 mJ/m^2 in this way [25]. The addition of maleated ethylene-propylene-diene elastomer (MAEPDM), on the other hand, results in a large extent of embedding. Properties change considerably with structure even at the same composition. Stiffness was shown to depend mainly on the extent of embedding, while impact resistance was influenced also by other factors including micromechanical deformation processes occurring around the inclusions (elastomer, filler) [26].

It seems to be obvious to use wood and/or natural fibers to replace mineral fillers or glass fibers also in such composites. However, wood flour differs considerably from traditional reinforcements. Wood particles are large, usually several 100 μm in size, which facilitates debonding, the separation of the matrix/filler interface already at small stresses [27–29]. A functionalized polymer coupling agent is needed practically always in order to achieve reasonable properties, at least in polyolefin composites. Besides debonding, large wood particles may initiate other micromechanical deformation processes during the deformation of the composites like fiber pull-out, or fiber fracture at strong interfacial adhesion [29, 30]. These differences compared to particulate fillers and glass fibers, and the tendency to replace traditional reinforcements with natural ones require more detailed study of the behavior of multicomponent materials containing wood fibers. Very few papers have been published in this area yet. A model study was carried out on the recycling of PP/PE blends by Clemons [31], and functionalized elastomers were used to modify structure and properties in PP/wood composites by Oksman and Clemons [32, 33]. Since impact resistance is one of the crucial properties in structural materials, the goal of our study was to investigate the effect of structure and interfacial adhesion on fracture toughness in wood reinforced multicomponent PP composites impact modified with elastomers. An attempt was made to control structure and adhesion by the use of functionalized polymers. Wood content changed in a wide, industrially relevant range. Besides the effect of wood content on fracture resistance, we tried also to identify the mechanism of failure in these composites.

2. Experimental

The polymer used in the study was the Tipplen H 781 F grade PP homopolymer (MFR = 0.7 g/10 min at 230°C and 2.16 kg load) produced by TVK, Tiszaújváros, Hungary. The Vistalon 706 ethylene-propylene-diene (EPR) elastomer (ethylene content: 65 wt%, Mooney viscosity ML1+4 at 125°C: 42) of Exxon Mobil, Houston, USA was used to increase impact resistance. The functionalized polymers applied for the control of structure and interfacial adhesion were the Orevac CA 100 grade maleated PP (MFI = 150–200 g/10 min at 230°C and 2.16 kg, MA content: 1.0 wt%) from Arkema, Puteaux, France and the Exxcellor VA 1803 maleated EPDM (ethylene content: 43 wt%, MFI = 3 g/10 min at 230°C and 2.16 kg, MA content: 0.5–1.0 wt%) from Exxon Mobil, Houston, USA. We hoped that the application of the first leads to separate dispersion, while the second to the complete encapsulation of wood particles. The Filtracel EFC 1000 wood flour was supplied by Rettenmaier and Söhne GmbH, Rosenberg, Germany. The wood was treated to remove waxes by the producer, it contained 70.4 wt% holocellulose, 28.7 wt% lignin and 0.9 wt% waxes. The filler had an average particle size of 210 μm as determined by laser light scattering. Scanning elec-

tron microscopic (SEM) analysis of particle geometry showed average particle length of 363 μm, diameter of 64 μm and aspect ratio of 6.8. MAPP was always added in 10 wt% calculated for the amount of wood [34], while 5, 10 and 20 wt% of the matrix polymer was replaced by impact modifier (EPR or MAEPDM). Wood content changed from 0 to 60 wt% in 10 wt% steps related to the total weight of the composites.

The composites were homogenized using a ThermoPrism TSE 24 (Thermo Fisher Sci. Inc., Waltham, USA) twin-screw extruder with a screw diameter of 24 mm and an *L*/*D* ratio of 28. Screw configuration included two kneading zones with different lengths and conveying elements. The polymer components were introduced into the hopper, while wood was added to the melt through a side feeder. Zone temperatures changed from 170 to 220°C in 10°C steps in the six zones of the extruder. The granulated material was dried for 4 hours at 105°C in an oven and then injection molded to standard ISO 527 1A tensile specimens using a Demag IntElect 50 machine (Demag Ergotech GmbH, Schwaig, Germany) at 170–180–190–200–210°C zone and 50°C mold temperatures, 50 mm/s injection rate, max 1300 bar holding pressure and 25 sec holding time. The samples were conditioned at 23°C and 50% RH for a week before testing.

Tensile testing was carried out with an Instron 5566 type machine (Instron Corp., Canton, USA). Stiffness was determined at 0.5 mm/min, while other tensile characteristics like yield stress, yield strain, tensile strength and elongation-at-break at 5 mm/min cross-head speed and 115 mm gauge length. Impact resistance was determined on notched Charpy specimens according to the ISO 179 standard at 2 mm notch depth. Instrumented impact testing was carried out using a Ceast Resil 5.5 instrument (CEAST S.p.A., Pianezza, Italy) with a 4 J hammer. The structure of the composites was studied by scanning electron microscopy using a Jeol JSM 6380 LA apparatus (JEOL Ltd., Tokyo, Japan). The distribution of the components in the matrix was determined on fracture surfaces created at liquid nitrogen temperature. Samples containing elastomer were etched in n-hexane for 1 min. SEM micrographs were recorded also on surfaces created in the tensile or impact test in order to determine the mechanism of failure. Etching was used when appropriate.

3. Results and discussion

The combination of all the compositional variables resulted in a very large number of composites. As a consequence we refrain from the presentation of all results and focus our attention on materials containing 20 wt% elastomer. However, all the results are presented in figures showing general correlations. In the first two sections we present tensile properties and structure, while impact resistance is analyzed in detail in the next part of the paper. General correlations and practical consequences are discussed in the last section.

3.1. Tensile properties

Model calculations proved that thermodynamics favors the formation of embedded structure. On the other hand, weak interaction and large shear destroys the embedded structure formed, separate the filler and elastomer from each other [17]. Besides being an important characteristic of structural materials, the stiffness of PP composites containing an elastomer and a reinforcement at the same time offers valuable information also about structure. The elastomer decreases stiffness, but otherwise the effect of the components is additive in the case of separate dispersion. On the other hand, embedding results in additional decrease of stiffness, the extent of which can be used for the estimation of the amount of embedded particles [26, 35]. The dependence of the Young's modulus of the composites is presented in Figure 1. The wood flour used in our study reinforces PP considerably and interfacial adhesion does not influence stiffness much. These observations are in complete agreement with our earlier results [36–38]. A slight deviation is observed from the expected tendency at large, 50 and 60 wt% filler content, which indicates a small extent of aggregation. The separate dispersion of the components is expected when both MAPP and EPR are added (▽), and the correlation is practically parallel to that of the PP/wood/MAPP composites (○) proving that the expectation is fulfilled. At large wood content the stiffness of the PP/wood/EPR (△) composites is somewhat smaller than in the presence of MAPP indicating a small extent of embedding or larger extent of aggregation. However, this latter seems to be less probable. MAEPDM was expected to encapsulate the particles completely [31, 32]. The composition dependence of the stiffness of the compos-

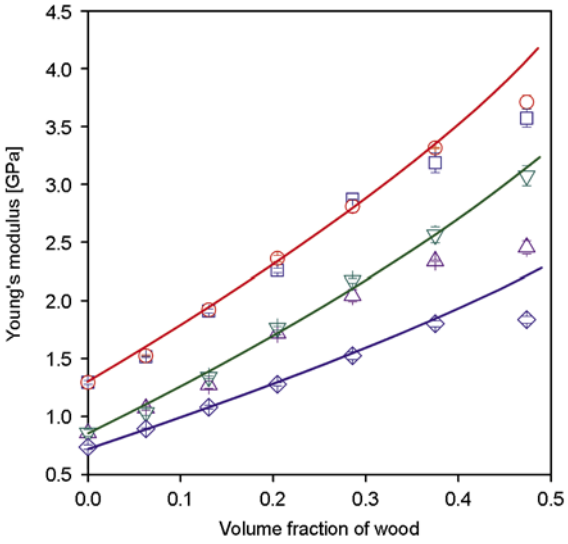


Figure 1. Stiffness of multicomponent PP composites plotted against their wood content. Elastomer content is 20 wt%, that of MAPP is 10 wt%. Symbols: (□) PP/wood, (○) PP/wood/MAPP, (Δ) PP/wood/EPR, (▽) PP/wood/EPR/MAPP, (◇) PP/wood/MAEPDM.

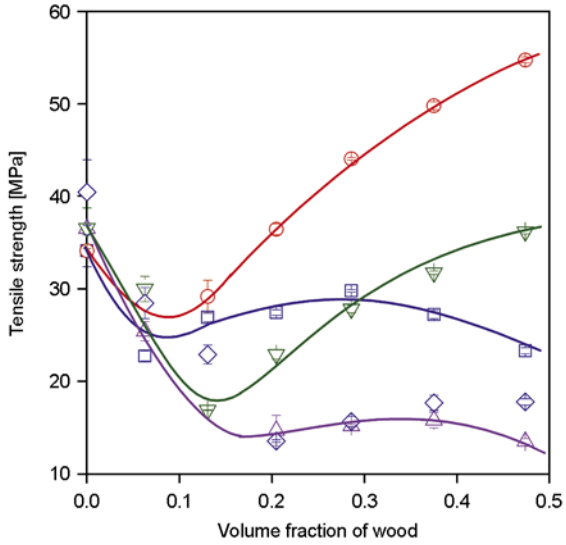


Figure 2. Effect of composition and coupling on the tensile strength of PP/wood composites. Composition and symbols are the same as in Figure 1.

ites containing this component does not confirm this expectation (◇). Wood reinforces PP also in this case and the increase in modulus is only slightly smaller in the presence of MAEPDM than with EPR that indicates a somewhat larger, but still small extent of embedding. We may conclude from the analysis of the composition dependence of stiffness that only small extent of embedding occurs in our composites and the separate distribution of the components dominates.

As shown above, stiffness does not depend very much on interfacial adhesion; its effect cannot be deduced from the composition dependence of Young's modulus. Properties measured at larger deformations, like tensile strength, show changes in interactions very sensitively. Tensile strength is plotted against wood content for the same five series of composites in Figure 2. The effect of adhesion and elastomer modification can be clearly seen in the figure. We refrained from drawing lines through all series for better clarity; lines are drawn anyway only to guide the eye and they are not fitted correlations. If we compare PP/wood composites with (○) and without (□) MAPP, the effect of adhesion becomes obvious. Strength increases drastically with increasing wood content in the first case, while it remains constant or slightly decreases in the second. The incorporation of the elastomer decreases strength, but the relative effect of adhesion, i.e. the presence or absence of MAPP, remains the same. Strength is very small in composites containing EPR without MAPP (Δ), while a considerable increase in strength is observed at strong adhesion (▽). These results also support our assumption about the separate distribution of the components. The effect of MAEPDM is very similar to that of EPR; composite strength is very small in its presence (◇). We must also comment on the strange composition dependence of strength with the minimum at small and the increase or leveling off at larger wood content. The deformability of the matrix (with or without elas-

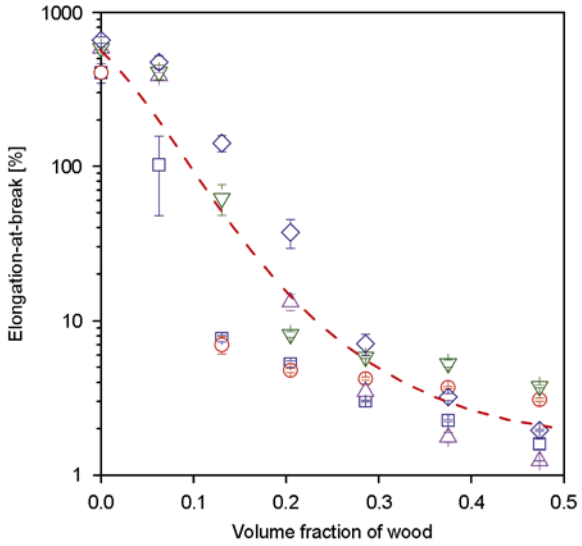


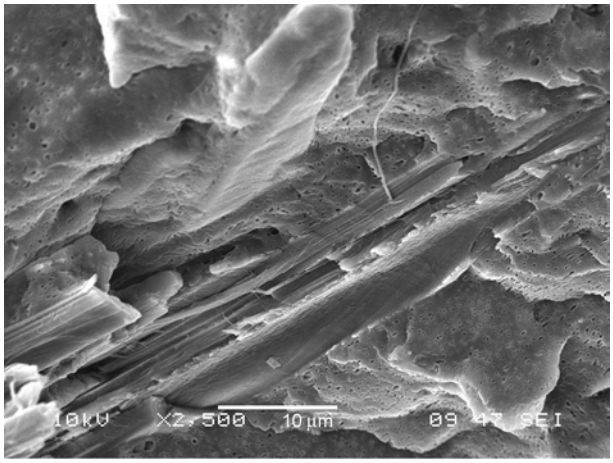
Figure 3. Dependence of the deformability of PP/wood composites on composition and interfacial adhesion. Composition and symbols are the same as in Figure 1.

tomer) and the composites containing various amounts of wood is extremely different; it covers a wide range between 1000 and 2% (Figure 3). Specimen cross-section changes at large elongations and strain hardening increases strength thus complicating the comparison of engineering strength values. Nevertheless, Figure 3 indicates that the presence of the elastomer increases the deformability of the samples, thus we may expect larger impact resistance for these composites.

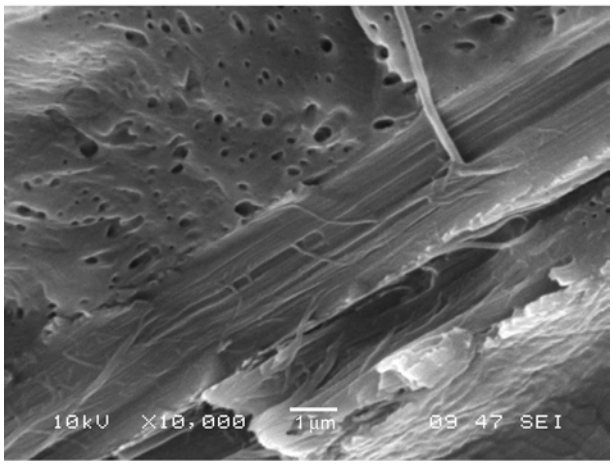
3.2. Structure

Structure is one of the major factors determining the properties and performance of heterogeneous materials. The structure of the multicomponent multiphase materials in question is rather complicated. The dispersion of the components, aggregation, the orientation of the fibers, and crystalline morphology are the main factors to be considered in the interpretation of composite behavior. One of the most important of these issues is the distribution and possible embedding of the fibers into the elastomer. The composition dependence of stiffness, and that of the other properties studied, indicates that the components are separately dispersed in most composites and only a small extent of encapsulation may occur mostly at large fiber and elastomer content.

In order to check this conclusion drawn from the composition dependence of tensile properties, the structure of the composites was studied also by scanning electron microscopy. The distribution of the components can be determined quite easily on fracture surfaces etched with n-hexane [13]. The elastomer is removed by the solvent during etching leaving holes behind, thus the discrimination of the phases becomes quite easy. Embedding can be detected by the apparently smaller number and total area of holes, and by voids appearing around wood particles. We refrain from the presentation of a large number of micrographs and show only two structures recorded on composites, which are supposed to have the two boundary structures, i.e. separate distribution of the components or complete encapsulation. The first structure should be obtained when both an elastomer and MAPP are added to the PP/wood composite. This structure is shown in Figure 4a. The micrograph verifies our assumption; the elastomer is distributed as submicron sized particles



a)



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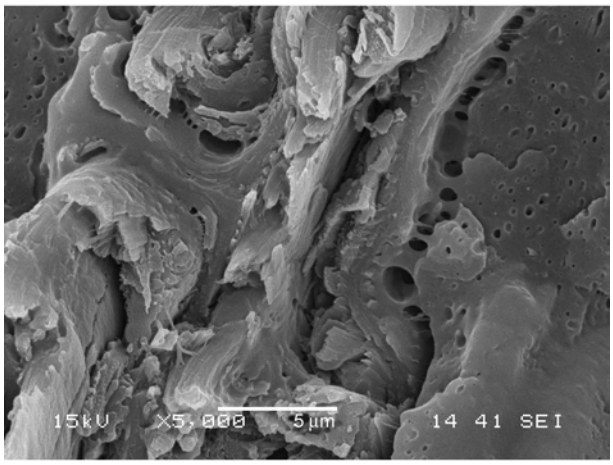

c)

Figure 4. Distribution of the components in PP/wood/elastomer composites in the presence of different functionalized polymers. a) PP/wood (29 vol%)/EPR/MAPP, b) the same as a) at larger magnification, c) PP/wood (29 vol%)/MAEPDM.

independently of the large wood particles. Unfortunately, the dissimilar dimensions of the dispersed components make the analysis of the structure quite complicated. The same part of the composite is

shown in Figure 4b in larger magnification. The micrograph clearly shows that wood is firmly embedded in the PP matrix and elastomer particles are not located on its surface. The other boundary structure, complete embedding, could not be verified in the same way. Figure 4c presents a SEM micrograph taken from the fracture surface of a PP composite containing MAEPDM besides wood. The structure is very similar to that shown in Figure 4a. A large number of small elastomer droplets are visible on the surface. This does not exclude the possibility of fiber encapsulation, but the extent of the latter must be small. A long thin crack runs around the large wood particle, which might be interpreted as dissolved MAEPDM elastomer. However, it is apparently located in the matrix and not on the surface of the wood, and the number of separately dispersed elastomer particles strongly denies the formation of embedded structure. Accordingly, the SEM study confirmed our previous conclusion drawn from the composition dependence of tensile properties that elastomer and wood are distributed mostly separately in the PP matrix.

3.3. Fracture toughness

Impact resistance might be the crucial property of PP/wood composites used in certain application areas as structural materials. Reports in the literature indicate that impact resistance often decreases as an effect of wood reinforcement [38–44] similarly to many particulate filled composites [45–51]. On the other hand, this decrease was compensated by the incorporation of an elastomer into these latter materials. The composition dependence of notched Charpy impact strength is presented for the five series of composites in Figure 5. Without elastomer fracture toughness is relatively small and it appears to go through a slight maximum with increasing wood content. The effect is stronger for PP/wood composites not containing MAPP than in those prepared with it, i.e. at good adhesion. The elastomer increases impact resistance considerably, as expected. However, fracture toughness decreases drastically with increasing wood content almost to the same level of PP/wood composites at the end of the composition range. Contrary to tensile strength, adhesion seems to have only a small effect on impact toughness; obviously other factor or factors determine the resistance of the material against fracture.

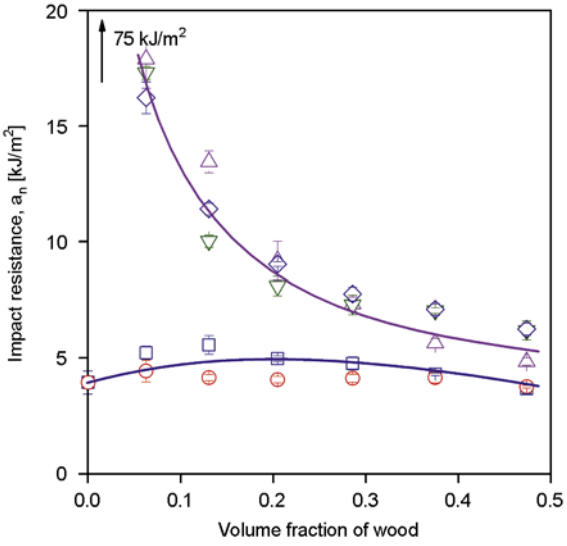

Figure 5. Effect of composition and interfacial adhesion on the impact resistance of multicomponent PP/wood composites. Composition and symbols are the same as in Figure 1.

We hoped that instrumented impact testing supplies additional information about the fracture process itself and about the factors determining fracture resistance. Selected force vs. time traces are presented in Figure 6 to demonstrate the effect of the various additives and factors on the fracture process. Neat PP fails by brittle fracture (Figure 6a). The maximum of the force vs. time traces and the critical stress intensity factor K_{Ic} is related to fracture initiation, while the area under the traces depends also on crack propagation. Fracture is initiated at a relatively small force and catastrophic failure occurs in a few milliseconds with small energy consumption. The presence of a small amount of wood

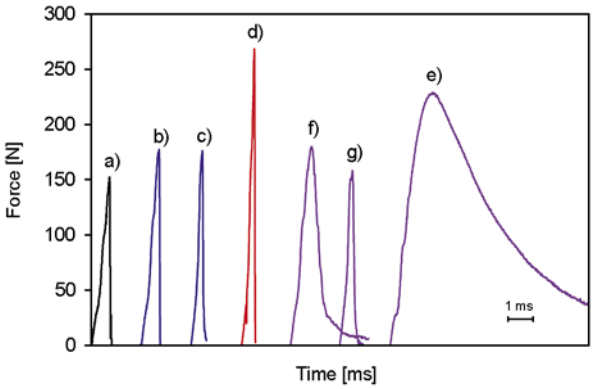

Figure 6. Force vs. time traces recorded by instrumented impact testing on selected multicomponent PP/wood composites. Elastomer content is 20 wt%. a) PP, b) PP/6 vol% wood, c) PP/47 vol% wood, d) PP/47 vol% wood/MAPP, e) PP/EPR, f) PP/13 vol% wood/EPR, g) PP/47 vol% wood/EPR.

increases initiation force and does not change the time to failure much, which leads to increasing fracture energy (Figure 6b). We assume that debonding is the dominating micromechanical deformation process in these composites, which requires surplus energy consumed by the debonding process itself and the subsequent plastic deformation. At larger amount of wood (47 vol%, Figure 6c) stiffness and initiation force increase further, but increased stiffness leads to smaller plastic deformation and reduced energy of fracture (see also the slight maximum in Figure 5). Improved adhesion, i.e. the presence of MAPP, results in a significant increase in initiation force, but increased stiffness reduces the resistance against crack propagation thus overall fracture resistance remains practically constant (Figure 6d). The polymer containing the elastomer behaves completely differently. The elastomer increases the resistance against initiation (see Fmax~240 N), but catastrophic failure does not occur, the propagation of the crack needs constant energy supply (Figure 6e). The addition of wood to the PP/elastomer blend increases stiffness and facilitates crack propagation (Figure 6f) and at large wood content the traces, thus the fracture process, becomes similar, if not the same, as without the elastomer (Figure 6g). Both initiation force and the time to fracture decrease significantly with increasing wood content.

The comparison of the force vs. time traces of Figure 6 indicates that the two components change both crack initiation and propagation. Wood content and adhesion have larger effect on the first, while elastomer on the second. The critical stress intensity factor, K_{Ic}, was calculated by Equation (1) [52]:

$$K_{Ic} = \sigma_F Y a^{1/2} \tag{1}$$

where σ_F is the maximum force recorded during fracture (see Figure 6), a the depth of the notch and Y is a factor depending on the dimensions of the specimen and on loading conditions. The effect of the components on the critical stress intensity factor, K_{Ic}, is presented in Figure 7. We can clearly see the influence of the various processes and factors on the variation of K_{Ic}. The combined effect of reinforcement and increasing stiffness is reflected in the maximum of the correlation obtained for the PP/wood composites (□). The continuously increasing K_{Ic} of the PP/wood/MAPP composite indicates that debonding must play an important role in failure (○).

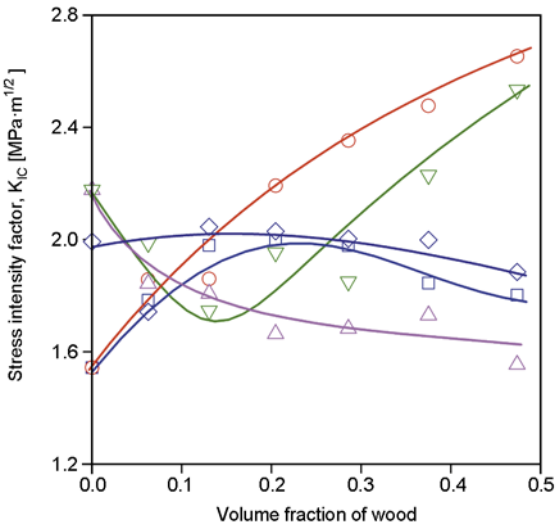

Figure 7. Dependence of the critical stress intensity factor (K_{Ic}) on composition and interfacial adhesion. Composition and symbols are the same as in Figure 1.

Increasing wood content leads to decreasing initiation resistance in the presence of the elastomer (△) and the combined effect of elastomer content, adhesion and wood content is shown by the correlation obtained for PP/wood/EPR/MAPP composites (▽). Although the critical stress intensity factor changes in a wide range, crack propagation seems to dominate fracture resistance and the effect of wood is stronger than that of the elastomer when both components are present. Obviously crack propagation becomes fast and plastic deformation small when the matrix contains large amounts of wood.

Further information can be obtained about the effect of the main factors, if we analyze the composition dependence of fracture resistance with the help of a simple model developed earlier [53], shown in Equation (2):

$$a_n = \frac{a_{n0}}{E/E_0} \frac{1-\varphi}{1+2.5\varphi} \exp(B\varphi) \tag{2}$$

where a_n and a_{n0} are the impact resistance of the composites and the matrix, respectively, E/E_0 is the relative stiffness of the composites, φ is the volume fraction of the dispersed component and B expresses the effect of this latter on impact resistance. According to the model impact resistance is influenced by four factors: matrix property (a_{n0}), the decrease of the deformability of the polymer with increasing amount of reinforcement (E/E_0), changing load-bearing cross-section [$(1-\varphi)/(1+2.5\varphi)$], interfa-

cial interactions and all additional factors [exp($B\varphi$)]. The model proved to be valid for a large number of particulate filled, elastomer modified and multicomponent materials [53]. In these latter the analysis is somewhat difficult since different approaches can be adopted. PP can be regarded as the matrix, the role of the elastomer ignored and only fiber content used as independent variable. This route is obviously wrong because of the influence of the elastomer on both fracture initiation, but especially on crack propagation. The PP/elastomer blend can be also regarded as matrix, while the third possibility is to investigate the combined effect of the additives in the PP matrix. Since the analysis of structure indicated the separate dispersion of the components, we followed the last route.

If we transform Equation (2) to calculate reduced impact strength by dividing composite impact strength with the factors accounting for matrix property, deformability and load-bearing cross-section we arrive to Equation (3):

$$a_{\mathrm{nred}} = a_{\mathrm{n}} \frac{E}{E_0} \frac{1 + 2.5\varphi}{1 - \varphi} = a_{\mathrm{n0}} \exp(B\varphi) \qquad (3)$$

and, if we plot the natural logarithm of this quantity against filler content, we should obtain a straight line the slope of which is parameter B. This latter expresses the effect of the dispersed component on fracture resistance compared to zero effect, i.e. a composite containing the amount of holes corresponding to φ. This means that a filler, reinforcement or other component can have a positive effect on impact resistance even if the actual numbers decrease compared to the matrix value. B is influenced by interfacial interactions, but also by structural effects like particle or matrix orientation, aggregation, etc.

The results obtained for five series are presented in in Figure 8. We can see two sets of lines, one with large slopes corresponding to two-component PP/elastomer blends, and the other to composites containing both elastomer and wood. The combined amount of wood and elastomer is used as compositional variable for these latter composites. We can see that straight lines are obtained in all cases indeed. The slope for the PP/elastomer blends is very large and similar to each other in agreement

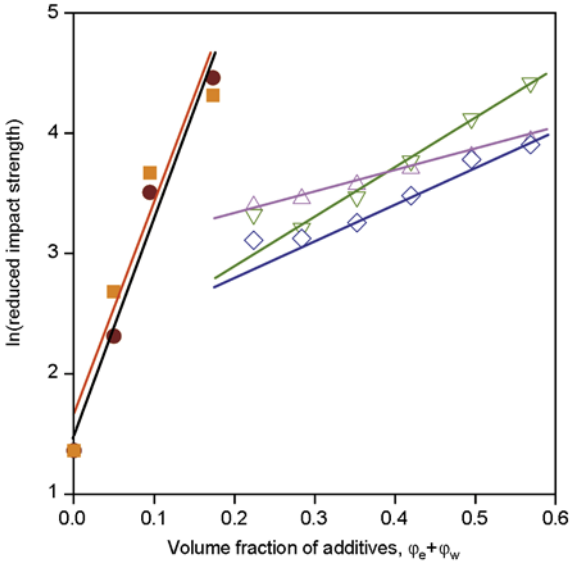

Figure 8. Model calculations carried out for the analysis of the effect of components on the impact resistance of three-component PP/wood/elastomer composites (see Equations (2) and (3) and the calculated parameters in Table 1). Symbols: (●) PP/EPR, (■) PP/MAEPDM blend; the rest of the symbols are the same as in Figure 1.

with the known fact that elastomers increase the impact resistance of PP considerably. The EPR used has a slightly larger effect than the MAEPDM selected. More interesting are the three-component PP/wood/elastomer composites. The smaller slopes express the weaker effect of wood on impact resistance, while the intercepts which differ from the corresponding matrix property express the combined effect of the elastomer and the orientation of the fibers. The slope of the line for the PP/wood/elastomer composites prepared without MAPP (△) shows that wood has very little positive effect on impact resistance and that it comes mainly from the energy needed for debonding and from a slight hindrance of crack propagation. Adhesion influences the fracture process strongly and the effect of wood on it (▽), B increases considerably compared to the previous case. This change is caused, in all probability, by the increase of initiation force (see Figure 7). On the other hand, the smaller intersection indicates that adhesion counteracts the positive effect of the elastomer. Parameter B of the composites containing MAEPDM is only slightly larger than that obtained for EPR, which confirms the similarity of structures and the separate distribution of the com-

Table 1. Effect of elastomer impact modifier and wood reinforcement on the impact resistance of three-component PP/wood composites; results of model calculations (see Equations (2) and (3))

Component			Intersection	a_{n0c}[a] [kJ/m^2]	B	R^{2b}
Wood	Elastomer	MAPP				
–	EPR	–	1.46	4.32	18.22	0.9746
–	MAEPDM	–	1.67	5.31	16.85	0.9269
+	EPR[c]	–	3.02	20.44	1.61	0.9951
+	MAEPDM[c]	–	2.27	9.67	2.93	0.9822
+	EPR[c]	+	1.97	7.20	4.30	0.9989

[a]calculated impact resistance of the matrix
[b]determination coefficient, goodness of the linear fit
[c]at 20 wt% elastomer content

ponents. The parameters determined by the model calculations are compiled in Table 1 and support our considerations presented above. We can conclude from all these results that although the presence of wood improves resistance against crack initiation, it facilitates crack propagation very much and thus becomes the dominating factor determining fracture toughness at large wood content.

3.4. Correlations, consequences

The application of multicomponent materials in practice indicates that the approach of the simultaneous use of an elastomer and a reinforcing filler or fiber results in composites which, at least in certain cases, have large stiffness and considerable impact resistance at the same time. The materials prepared in this study failed to meet this requirement espe-

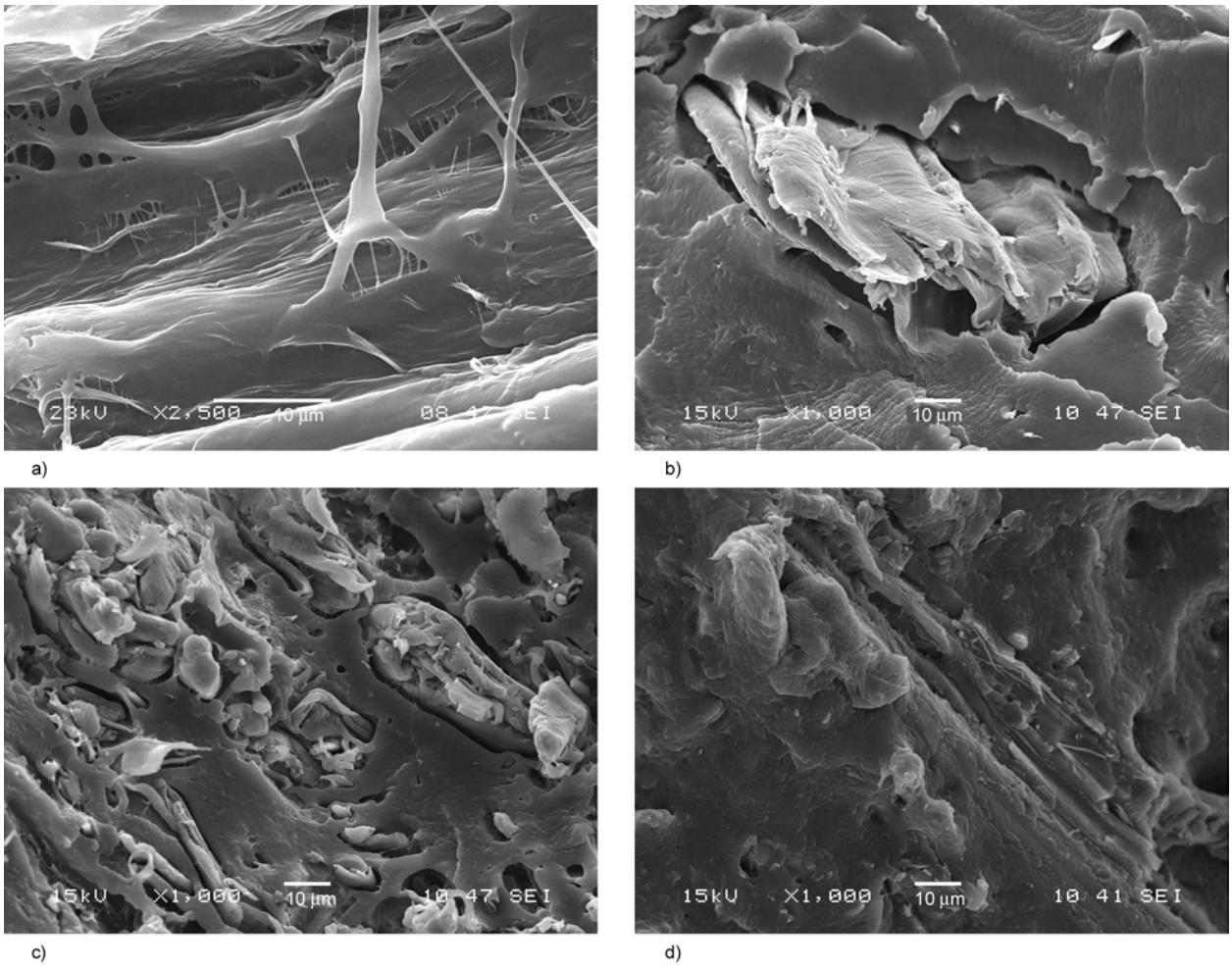

Figure 9. SEM micrographs recorded on the fracture surface of multicomponent PP/wood composites; study of deformation and failure mechanism. The surfaces were created in impact testing. a) PP/20 wt% EPR, b) PP/6 vol% wood/20 wt% elastomer, c) PP/29 vol% wood/20 wt% elastomer, d) PP/29 vol% wood/20 wt% elastomer/MAPP.

cially in the usual range of wood contents. Previous sections showed the main factors influencing fracture toughness. In a further attempt to reveal the reason for the lack of success, we analyzed the fracture surface of various samples by SEM. Only a few examples are shown here to support conclusions drawn in previous sections. Figure 9a shows the fracture surface of a specimen prepared at large elastomer content. We can see that the presence of the elastomer results in considerable plastic deformation as expected. This effect depends on the particle size of the droplets, their dispersion, the properties of the elastomer and interaction. Differences in these factors explain the dissimilar effect of the EPR and the MAEPDM used here. Wood on the other hand, decreases fracture resistance generally, in spite of the fact that debonding consumes energy. The micrograph in Figure 9b proves that very limited plastic deformation occurs in the presence of even a small amount (6 vol%) of wood and the dominating deformation process is debonding. The fact that debonding is dominating in composites not containing MAPP, i.e. at poor adhesion, is shown by Figure 9c in which debonding dominates accompanied by limited pull-out and fiber fracture. At strong adhesion, the main deformation process is the fracture of the fibers (Figure 9d) which is further facilitated by the continuously increasing stiffness with increasing fiber content. Larger stiffness results in smaller deformability in spite of the presence of the elastomer and large fiber content increases the probability of fiber related processes. The domination of these latter and the additive effect of the two components are demonstrated quite convincingly by Figure 10 in which impact resistance is plotted against the relative ratio of the two components, i.e. elastomer (φ_e) and wood (φ_w). A unique and very close correlation is obtained with larger deviations at large elastomer and small wood content. In order to address our main question, i.e. the possibility of simultaneously increasing impact resistance and stiffness, the former property is often plotted against the latter. The correlation of the two quantities is presented for the series of composites discussed throughout this paper in Figure 11. Supporting very much the conclusions of the previous figure, the composites can be divided into two groups, to those containing an elastomeric component and to the two prepared without it. We can see that impact resistance is dominated by fiber related processes, debonding or fiber fracture, and failure is accompanied by very small energy consumption in the latter. This demonstrates again the importance of crack propagation against crack initiation, since resistance against the latter increased quite significantly with fiber content especially in the case of good adhesion (see Figure 7). The other three sets of composites show the usual inverse correlation of

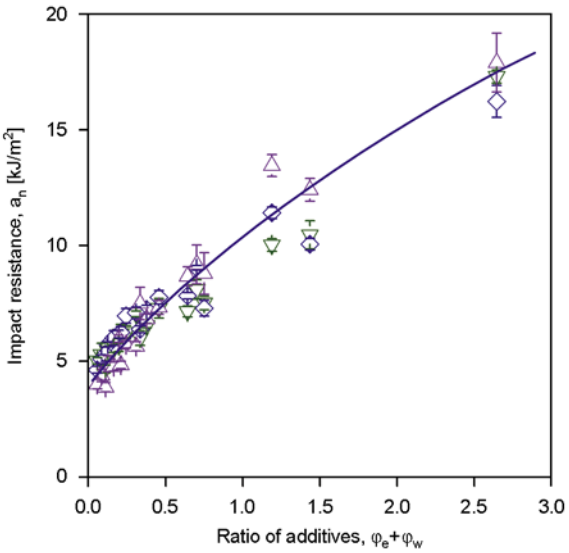

Figure 10. Impact resistance of multicomponent PP/wood composites plotted against the ratio of elastomer and wood volume fractions (φ_e/φ_w). Symbols are the same as in Figure 1. Results obtained at all elastomer contents (5, 10 and 20 wt%) are plotted in the figure.

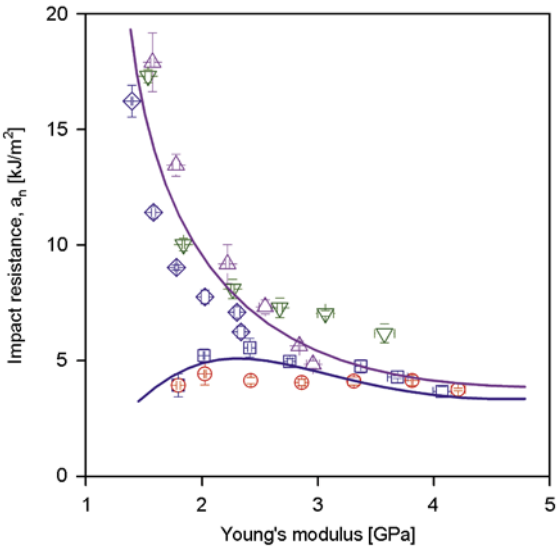

Figure 11. Correlation of impact resistance and stiffness for the five series of composites discussed in the paper. Composition and symbols are the same as in Figure 1.

the two properties characteristic for most structural materials. Very little deviation is observed from the general tendency, but based on the figure we may conclude that separate dispersion is slightly more advantageous than the embedding of the particles through the use of a functionalized elastomer. Figure 11 clearly proves that wood cannot be used in the usual large amounts in composites intended for application in which large stiffness and toughness is required. Failure occurs very easily even at small wood contents because of easy debonding and fracture of the particles caused by their large size. Further study is needed, however, to check the effect of embedding, since all results indicated only limited extent of encapsulation in our composites in spite of the use of the functionalized elastomer.

4. Conclusions

The study of the structure of three-component PP/wood/elastomer composites showed that the components are dispersed independently of each other even when a functionalized elastomer is used for impact modification, at least under the conditions of this study. The stiffness of the composites increases with wood content, but good adhesion, i.e. coupling is needed to improve strength. Impact resistance does not change much as a function of wood content in PP/wood composites, but decreases drastically from the very high level of the PP/elastomer blend to almost the same value as obtained without the impact modifier. Increasing stiffness and fiber related micromechanical deformation processes lead to small fracture toughness at large wood content. These processes depend mainly on PP/wood adhesion; debonding and pull-out take place at poor adhesion, while fiber fracture dominates when adhesion is strong. Composites with sufficiently large impact resistance cannot be prepared in the usual range of wood contents (50–60 wt%). Separate dispersion of the components seems to favor somewhat larger impact resistance, but the effect is slight and needs further checking.

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