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Polypropylene hybrid composites: structure and properties

Thesis book

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1. Introduction

Plastics are the preferred materials in most applications because of their relatively low weight, low price and ease of processing. Polypropylene (PP) is one of the favored materials in the automotive industry, but it is used extensively in many other areas as well. Besides being relatively cheap, its property profile is excellent, it is light, stiff with good strength and acceptable deformability. The growth rate of PP production is one of the largest among all commodity polymers[1]. Its further advantage is that it can be modified in various ways to extend its property profile even further.

Structural applications, especially in the automotive industry, often require large stiffness and impact resistance simultaneously. However, an inverse correlation exists between these two properties for most structural materials including plastics, metals and ceramics[2] thus the requirement is difficult to satisfy. The impact strength of polypropylene homopolymers is usually small, around 2 kJ/m^2; the requirement is often 10-15 kJ/m^2 or larger. The combination of a stiffness of 2-4 GPa and impact resistance larger than 15 kJ/m^2 is often the targeted property profile of the materials used in the automotive industry.

Our research group was requested by Borealis AG, which is one of the largest manufacturers of polyolefins in the world, to develop fiber reinforced PP-based composite material that meets the requirements mentioned above. The Laboratory of Plastics and Rubber Technology of the Department of Physical Chemistry and Materials Science at the Budapest University of Technology and Economics together with the Polymer Physics Research Group of the Institute

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[1] Talarico G, De R, Claudio, Auriemma F: Tacticity, Regio and Stereoregularity. In: Karger-Kocsis J, Bárány T, editors. Polypropylene Handbook: Morphology, Blends and Composites. 1 ed: Springer International Publishing, p. 1-35 (2019)

[2] Callister WD, Rethwisch DG: Materials Science and Engineering: An Introduction. New York: John Wiley & Son (2007)

of Materials and Environmental Chemistry at the Eötvös Lóránd Research Network have huge experience in the development and study of heterogeneous polymeric materials including short fiber reinforced composites. Following the international trends in polymer science and industry, a number of projects related to fiber reinforced composites have been started and completed at our Laboratory in the last years.

2. Background

As mentioned in the introduction, the aim of the research work was to produce composites with large stiffness and impact resistance simultaneously. Impact resistance can be increased by blending, by the introduction of elastomers; often ethylene-propylene (EPR) or ethylene-propylene-diene (EPDM) copolymers are used for the modification of PP[3]. However, the addition of elastomers decreases the stiffness of the material to below 1 GPa at large elastomer content, which is not acceptable in many applications[3]. Particulate fillers increase its stiffness and heat deflection temperature[4], while fiber modification usually results in the simultaneous increase of stiffness and strength[5]. In this Thesis we focus on short fiber reinforced composites. The principle of fiber reinforced composites is simple; stiff and strong fibers carry the load while the polymer matrix transmits it among the fibers. The stiffness and strength of the fibers traditionally used in composites are at least two orders of magnitude larger than the corresponding properties of the matrix used. The properties of all heter-

[3] Yu CT, Metzler DK: Metallocene plastomers as polypropylene impact modifiers. In: Karian H, editor. Handbook of polypropylene and polypropylene composites, revised and expanded. Boca Raton: CRC Press, p. 200-250 (2003)

[4] Móczó J, Pukánszky B: Particulate filled polypropylene: structure and properties. In: Karger-Kocsis J, Bárány T, editors. Polypropylene handbook: morphology, blends and composites. Cham: Springer International Publishing, p. 357-417 (2019)

[5] Chu FP: Glass fiber–reinforced polypropylene. In: Karian H, editor. Handbook of polypropylene and polypropylene composites, revised and expanded. Boca Raton: CRC Press, p. 281-351 (2003)

ogeneous polymers are determined by the same four factors: component properties, composition, structure and interfacial adhesion. All four are equally important in determining composite properties. In order to achieve optimum performance and economics these factors must be optimized.

Glass (GF) and carbon fiber (CF) reinforced polymers have been used for decades[4,6], but increasing environmental awareness and some economic aspects created much interest in natural fiber and wood reinforcement[7]. These fibers are not as stiff and strong as glass or carbon[8], but they have considerable advantages including their natural origin, environmental benefits, small density and low price[9]. The proper selection of the matrix polymer, the fiber and composition may lead to the required strength and stiffness for most applications however, impact resistance cannot be increased by this approach. The answer of the industry to the problem is the simultaneous use of an elastomer to increase impact resistance and a filler or fiber to improve stiffness[9,10]. The combination of the three component may result in complicated structures, and properties can vary in a wide range depending on component properties and structure[11]. Optimization resulted in several commercial grades with acceptable properties, which contain fillers or fibers to improve stiffness. Unfortunately, attempts to use wood in such multicomponent materials proved unsuccessful, although the

[6] Hopmann C, Michaeli W, Puch F: Experimental investigation on the influence of the composition on the morphology and the mechanical properties of short glass fiber-reinforced polypropylene nanocomposites. *Polym Compos* **33**, 2228-2235 (2012)

[7] Suddell BC, Evans WJ: Natural fiber composites in automotive applications. In: Mohanty AK, Mishra M, Drzal LT, editors. Natural fibers, biopolymers, and biocomposites. Boca Raton: CRC Press, p. 231-259 (2005)

[8] Clemons C: Raw materials for wood-polymer composites. In: Oksman K, Sain M, editors. Wood-polymer composites. Boca Raton: CRC Press LLC, p. 1-22 (2008)

[9] Sang L, Zheng G, Hou W, Yang X, Wei Z: Crystallization and mechanical properties of basalt fiber-reinforced polypropylene composites with different elastomers. *J Thermal Anal Calorym* **134**, 1531-1543 (2018)

[10] Stamhuis JE: Mechanical properties and morphology of polypropylene composites. Talc-filled, elastomer-modified polypropylene. *Polym Compos* **5**, 202-207 (1984)

[11] Kolárik J, Lednicky F, Jancár J, Pukánszky B: Phase structure of ternary composites consisting of polypropylene/elastomer/filler. Effect of functionalized components. *Polym Commun* **31**, 201-204 (1990)

stiffness of the material reached the desired level, its impact resistance remained invariably small[12,13,14,15]. The detailed analysis of local deformation and failure processes revealed that depending on the strength of interfacial adhesion either the debonding of large wood particles or their fracture led to the premature, catastrophic failure of the composites at very low energy consumption[16,17].

The previous approaches, as the addition of elastomer or the use of stiff fibers, to achieve the requirements have not worked, therefore we introduced a completely new approach to produce hybrid composites using various synthetic or natural stiff fibers with polymeric fibers in composites to increase stiffness and impact resistance simultaneously. The application of polymeric fibers is not widespread in the field of composite development, but because of their unique characteristics they provide quite good impact resistance for composite materials.

3. Goals

The main goal of this study was to develop composite materials which have both large stiffness and impact resistance. These composites can be used at several areas in the industry as we hope. Numerous papers have been published

[12] Keledi G, Sudár A, Burgstaller C, Renner K, Móczó J, Pukánszky B: Tensile and impact properties of three-component PP/wood/elastomer composites. *Express Polym Lett* **6**, 224-236 (2012)
[13] Oksman K, Clemons C: Effects of elastomers and coupling agent on impact performance of wood flour-filled polypropylene. Fourth International Conference on Woodfiber-Plastic Composites. Madison: Forest Products Society, p. 144-155 (1997)
[14] Sudár A, Burgstaller C, Renner K, Móczó J, Pukánszky B: Wood fiber reinforced multicomponent, multiphase PP composites: Structure, properties, failure mechanism. 103, 106-112 (2014)
[15] Sudár A, Renner K, Móczó J, Lummerstorfer T, Burgstaller C, Jerabek M, Gahleitner M, Doshev P, Pukánszky B: Fracture resistance of hybrid PP/elastomer/wood composites. *Compos Struct* **141**, 146-154 (2016)
[16] Sudár A, Burgstaller C, Renner K, Móczó J, Pukánszky B: Wood fiber reinforced multicomponent, multiphase PP composites: Structure, properties, failure mechanism. 103, 106-112 (2014)
[17] Sudár A, Renner K, Móczó J, Lummerstorfer T, Burgstaller C, Jerabek M, Gahleitner M, Doshev P, Pukánszky B: Fracture resistance of hybrid PP/elastomer/wood composites. *Compos Struct* **141**, 146-154 (2016)

on the study of the mechanical properties of short fiber reinforced polymer composites, but very few presented a detailed analysis of the local deformation processes taking place in them under external load. The better understanding of these processes and correlations is essential in the development of composite materials. In this Thesis we focus among others on the identification of the deformation processes occurring in natural and synthetic short fiber reinforced composites. One of our goals was to check that whether wood flour, which is derived from natural and renewable resources could replace carbon or glass fibers in composites. An another unanswered question was the effect of the flexible polymer fibers on the mechanical properties of composites and the determination of the local deformation processes occurring around the fibers during the failure, which is essential to know for the development of hybrid composites.

4. Materials and methods

Two polypropylene grades were used as matrix in the experiments. One was a homopolymer (hPP), the other polymer, a heterophasic ethylene-propylene copolymer (ePP) or reactor blend, containing 32 wt% elastomer. A polypropylene functionalized with maleic anhydride (MAPP) was used for coupling to improve interfacial adhesion. Five different polymer fibers were applied in the study, four of them were poly(vinyl alcohol) (PVA) and one of them was poly(ethylene terephthalate) (PET) fiber. Various synthetic and natural fibers were applied as reinforcements, carbon fibers (CF), glass fibers (GF), wood flour (W), flax (F) and sugar palm fiber (SP). The particle size of the wood fibers was determined by laser light scattering, while in the other cases the fiber length and aspect ratio were determined by measuring fibers individually on micrographs recorded by scanning electron (SEM) or digital optical microscopy (DOM).

The composition of composites containing only one type of fiber varied from 0 to 60 wt% in 5 wt% steps. The hybrid composites always contained 20 wt% of the reinforcing stiff fiber (carbon, glass, wood, flax, sugar palm) and the amount of the polymer fiber (PET, PVA) changed from 0 to 40 wt% in 5 wt% steps. The components, i.e. the fibers and the matrix polymer were homogenized in a twin-screw compounder. Wood flour, flax and sugar palm fibers were dried before extrusion at 105 °C for 4 hours, while the PVA fibers at 80 °C for 4 hours in a vacuum oven. Extrusion was repeated once in order to increase homogeneity. The granulated composites were injection molded into standard tensile bars of 4 mm thickness. The specimens were stored at ambient temperature (25 °C, 50 % RH) for one week before further testing.

The mechanical properties of the composites were characterized by tensile and impact testing. Local deformation processes were followed by acoustic emission (AE) measurements. Impact resistance was characterized by the notched Charpy impact strength. The unnotched impact strength of the specimens was also determined under the same conditions. Instrumented impact testing was carried out on notched specimens and also on unnotched specimens. The structure of the composites and deformation mechanisms were studied by scanning electron microscopy by recording micrographs on fracture surfaces created during tensile and fracture testing, respectively. In certain cases, the possible attrition of the fibers was checked on compression molded thin films by digital optical microscopy. The thermal properties of the PVA fibers were characterized by differential scanning calorimetry (DSC).

5. Results

Since to our greatest surprise hardly any publication is available in the open literature which compares wood to traditional fibers, i.e. to glass or carbon, therefore in the first stage of the work we studied the effect of various fibers as reinforcements for PP used in structural applications and examined the possibility whether carbon or glass fibers can be replaced with wood flour. The attention was mainly focused on the stiffness and impact resistance of the composites. Besides the identification of the most important factors determining these properties as well as the study of the effect of coupling on them, special attention was paid to local deformation processes and their relationship to macroscopic properties. The results proved that all three fibers have advantages and drawbacks. The stiffness of the composites depends on the modulus of the fiber, their aspect ratio, orientation and on composition, but coupling does not influence it practically at all. Properties measured at larger deformations, on the other hand, are influenced quite strongly by interfacial adhesion and local deformation processes related to the fibers. The effect of adhesion depends on the type of the fiber. Large wood particles debond from the matrix very easily without coupling, while their fracture is the dominating process at good adhesion, which leads to immediate failure in PP/wood composites. Increased interfacial adhesion improves reinforcement, but decreases impact resistance in carbon fiber reinforced composites. The glass fiber used in the study offers good adhesion even without coupling. Due to the natural and renewable character as well as low price of wood, it is a good reinforcement when the increase of stiffness is the main goal of modification, but it cannot be used in applications in which impact resistance is important. Carbon fibers are rather expensive and their composites are very stiff, but otherwise they possess intermediate properties. If adhesion is good, glass fibers offer a good balance of properties including stiffness, strength and impact resistance as demonstrated by **Fig. 1**.

The use of wood, glass and carbon fibers lead to an increase of impact strength, and the originally small, 2 kJ/m^2, impact resistance of PP increased to 4 kJ/m^2 in the presence of wood or carbon fiber, and even to 8 kJ/m^2 upon reinforcement with glass fibers. However, the required impact strength is often much larger, exceeds 15 or 20 kJ/m^2. Therefore, we tried a new concept to increase the fracture resistance of the composites using polymer fibers. The results showed that these fibers increase stiffness only moderately; because of the flexibility of the fibers, they do not orientate in the direction of the load. Consequently, the aspect ratio of the fibers is less important than in the case of stiff fibers like glass and carbon. Composite strength changes in a wider range and it depends on coupling. PP composites containing PVA and flax fibers have the largest strength when adhesion is good. The deformability of the composites also varies considerably, more plastic deformation occurs in composites prepared with the PET and PVA fibers. Compared to traditional, stiff fibers, impact resistance can be improved significantly with PET and PVA fibers; an impact strength as large as 30 kJ/m^2 can be

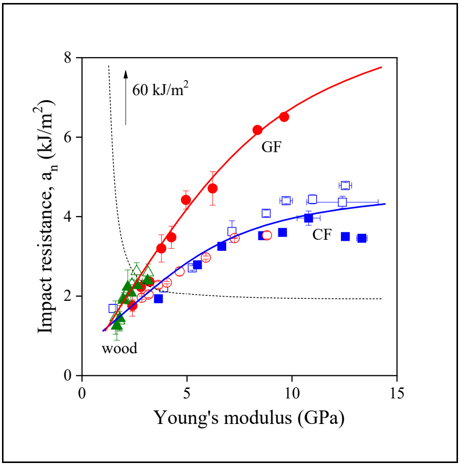

Fig. 1 *Correlation between the impact resistance and stiffness of fiber reinforced PP composites. Symbols: (□,■) CF, (○,●) GF, (△,▲) wood flour. Empty: without coupling, poor adhesion; full: with MAPP, good adhesion. The broken line indicates the general correlation of the two properties determined in a previous study*[17].

achieved with PVA fibers as demonstrated by **Fig. 2**. Fiber type and coupling influence the local deformation processes occurring around the fibers considerably, but does not change fracture energy much. The combination of debonding and subsequent plastic deformation is the most efficient in the improvement of impact strength. Fiber fracture dominates in wood and flax composites at good adhesion, but it does not consume sufficient energy to increase impact resistance.

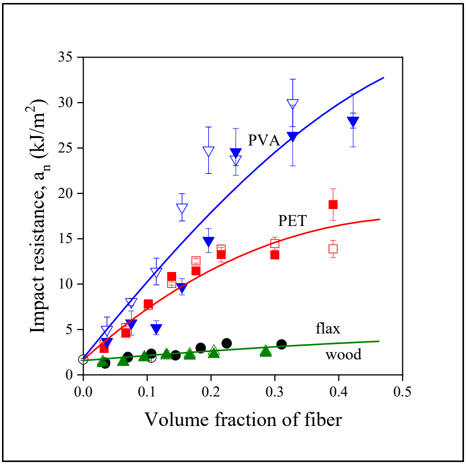

Fig. 2 *Correlation between the notched Charpy impact resistance of PP/organic fiber composites and fiber content. Effect of coupling. Symbols:* (△,▲) *wood,* (○,●) *flax,* (□,■) *PET,* (▽,▼) *PVA; empty symbols: without MAPP, poor adhesion; full symbols, with MAPP, good adhesion.*

The use of polymeric fibers influences the impact resistance of PP considerably. Since only a few publications are available on synthetic fiber reinforced PP composites[18,19,20], it seems to be obvious to explore further the possibilities of modifying polypropylene with polymeric fibers. Two polymer matrices were used, the characteristics of which differed widely, one was a homopolymer (hPP), while the other polymer was a heterophasic ethylene-propylene copolymer (ePP). The influence of coupling was also studied. The results

[18] Asgari M, Masoomi M: Thermal and impact study of PP/PET fibre composites compatibilized with glycidyl methacrylate and maleic anhydride. *Compos Part B Eng* **43**, 1164-1170 (2012)
[19] Santos P, Pezzin SH: Mechanical properties of polypropylene reinforced with recycled-PET fibres. *J Mater Process Technol* **143-144**, 517-520 (2003)
[20] López-Manchado MA, Arroyo M: Effect of the incorporation of pet fibers on the properties of thermoplastic elastomer based on PP/elastomer blends. *Polymer* **42**, 6557-6563 (2001)

showed that different inherent matrix characteristics result in dissimilar combination of properties. The PVA fiber proved to be more efficient as reinforcement in both matrices than the PET fiber; the former increases both stiffness and strength more than the latter. The strength of the composites depends very much on interfacial adhesion. The effect of the factors studied (fiber and matrix type, interfacial adhesion) is stronger and more complicated in the case of impact resistance. Both fibers improve impact strength in the hPP matrix (see **Fig. 3**), but the effect depends on fiber type and interfacial adhesion. The changes in impact resistance cover a wide range in the ePP matrix; impact strength may increase or decrease depending on fiber type and adhesion, however, it remains always smaller than the fracture resistance of the matrix. Interfacial adhesion influences local deformation processes very strongly and these processes determine the macroscopic properties of the composites. Shear yielding and cavitation occur in the matrix, while debonding, fiber pullout and fracture are the main deformation mechanisms in the composites. Shear yielding induced by the debonding of the fibers consumes much energy, the fracture of the fibers is less efficient in the increase of impact resistance, while cavitation in the ePP polymer results in inferior impact strength. A combination of stiffness and impact resistance in excess of 3 GPa and 40 kJ/m^2, respectively, can be obtained in PP homopolymers modified with PVA fibers,

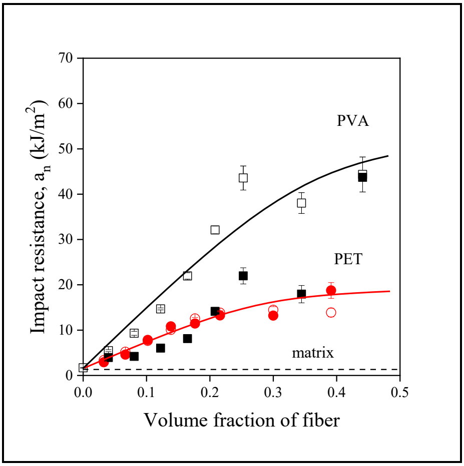

Fig. 3 *Influence of composition on the notched Charpy impact resistance of fiber reinforced PP composites. Symbols: (□,■) hPP/PVA, (○,●) hPP/PET; empty symbols: poor adhesion (without MAPP), full symbol: good adhesion (with MAPP).*

while this type of modification is less advantageous for elastomer modified PP grades.

We achieved considerable improvement in the stiffness of PP composites by using carbon or glass fibers, but the impact resistance of these composites remained relatively small. We proved that using polymeric fibers in polypropylene composites improves impact resistance significantly, but it increases the stiffness only slightly. The combination of traditional reinforcements and polymeric fibers could be a beneficial and new concept in composite development in order to prepare composites with large stiffness and impact resistance. We prepared hybrid PP composites containing glass or carbon fibers and a polymer, PET, fiber simultaneously. PP/wood/PET hybrid composites were used as reference in the study. The results proved that improved impact resistance can be achieved in this way with all stiff fibers and not only for wood as shown earlier[21]. Approximately the same impact resistance can be achieved with PET fibers, which depends on the amount of PET fiber used; impact strength as large as 15 kJ/m^2 can be obtained with the approach. The large impact resistance is the result of local deformation processes initiated by the polymeric fiber. Irrespectively of the strength of interfacial adhesion, the main local process is the debonding and/or pullout of the PET fibers, which facilitates also the plastic deformation of the matrix polymer. The combination of these local processes results in large energy consumption and increased impact resistance. Other properties depend on the type of the reinforcement and on interfacial adhesion. The application of carbon fibers results in composites with large stiffness, while the strength of glass fiber reinforced composites is the largest at good adhesion. Impact resistance is plotted against the stiffness of the composites in **Fig 4**. Except

[21] Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Pukánszky B, Renner K: Impact modification of PP/wood composites: A new approach using hybrid fibers. *Express Polym Lett* **13**, 223-234 (2019)

for the CF composites, the two quantities are independent of each other; the increase of impact resistance not being accompanied by a decrease in stiffness. The proper selection of components, composition and interfacial adhesion allows the adjustment of overall composite properties in a relatively wide range.

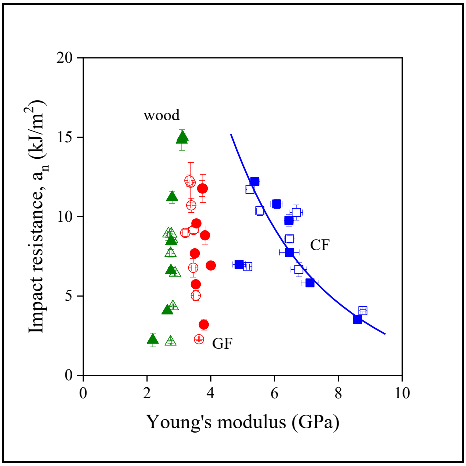

Fig. 4 *Correlation between the Young's modulus and impact resistance of hybrid PP composites containing PET fiber. Symbols: (□,■) CF, (○,●) GF, (△,▲) wood; empty symbols without MAPP (poor adhesion), full symbols with MAPP (good adhesion).*

Environmental considerations and the outstanding properties of wood/PET hybrid composites encouraged us to verify the validity of the approach of hybridization also for PP composites reinforced not only with wood but also with other natural fibers. Although, an attempt was made by Sudár et al.[20] to use a PP reactor blend containing as much as 33 wt% elastomer to improve the impact resistance of PP/wood composites, fracture strength remained at a low level, despite the large elastomer content. Many attempts were made earlier also to combine traditional reinforcements and natural fibers[22,23,24], but composites with prominently good properties could not be produced. In our work

[22] Kalaprasad G, Thomas S, Pavithran C, Neelakantan NR, Balakrishnan S: Hybrid effect in the mechanical properties of short sisal/glass hybrid fiber reinforced low density polyethylene composites. *J Reinf Plast Compos* **15**, 48-73 (1996)

[23] Rozman HD, Tay GS, Kumar RN, Abubakar A, Ismail H, Ishak ZAM: Polypropylene hybrid composites: a preliminary study on the use of glass and coconut fiber as reinforcements in polypropylene composites. *Polym Plast Technol Eng* **38**, 997-1011 (1999)

[24] Samal SK, Mohanty S, Nayak SK: Polypropylene–bamboo/glass fiber hybrid composites: fabrication and analysis of mechanical, morphological, thermal, and dynamic mechanical behavior. *J Reinf Plast Compos* (2008)

we verified the validity of the approach of hybridization also for PP composites reinforced with natural and PET fibers. Flax and sugar palm fibers were applied as reinforcements with wood flour being used as reference. The attention was focused less on conventional properties, stiffness and strength, but more on fracture resistance. Local deformation processes were analyzed to determine the mechanism of deformation and failure. This study proved that the concept works with other natural fibers as well; the impact strength of also flax and sugar palm fiber reinforced composites could be improved considerably. The mechanism of impact modification is always the same, the debonding or pullout of the PET fibers initiates the local yielding of the matrix, which consumes considerable energy. The fracture of PET fibers might also contribute to energy absorption. The type of natural fiber used as reinforcement does not influence the effect, the amount of PET fibers determines the fracture resistance of the composites as demonstrated by **Fig. 5**. Improving interfacial adhesion by coupling increases strength and slightly improves impact resistance.

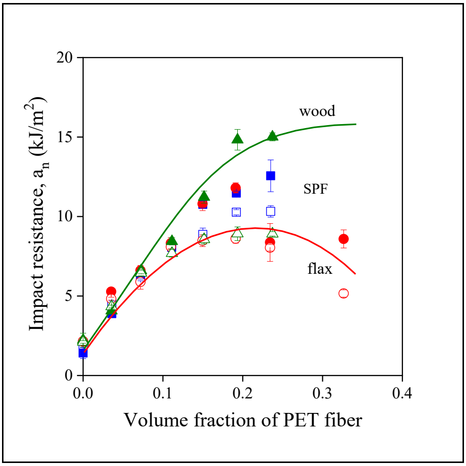

Fig. 5 *Composition dependence of the impact resistance of hybrid PP composites containing PET fiber. Natural fiber content 20 wt%. Symbols: (△,▲) wood, (○,●) flax, (□,■) sugar palm fiber; empty symbols: poor adhesion, without MAPP, full symbols: good adhesion, with MAPP.*

In our previous projects we found that synthetic polymer fibers, PET and PVA, increased the impact resistance of PP when they were used as single additives, and the PET fibers proved efficient impact modifiers in hybrid composites reinforced either with traditional glass or carbon fibers, or with various natural fibers like flax, sugar palm fiber or wood flour as well. Although PVA fibers were also effective in two-component PP composites, no proof is so far available that they improve impact resistance in the presence of reinforcing fibers as well. Consequently, one of our goals was to check and – if possible – verify the concept of using PVA fibers for the impact modification of PP composites reinforced with the most frequently used fibers, i.e. with glass, carbon and wood. The results obtained prove that the novel concept of using synthetic fibers for the purpose can be applied successfully also with PVA fibers. The impact resistance of the composites increases with increasing PVA fiber content (see **Fig. 6**). The extent of improvement in impact strength depends on fiber type and content, but also on interfacial adhesion which strongly influences the local deformation processes occurring around the fibers during fracture. Both the reinforcing and the synthetic fiber take part in these processes and contribute to energy consumption. Debonding and the subsequent plastic deformation of the matrix consumes energy the most efficiently, but the fracture of the PVA fibers also requires energy, thus PVA fibers improve impact resistance both at poor and good adhesion. The approach allows the design of materials for structural applications, the combination of a stiffness of 4-6 GPa and an impact resistance of 20-25 kJ/m^2 exceeds the properties of most PP composites available on the market.

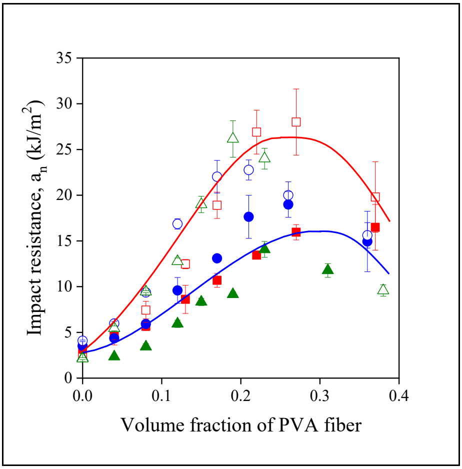

Fig. 6 *Notched Charpy impact resistance plotted against the PVA fiber content of hybrid PP composites. Symbols: (△,▲) wood, (□,■) GF, (○,●) CF; empty symbols: without MAPP, poor adhesion; full symbols: with MAPP, good adhesion.*

6. New scientific results

1. In spite of numerous claims and statements published about the benefits and superior performance of natural fibers, by the comparative study of three reinforcing fibers in polypropylene we pointed out the first time, that traditional glass or carbon fibers cannot be replaced automatically with a natural reinforcement. All fibers have advantages and drawbacks and must be selected according to the requirements of the application in mind. Wood has environmental benefits, carbon fibers provide large stiffness, while glass fibers offer a good balance of properties.

2. The comparison of the effect of four organic fibers on the properties of polypropylene showed that although such fibers do not increase the stiffness of the polymer very much, synthetic polymer fibers improve impact resistance considerably. The observation led to the development of the novel concept of impact modification with synthetic fibers which has not been used before.

3. By the study of the effect of two different synthetic fibers on the reinforcement and impact modification of PP in two different matrix polymers, we proved that the extent of reinforcement and impact modification depends on the properties of the matrix polymer and on interfacial adhesion. The novel principle of impact modification with synthetic fibers can be applied successfully in stiff PP matrices (homopolymers), but it is less efficient in random copolymers or reactor blends.

4. With the help of systematic experiments we proved that the novel concept of impact modification with synthetic fibers can be used also in hybrid

composites containing a reinforcing and a synthetic fiber. The combination of the two fibers results in large stiffness and impact resistance simultaneously, very much sought for by the industry. We also showed that the mechanism of impact modification is the debonding of the fibers and the subsequent plastic deformation of the matrix polymer.

5. In another series of experiments we showed that the new approach of impact modification can be applied in hybrid composites reinforced with any kind of natural fibers. The extent of impact modification does not depend on the type of the natural reinforcement, but only on the amount of the synthetic fiber (PET), since the former does not contribute to impact resistance.

6. By the analysis and modeling of the fracture process in PP hybrid composites we proved that both the reinforcing and the synthetic fiber can contribute to the modification of impact resistance. The contribution depends on the characteristics of the components and interfacial adhesion that determines the local deformation processes initiated around the fibers by the external load. We showed that even better property combinations can be achieved with PVA than with PET fibers.

7. During the work done in the thesis we paid increased attention to local, micromechanical deformation processes and pointed out that several of such processes (yielding, cavitation, debonding, fiber fracture, pullout) can take place simultaneously or consecutively in polymer composites during deformation and that these processes determine the macroscopic properties of the composites. Debonding and pullout accompanied by the

plastic deformation of the matrix consume much energy and result in efficient impact modification. In spite of their importance, local, micromechanical processes are largely neglected in the study of polymer composites in the literature.

7. Possible applications

Our research group have been taking part in international project together with Borealis AG, which is one of the largest manufacturers of polyolefins in the world producing also short fiber reinforced raw materials. The purpose of the cooperation is to develop materials which satisfy the requirements mentioned earlier for composite materials. The participation of industry offers the possibility to introduce the results into practice. Composites developed by us are already potential raw materials for bumpers in the automotive industry. The research work began in 2014, and since then Borealis was granted a number of patents based on the results of our research work[25].

8. Publications

8.1 The Thesis is based on the following papers

1. Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Faludi G, Móczó J, Pukánszky B: Comparative study of fiber reinforced PP composites: Effect of fiber type, coupling and failure mechanisms. *Compos Part A Appl Sci Manuf* **133**, 105895, DOI: https://doi.org/10.1016/j.compositesa.2020.105895 (2020), IF: 6.444, C: 3

2. Várdai R, Ferdinánd M, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Faludi G, Móczó J, Pukánszky B: Effect of various organic fibers on the stiffness, strength and impact resistance of polypropylene: a comparison. *Polym Int* **70**, 145-153, DOI: https://doi.org/10.1002/pi.6105 (2021), IF: 2.574, C: 0

[25] Lummerstorfer T., Jerabek M., Hochradi S., Pretschuh C., Renner K., Sobczak L., Stockreiter W., Pukanszky B., Moczo J.: Fiber Reinforced Polypropylene Composite, *United States Patent*, Patent No.: US 10,752,762 B2 (2020)

3. Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Faludi G, Móczó J, Pukánszky B: Reinforcement of PP with polymer fibers: Effect of matrix characteristics, fiber type and interfacial adhesion. *Polymer* **190**, 122203, DOI: https://doi.org/10.1016/j.polymer.2020.122203 (2020), IF: 4.231, C: 4

4. Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Faludi G, Móczó J, Pukánszky B: Impact modification of fiber reinforced polypropylene composites with flexible poly(ethylene terephthalate) fibers, *Polym Int*, DOI: https://doi.org/10.1002/pi.6210 (2021), IF: 2.574, C: 0

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8.2 Manuscripts used for the preparation of the Thesis

1. Várdai R, Ferdinánd M, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Faludi G, Móczó J, Pukánszky B: Impact modification of hybrid PP composites with PVA fibers. Submitted to *Compos Struct* (2021)

8.3 Other publications related to the Thesis

1. Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Pukánszky B, Renner K: Impact modification of PP/wood composites: A new approach using hybrid fibers. *Express Polym Lett* **13**, 223-234, DOI: https://doi.org/10.3144/expresspolymlett.2019.19 (2019), IF: 3.292, C: 9

8.4 Other publications not related to the Thesis

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8.5 Conference presentations

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2. Várdai R, Renner K, Pukánszky B: Deformációs folyamatok hatása PP/faliszt kompozitok ütésállóságára, MTA Műanyag és Természetes Polimerek Munkabizottsági ülése, April 27, 2016, Budapest, Hungary
3. Renner K, Várdai R, Lummerstorfer T, Jerabek M, Pretschuh C, Doshev P, Gahleitner M, Móczó J, Pukánszky B: Deformation processes in PP based hybrid composites, Eurofillers 2017, April 26, 2017, Heraklion, Greece
4. Várdai R, Renner K, Móczó J, Pukánszky B: Szerkezet, tulajdonság összefüggések keresése hagyományos és hibrid kompozitokban, MTA TTK AKI szeminárium, June 6, 2017, Budapest, Hungary
5. Várdai R, Horváth F, Balogh R: High resolution deposition of the conductive polymer paths on the flexible substrate for electronics and regenerative medicine, Soft Skills Training for Young Scientists, Project Proposal Competition, October 16, 2017, Bratislava, Slovakia
6. Várdai R, Renner K: Szálerősítésű hibrid kompozitok: ütésállóság növelése PET szál segítségével, SPE MSC Diplomadíj pályázat, November 16, 2017, Budapest, Hungary
7. Várdai R, Pukánszky B, Renner K: Polimer szál erősítésű kompozitok: szerkezet, tulajdonságok, deformáció, MTA TTK AKI szeminárium, March 13, 2018, Budapest, Hungary
8. Lummerstorfer T, Várdai R, Renner K, Jerabek M, Doshev P, Gahleitner M, Pukánszky B: Heterophasic PP/wood composites: interfacial interactions and their relation to deformation processes, BiPoCo 2018, September 3, 2018, Balatonfüred, Hungary
9. Kanyó L, Várdai R, Renner K: Damage process in PP/wood composites analyzed by acoustic emission, BiPoCo 2018, September 5, 2018, Balatonfüred, Hungary
10. Kovács Á, Várdai R, Faludi G, Renner K: Deformation processes and impact resistance in poly(lactic acid) based composites, BiPoCo 2018, September 5, 2018, Balatonfüred, Hungary
11. Várdai R, Lummerstorfer T, Pretschuh C, Jerabek M, Gahleitner M, Pukánszky B, Renner K: Impact modification of PP/wood composites: a new approach using PET fibers, BiPoCo 2018, September 5, 2018, Balatonfüred, Hungary

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13. Várdai R, Móczó J, Pukánszky B: Biopolymers, Their Blends, Composites and Application, IC-AMME 2018, September 27, 2018, Bali, Indonesia
14. Várdai R, Kovács B Z, Faludi G, Móczó J, Pukánszky B: Természetes szál-erősítésű kompozitok ütésállóságának módosítása PET szál segítségével, MTA TTK AKI szeminárium, April 9, 2019, Budapest, Hungary
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