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Polymer composites reinforced lignocellulosic fiber: Particle characteristics, interactions, performance

Thesis book

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1. Introduction

Plastics play a very important role in industry and in our everyday life. They are versatile and satisfy all expectations from cheap, commodity products to high-tech applications like the aerospace industry, electronics or healthcare. Nevertheless, the abundant use of commodity plastics results in a large amount of consumer waste which raises environmental concerns in the public. The increasing environmental awareness of the population pushes industry towards the application of more environmentally friendly materials. The use of natural polymers like cellulose and starch has become more frequent, and numerous polymeric materials are produced based on raw materials derived from natural resources. Some of these, e.g. bio-polyethylene or bio-poly(ethylene terephthalate), are completely identical with their conventional counterparts, thus are not degradable. Poly(lactic acid), PLA, on the other hand, is entirely compostable, while it also has acceptable properties and price. Not surprisingly, the interest in this material increased tremendously both in academia and industry in recent years.

Although PLA and other biopolymers gained considerable importance recently, these materials have several drawbacks as well. Their processability is usually inferior compared with that of commodity polymers, they are heat and moisture sensitive and their properties often do not meet the requirements of demanding applications. The physical ageing of PLA leads to a rapid change of properties and to brittleness, and its low glass transition temperature limits the application of this material in many areas. As a consequence, similarly to other biopolymers, PLA is modified by various means. Deformability and brittleness is compensated by plasticization or by the use of impact modifiers. Dimensional stability and stiffness is improved by the addition of mineral fillers or short fibers. Natural fibers are used with increasing frequency to modify biopolymers,

and the combination leads to environmentally friendly materials. However, modification also results in new problems, thus the thorough study of these materials is essential for future applications.

The preparation and use of polymers containing natural fillers or reinforcements is not new in the plastics industry, but these materials went through a revival in recent years all over the world. Composites containing lignocellulosic components are known since the 1900s, especially in the building and furniture industry[1], but they are used in other areas as well. After recognizing the advantages of natural fillers and fibers, more and more research groups started to work on the replacement of glass and carbon fibers in composites and to study natural fiber reinforced thermoset resin composites[2,3,4].

Increasing quantities of thermoplastic polymers are used also as matrix materials in wood/plastic composites (WPC) in recent years. One of the main advantages of these materials is that products can be manufactured with traditional thermoplastic processing technologies for a wide range of applications. Commodity polymers are used the most often as matrices for the production of such composites[5]; the selection of the polymer depends on the intended application.

Natural reinforcements have numerous advantages. They are produced and are available in large quantities[6], and their price is low compared to traditional reinforcements and competes even with that of mineral fillers. They have

[1] Youngquist, J.A.: *Wood Handbook. Wood as an Engineering Material.* Forest Products Laboratory, Madison, 1999.
[2] Abdelmouleh, M., Boufi, S., Belgacem, M.N., Dufresne, A., Gandini, A., *J. Appl. Polym. Sci.* **98,** 974-984 (2005)
[3] Gassan, J., Bledzki, A.K., *Compos. Sci. Technol.* **59,** 1303-1309 (1999)
[4] Mishra, S., Naik, J.B., Patil, Y.P., *Compos. Sci. Technol.* **60,** 1729-1735 (2000)
[5] Markarian. J., *Plast. Additives Compound.* **4,** 18-21 (2002)
[6] Suddell, B.C.; Evans, W.J.: in *Natural Fibers, Biopolymers, and Biocomposites*, Mohanty, A.K, Mishra, M., Drzal, L.T., eds. CRC Press, Boca Raton, pp. 231-259, 2005.

large stiffness and strength, as well as low density. They are produced from renewable resources, often as byproducts. They can degrade biologically and the waste can be handled easily. On the other hand, these reinforcements are sensitive to humidity and heat, they have poor transverse strength and very poor adhesion to the matrix because of their small surface energy.

2. Background

Although thermoplastic composites reinforced with natural fibers are used in many areas, extensive research is still going on with the goal to compensate for the weaknesses of such composites, to improve properties and to decrease price. Optimization of price and performance is possible only if the factors determining the properties of the composites are known. Natural fiber reinforced polymers are heterogeneous materials. Consequently, their properties are determined by the characteristics of the components, composition, interfacial interactions and structure. All four factors are equally important and must be adjusted to achieve optimum performance and economics.

The characteristics of the matrix strongly influence the effect of the fiber on composite properties; stiffness is probably the most important matrix characteristic, while the effect of fiber properties is much more complicated and ambiguous. Chemical composition, particle characteristics, surface energy and other properties all play important roles in the determination of composite properties. Particle characteristics and particle shape are especially important in wood composites. Small particles often aggregate, while large particles adhere to the matrix very weakly and debonding, i.e. the separation of the matrix and the reinforcement, occurs very easily. Anisotropic particles orientate during processing and the extent of reinforcement depends very much on the relative direction of orientation and load.

Composition, i.e. the fiber content of composites may change in a wide range. The goal of using fibers is usually to improve stiffness and strength. This requires the introduction of the largest possible amount of filler or fiber into the polymer, but the improvement of the targeted property may be accompanied by the deterioration of others. Since various properties depend on fiber content in different ways, composite properties must always be determined as a function of composition.

The structure of particulate filled and short fiber reinforced polymers often receives less attention than it merits. The homogeneous distribution of particles in the polymer matrix is assumed in most cases. This, however, rarely occurs and often special, particle related structures develop in composites. In relation with WPC, the most significant are aggregation, orientation and attrition, i.e. the fracture of the fibers during processing. Aggregation occurs frequently and dominates at high surface energy and small particle size. Orientation is probably an even more important structural phenomenon in wood composites. All processing technologies orient anisotropic particles in smaller or larger extent, resulting in increased stiffness and strength in the direction of orientation.

Interfacial interactions are crucial in all composites, but especially in fiber reinforced polymers. Strong particle-particle interactions result in aggregation, while matrix/filler interactions lead to the development of an interphase with properties different from those of both components. Secondary forces play a crucial role in the development of both. These interactions can be modified by either physical or chemical means. One often applied, relatively old physical method is mercerization, i.e. the alkaline treatment of cellulose fibers, which might increase their tensile strength[7]. The surface modification of the fillers is perhaps even more widely applied. This method involves coating the fibers with

[7] Gassan, J., Bledzki, A.K., *J. Appl. Polym. Sci.* **71,** 623-629 (1999)

a small molecular weight nonpolar organic compound, in order to decrease particle-particle interactions. Unfortunately this approach also decreases composite strength as a consequence[8].

The purpose of chemical modification is either to decrease reactivity and polarity of the fibers, or chemical coupling. Etherification and esterification[9,10] belong to the first category. Exchanging the surface hydroxyls of cellulose to less polar groups results in decreased water absorption and aggregation, but also in weaker interfacial interactions. In order to achieve the necessary stress transfer, however, strong interaction is needed that is usually achieved by the creation of covalent bonds between the fiber and the matrix. Treatment with functionalized polymers is often applied in composites with polyolefin matrices. Their reactive groups react chemically with the surface of the fiber, while the polymer backbone of the functional polymer diffuse into the matrix, increasing the strength of the composite considerably[11,12,13]. Coupling agents[14,15], on the other hand, create covalent bonds between the fiber and the matrix by reacting chemically with both. The coupling agent must be selected accordingly, it must have functional groups to react with both components.

Interactions very often determine the local deformation processes occurring around heterogeneities. One may question the importance of these processes, but they determine the final properties, failure and performance of the entire composite. Consequently, controlling them and increasing the initiation

[8] Dányádi, L., Móczó, J., Pukánszky, B., *Compos. Part A, Appl. S.* **41,** 199-206 (2010)
[9] Baiardo, M., Frisoni, G., Scandola, M., Licciardello, A., *J. Appl. Polym. Sci.* **83,** 38-45 (2002)
[10] Freire, C.S.R., Silvestre, A.J.D., Neto, C.P., Belgacem, M.N., Gandini, A., *J. Appl. Polym. Sci.* **100,** 1093-1102 (2006)
[11] Bledzki, A.K., Fink, H.P., Specht, K., *J. Appl. Polym. Sci.* **93,** 2150-2156 (2004)
[12] Karnani, R., Krishnan, M., Narayan, R., *Polym. Eng. Sci.* **37,** 476-483 (1997)
[13] Felix, J.M., Gatenholm, P., *J. Appl. Polym. Sci.* **42,** 609-620 (1991)
[14] Gassan, J., Gutowski, V.S., Bledzki, A.K., *Macromol. Mater. Eng.* **283,** 132-139 (2000)
[15] Maldas, D., Kokta, B.V., Daneault, C., *J. Appl. Polym. Sci.* **37,** 751-775 (1989)

stress of the dominating process is of very large importance. Since often the fracture of the fibers initiate these processes and results in catastrophic failure, the selection of fibers with larger inherent strength or the improvement of this latter characteristic would lead to composites with better performance.

As natural fiber reinforced polymer composites represent a relatively new group of materials, reliable information is extremely limited on the effect of the above mentioned factors on their final properties. Reported data on interfacial adhesion are often controversial, structural phenomena are scarcely investigated and even less is known about the effect of the very complex structure of cellulose fibers on the deformation and failure of the composites. These latter are also neglected in most studies that are generally directed toward the development of materials with acceptable properties on a trial and error basis. The knowledge of the basic processes leading to the failure of the material, however, would allow their control as well as the improvement of composite characteristics, thus these could be optimized and adjusted to the intended application.

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 Institute of Materials and Environmental Chemistry at the Hungarian Academy of Sciences have vast experience in the development and study of heterogeneous polymeric materials including short fiber reinforced composites. Several papers have been published, and two PhD theses were also prepared in the Laboratory on this topic in recent years. The first dealt with interfacial interactions in polypropylene (PP)/wood composites[16]

[16]Dányádi, L.: *Interfacial interactions in fiber reinforced thermoplastic composites, PhD Thesis,* Budapest University of Technology and Economics, Budapest, 2009.

while the second focused on micromechanical deformation processes in heterogeneous polymer systems in general, and in wood reinforced PP in particular[17]. The information obtained in these studies resulted in deep knowledge on such materials and paved the way for this thesis. The Laboratory also participated in the Forest Resource Sustainability through Bio-Based-Composite Development (Forbioplast) FP7 project that started in 2008. The main role of our group was to explore possibilities and to develop methods for the improvement of the properties of PP/wood and PLA/wood composites through the modification of interfacial interactions and wood characteristics. Scientific results were summarized in publications, but some of them found their way into practical applications and even a patent is under preparation. Quite a few BSc and MSc theses form the basis of this work, which in itself is one of the most important results of the project.

3. Goals

The use of thermoplastic composites reinforced with natural fibers is now a mature technology with a considerable growth rate. Although the products are used in many areas, extensive research is still going on aiming to compensate for the weaknesses of such composites, to improve properties and decrease price. Optimization of price and performance is possible only if the factors determining the properties of the composites are known. Besides improving existing products and technologies, scientists and industry are looking for new advances as well. The interest turned towards new areas in recent years and although much research is done on WPC composites based on commodity polymers, the focus of attention shifted towards bio-related materials derived from renewable re-

[17]Renner, K.: *Micromechanical deformation processes in polymer composites, PhD Thesis*, Budapest University of Technology and Economics, Budapest, 2010.

sources. These new materials include all kinds of natural polymers and their derivatives like starch, cellulose, cellulose acetate, poly(lactic acid) (PLA), natural fibers and wood. As a consequence, the main goal of this Thesis is to extend knowledge on factors determining the properties of biocomposites reinforced with natural fibers, to establish structure-property correlations in such materials, to study and modify interfacial interactions and analyze them quantitatively, and to identify specific questions related to such fibers. In order to achieve these goals we utilize the knowledge compiled by the group on heterogeneous polymers up to now, and extend them to the new problems and materials. The final goal of the work was to establish guidelines for the development of new materials based entirely on bio-resources, and specifically on PLA and wood.

4. Materials and methods

Composites were prepared in a wide composition range using six different lignocellulosic fibers as reinforcement in a PLA matrix. Particle size was determined by laser light scattering, while aspect ratio by individually measuring particles on micrographs recorded by scanning electron microscopy (SEM). The abbreviations used for labeling the fillers indicate their origin (wood, W; microcrystalline cellulose, MCC; corn cob, CC) and ten times their aspect ratio. Two reactive coupling agents, N,N-(1,3-phenylene dimaleimide) (BMI) and 1,1-(methylene-di-4,1-phenylene)bismaleimide (DBMI) were used to improve interfacial adhesion in PLA/wood composites. In some of the experiments a phenolic resin was also applied to impregnate the fibers.

Both the matrix and the fibers were dried before composite preparation. The components were homogenized using an internal mixer. Filler content changed in a wide range. The homogenized material was compression molded into plates. All specimens were kept in a room with controlled temperature and humidity for at least one week prior further testing. In some cases a coupling

agent was also added to the composition.

Mechanical properties were characterized by the tensile testing of specimens cut from the compression molded plates. Micromechanical deformation processes were followed by acoustic emission (AE) measurements. The particle characteristics of wood and the structure as well as the deformation mechanism of the composites were investigated by SEM. Failure mechanism was studied also by polarization optical microscopy (POM) on model composites prepared for the purpose. The water absorption of impregnated wood flour was determined by the measurement of weight.

5. Results

In the first stage of the work we investigated the effect of six commercially available natural fillers with varying chemical composition and particle characteristics on the mechanical performance and failure mechanism of poly(lactic acid). Since very little reliable information has been published on interfacial interactions in PLA/wood composites, considerable attention was paid to the estimation of the strength of adhesion between the components as well. Most of the papers available on this issue[18,19] claim that interaction is weak between PLA and wood, but the results are often contradictory. Unfortunately, the direct determination of interaction is impossible in such composites, therefore three different indirect methods were used for the quantitative estimation of interfacial adhesion. Mechanical and acoustic emission measurements as well as a SEM study all indicated that contrary to most claims published in the literature interfacial adhesion is rather strong between PLA and natural fibers. Strong interfacial adhesion results in weak dependence of the extent of reinforcement on the particle characteristics of the reinforcing fibers. Both acoustic emission

[18] Bax, B., Müssig, J., *Compos. Sci. Technol.* **68,** 1601-1607 (2008)
[19] Huda, M.S., Drzal, L.T., Misra, M., Mohanty, A.K., *J. Appl. Polym. Sci.* **102,** 4856-4869 (2006)

measurements and microscopy indicated that the dominating micromechanical deformation mechanism is the fracture of the fibers and close correlation was found between the initiation stress of fiber fracture, reinforcement and ultimate composite strength.

The results acquired in the first stage of the study also revealed that besides the fracture of the fibers several other deformation mechanisms, including debonding, also take place during the deformation of PLA/wood composites. As a consequence, composite properties might be improved by increasing interactions further. Coupling of the phases is one of the most effective methods to achieve this goal, therefore a series of experiments were designed in which two reactive coupling agents, BMI and DBMI were used to improve interfacial adhesion between PLA and wood. Both compounds proved to be effective, i.e. they improved the tensile strength of the composites, as demonstrated by Fig. 1. Besides strength, stiffness and deformability also increased, supplying further proof for coupling. Due to the larger flexibility of the molecule, DBMI proved to be a more efficient coupling agent than BMI in the studied composites. However, the effect of coupling is small, as only a few very large particles debond under the effect of external load. Smaller particles seem to adhere strongly to the matrix proving our earlier conclusion that interfacial adhesion is inherently strong in

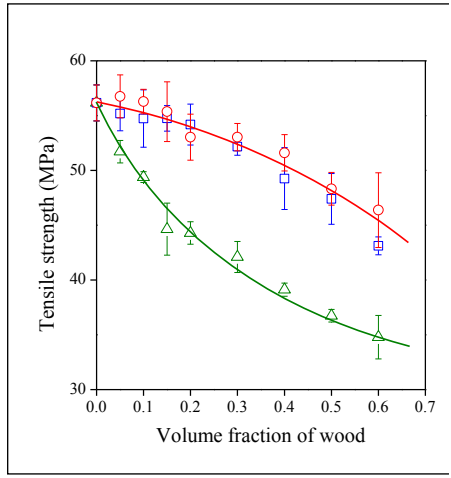
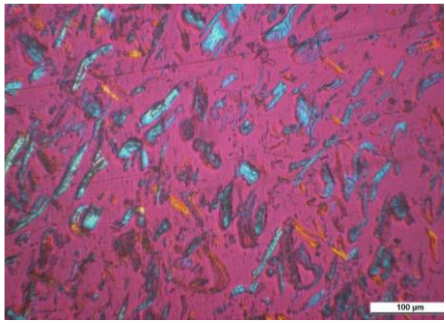


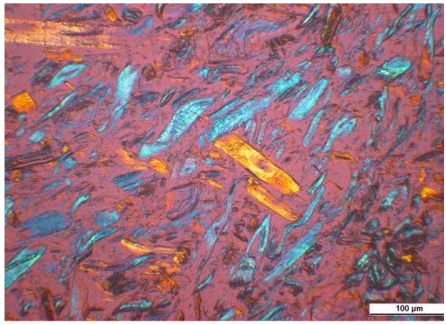
Fig. 1 *Effect of wood content and coupling on the tensile strength of PLA/wood composites; Symbols: □ BMI, ○ DBMI, △ no coupling.*

PLA/wood composites. Accordingly, the effect must depend on the size of the particles, increasing with increasing particle size.

Structural questions related to polymer/wood composites are rarely discussed in the literature. The main reason is that wood particles are large and their surface energy is small[20], thus aggregation is not expected in such composites. The small aspect ratio of the fibers decreases also the effect of orientation to an almost insignificant level. On the other hand, our earlier investigations showed that at the large wood contents used in industrial practice wood particles may touch each other purely from geometrical reasons and the effect as well as its extent depends on particle characteristics. The results obtained in the first stage indicated that the fiber having the largest aspect ratio (W126) behaves anomalously, when added to the PLA matrix above a certain fiber content; the modulus and especially the strength of the composites decreased drastically. As a consequence, we investigated the behavior of this fiber more in detail. Micromechanical testing showed that the mechanism of deformation and failure changes at this critical



a)



b)

Fig. 2 *POM micrograph recorded on W126 composites; a) 20, b) 40 vol% wood. Development of a fiber network at large wood content.*

[20] de Meijer, M.; Haemers, S.; Cobben, W.; Militz, H.: *Langmuir* **16,** 9352-9359 (2000)

composition, probably due to the formation of a fiber network (Fig. 2), which was later detected by microscopic analysis. The inherent strength of this network is very small because of the weak forces acting among the fibers, therefore the structure of the composites is very sensitive to processing conditions leading to decreased reproducibility as well as reliability. Our results reveal the importance of choosing processing conditions and composition very carefully when natural fibers with large aspect ratio are used for the preparation of composites.

Ground corn cob is used as a lignocellulosic fiber in industrial practice. After the grinding of the cob, the product is separated into fractions and the heavy fraction is used as filler. However, the properties of the various fractions differ strongly from each other and separation is not perfect. Our study focused on the possible effect of inherent filler properties and imperfect separation on the deformation and failure of PLA composites containing this filler. The different strength of the components was proved by direct measurements and also by the acoustic activity of the PLA composites prepared from the various fractions. Two consecutive micromechanical deformation processes were detected in composites containing the heavy fraction, which were assigned to the fracture of the soft and hard particles, respectively. The occurrence of the two processes confirms that the separation of the components is not perfect in the industrial technology used. The fracture of soft particles, however, does not result in the failure of the composites that is initiated either by the fracture of hard particles or by matrix cracking. Very large particles debond easily from the matrix resulting in catastrophic failure at very small stresses. At sufficiently large shear stresses large soft particles break easily during compounding thus, as a consequence of fiber attrition, reinforcement depending on interfacial adhesion was practically the same in all composites irrespectively of the initial characteristics of the fiber.

A large amount of information was compiled during our studies on polymer/wood composites. Rather surprisingly, practically the same final strength

was obtained in composites produced with different matrices despite the rather dissimilar properties of these polymers. As a consequence, we decided to compare a considerable number of thermoplastic polymer/wood composites prepared with various matrices and fibers in order to analyze this phenomenon. The results confirmed that the micromechanical deformation processes initiated by the fibers determine the performance of the composites. Debonding usually leads to the decrease of strength, but decreasing strength is not always associated with poor adhesion and debonding. The direction of property change with increasing wood content depends on component properties and interfacial adhesion. Good interfacial adhesion of the components often results in the fracture of the fibers in polymer/lignocellulosic fiber composites. Depending on their size and aspect ratio, fibers may fracture parallel or perpendicular to their axis. At good adhesion, the maximum strength achieved for a particular polymer/wood pair depends on the inherent strength of the fibers, which is larger for perpendicular than parallel fracture. Inherent fiber strength effective in a composite depends also on particle size; larger particles fail at a smaller stress, because of the larger number of possible flaws in them. As Fig. 3 demonstrates, very close correlation exists between the initiation stress of the dominating local deformation process and composite strength proving that these processes lead to the failure of the composite and determine its performance.

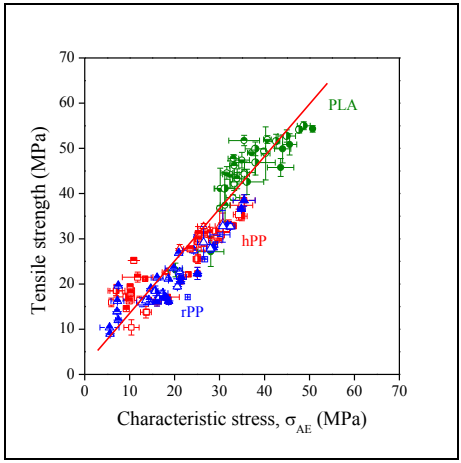


Fig. 3 *Close correlation between the initiation stress of the dominating deformation process and the strength of the composite. Symbols: PLA:* ◓◑◒○●, *rPP:* ◭◭◭△◭, *hPP:* ◓◨◒□■, *CC23:* ◓◭◒, *W35:* ◑◭◨, *W54* ◒◭◒, *W68* ○△□, *W125:* ●◭■

This conclusion led to the design of the next series of experiments aiming at the modification of the inherent strength of the fibers by impregnating wood particles with a phenol formaldehyde resin. Chemical modification of the lignocellulosic particles used as reinforcement proved to be a successful approach to improve the properties of PLA/wood composites. Treatment with a solution of 1 wt% resin resulted in a considerable increase of composite strength (see Fig. 4) and decrease of water absorption. Composite strength improved as a result of the increased inherent strength of wood, but interfacial adhesion might have been modified as well. When wood was treated with resin solutions of larger concentrations, the strength of the composites decreased, first slightly, then drastically to a very small value. The study of micromechanical deformation processes by acoustic emission measurements revealed that the mechanism of deformation changes at large resin contents. The resin forms a thick coating on the wood particles in this case, which breaks very easily leading to the catastrophic failure of the composites at very small loads. Although the approach proved to be successful and the treatment beneficial, further experiments are needed to optimize the properties of the resin, its amount and the technology of the treatment in order to achieve maximum effect.

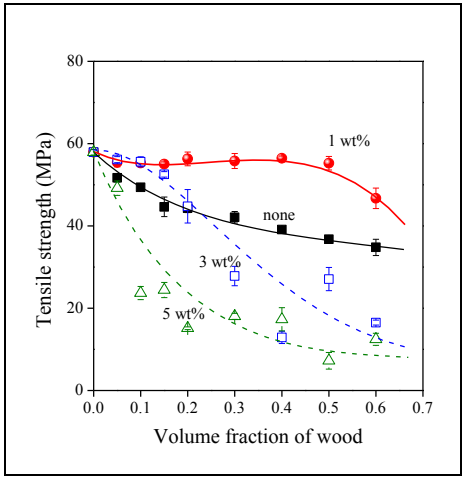


Fig. 4 *Dependence of the tensile strength of PLA/wood composites on wood content and on the concentration of the resin solution used for impregnation. Symbols:* ■ *neat,* ● *1,* □ *3,* △ *5 wt% resin solution.*

6. New scientific results

1. By detailed study and modeling we found that contrary to statements in the literature interfacial adhesion is not weak, but rather strong in PLA/wood composites. As an effect of strong adhesion the dominating micromechanical deformation process is the fracture of the fibers in these materials, although limited number of debonding also occurs. Because of strong adhesion, the effect of particle characteristics on reinforcement is weaker than in the PP/wood composites studied earlier.

2. We successfully coupled wood particles to PLA by applying two coupling agents, BMI and DBMI. However, the effect was small, because only a few very large particles debond under the effect of external load proving our earlier conclusion that in PLA/wood composites interfacial adhesion is inherently strong.

3. We proved that at large filler content long fibers with large aspect ratio may form a loose network, which is held together by weak forces. The strength of the network is small, thus network formation leads to the deterioration of stiffness and strength, and makes the structure and properties of the composites very sensitive to processing conditions.

4. In a detailed study of PLA/corn cob composites we showed that large and/or weak particles fracture during processing that changes particle size and its distribution resulting in completely different composite properties as deduced from the original size of the particles.

5. By the detailed study of the deformation and failure processes of a large number of PLA/lignocellulosic composites we proved that several micromechanical deformation processes may occur simultaneously and/or consecutively in these materials. Not all of them lead to the catastrophic failure

of the composites, but the dominating one usually determines composite properties.

6. Because of good interfacial adhesion the inherent strength of the fibers limits the maximum strength achieved in PLA/wood composites. As a result of a detailed study we pointed out that the upper limit of strength is independent of the matrix used, and depends only on particle characteristics and the direction of fracture. Small particles and fracture perpendicular to fiber axis result in large composite strength.

7. We proved that the inherent strength of the fibers can be improved by impregnation with an appropriate phenolic resin. Excessive amounts of the resin lead to the deterioration of properties thus the extent of impregnation must be optimized to achieve maximum effect.

7. Publications

7.1 The thesis is based on the following papers

1. Faludi, G.; Dora, G.; Imre, B.; Renner, K.; Móczó, J.; Pukánszky, B.: PLA/Lignocellulosic fiber composites: Particle characteristics, interfacial adhesion and failure mechanism, *J. Appl. Polym. Sci.* **131**(4), 1-41 (2014), IF: 1.64, C: 25 (DOI: 10.1002/app.39902)

2. Faludi, G.; Dora, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Improving interfacial adhesion in PLA/wood biocomposites, *Compos. Sci. Technol.* **89,** 77-82 (2013), IF: 3.57, C: 27 (http://dx.doi.org/10.1016/j.compscitech.2013.09.009)

3. Faludi, G.; Hári, J.; Renner, K.; Móczó, J.; Pukánszky, B.: Fiber association and network formation in PLA/lignocellulosic fiber composites, *Compos. Sci. Technol.* **77,** 67-73 (2013), IF: 3.57, C: 14 (http://dx.doi.org/10.1016/j.compscitech.2013.01.006)

4. Faludi, G.; Dora, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Biocomposite from polylactic acid and lignocellulosic fibers: Structure-property correlations, *Carbohydr. Polym.* **92** (2), 1767-1775 (2013), IF: 4.07, C: 26 (http://dx.doi.org/10.1016/j.carbpol.2012.11.006)

5. Faludi, G.; Link, Z.; Renner, K.; Móczó, J.; Pukánszky, B.: Factors determining the performance of thermoplastic polymer/wood composites; the limiting role of fiber fracture, *Mater. Des.* **61,** 203-210 (2014), IF: 3.50, C: 6 (http://dx.doi.org/10.1016/j.matdes.2014.04.052)

6. Csizmadia, R.; Faludi, G.; Renner, K.; Móczó, J.; Pukánszky, B.: PLA/wood biocomposites: improving composite strength by chemical treatment of the fibers, *Compos. Part A* **53,** 46-53 (2013), IF: 3.07, C: 30 (http://dx.doi.org/10.1016/j.compositesa.2013.06.003)

7.2 Other publications related to the thesis

1. Csikós, Á.; Faludi, G.; Domján, A.; Renner, K.; Móczó, J.; Pukánszky, B.: Modification of interfacial adhesion with a functionalized polymer in PLA/wood composites, *Eur. Polym. J.* **68,** 592-600 (2015), IF: 3,485, C: 50 (https://doi.org/10.1016/j.eurpolymj.2015.03.032)

2. 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, *Polym.* **190**, (2020), IF: 4.231 (https://doi.org/10.1016/j.polymer.2020.122203)

3. 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* **133**, (2020), IF: 6.444 (https://doi.org/10.1016/j.compositesa.2020.105895)

4. 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.*, Accepted Article published online on 25 August, (2020), IF: 2.574

7.3 Other publications

1. Faludi, G.; Horváth, I.; Séta, L.: Hulladék PET felhasználásával PET palack előállítás, *Műanyag és Gumiipari Évkönyv* (2007)

2. Menyhárd, A.; Faludi, G.; Varga, J.: β-crystallization tendency and structure of polypropylene grafted by maleic anhydride and its blends with isotactic polypropylene. *J. Therm. Anal. Calorim.* **93,** 937-945 (2008), IF: 1.530, C: 27 (https://doi.org/10.1007/s10973-007-8569-7)

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7.4 Conference presentations

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2. Faludi, G.; Menyhárd, A.; Varga, J.: Polipropilén keverékek és β-nukleált változatainak előállítása és vizsgálata, OTDK, April 2-4, 2007, Szeged, Hungary
3. Faludi, G.; Menyhárd, A.; Varga, J.: Polipropilén keverékek és β-nukleált változatainak előállítása és vizsgálata, X. Doktori Kémiai Iskola, May 7-9, 2007, Mátraháza, Hungary

4. Menyhárd, A.; Varga, J.; Faludi, G.: The effect of compatibilizers on the crystallization, melting and polymorphic composition of β-nucleated isotactic polypropylene and polyamide 6 blends, Eurofillers Polymerblends, August 26-30, 2007, Zalakaros, Hungary
5. Menyhárd, A.; Varga, J.; Faludi, G.: Supermolecular structure and β-nucleation tendency of polypropylene grafted by maleic anhydride, The 9th Polish Seminar to the Memory of St. Bretsznajder with Foreign Participants, September 26-28, 2007, Plock, Poland
6. Dora, G.; Faludi, G.; Pukánszky, B.: Mikromechanikai deformációs folyamatok PLA alapú biokompozitokban, MTA Tudomány Napja Konferencia sorozat Doktoranduszok Fóruma, November 4, 2010, Debrecen, Hungary
7. Faludi, G.; Dora, G.; Sudár, A.; Renner, K.; Móczó, J.; Pukánszky, B.: Modification of interfacial interactions in wood flour filled PLA composite, ECCM14 14th European Conference on Composite Materials, June 7-10, 2010, Budapest, Hungary
8. Sudár, A.; Renner, K.; Burgstaller, C.; Faludi, G.; Pukánszky, B.: Recycled PP/wood composites – adhesion and micromechanical deformations, ECCM14 14th European Conference on Composite Materials, June 7-10, 2010, Budapest, Hungary
9. Sudár, A.; Faludi G.; Pukánszky, B.: Újrafeldolgozott polipropilén/fa kompozitok szerkezete és tulajdonságai, Oláh György Doktori Iskola Doktoráns Konferencia, February 4, 2010, Budapest, Hungary
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11. Renner, K.; Faludi, G.; Móczó, J.; Pukánszky, B.: Interfacial interactions in natural fiber reinforced PLA composites, International Conference on Interfaces & Interphases in Multicomponent Materials (IIMM), September 1-3, 2010, Sheffield, United Kingdom
12. Imre, B.; Faludi, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Micromechanical deformation processes in PLA based biocomposites, 6th International Conference on Modification, Degradation and Stabilization of Polymers, September 5-9, 2010, Athens, Greece
13. Dora, G.; Faludi, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Effect of the structure of the lignocellulosic fiber on the deformation and failure mechanism of PLA composites, Eurofillers Polymerblends, August 21-25, 2011, Dresden, Germany

14. Dora, G.; Faludi, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Factors determining the properties of PLA/wood composites: particle characteristics, interactions, reinforcement, 4th International Conference Polymeric Materials in Automotive PMA 2011, April 12-14, 2011, Bratislava, Slovakia
15. Faludi, G.; Renner, K.; Móczó, J.; Pukánszky, B.: Interactions and deformation processes in PLA/wood composites, 2nd Workshop Green Chemistry and Nanotechnologies in Polymer Chemistry, April, 2011, Riga, Latvia
16. Renner, K.; Faludi, G.; Móczó, J.; Pukánszky, B.: Wood flour reinforced PLA composites: structure, micromechanical deformations, properties, Workshop, December 1, 2011, Alessandria, Italy
17. Menyhárd, A.; Dora, G.; Faludi, G.; Horváth, Zs.; Varga, J.: Kinetics of competitive crystallization of β- and α-modifications in β-nucleated iPP studied by differential scanning calorimetry, MEDICTA 2011, July 24-27, 2011, Porto, Portugal
18. Faludi, G.; Csizmadia, R.; Renner, K.; Móczó, J.; Pukánszky. B.: Development of methods for chemical modification of wood, BiPoCo2012, May 27-31, 2012, Siófok, Hungary
19. Imre, B.; Faludi, G.; Pukánszky, B.: Present status and future of sustainable plastics in Hungary, Future of Bioplastics, 3rd International Plastic Conference, October 1-2, 2013, Warsaw, Poland
20. Renner, K.; Faludi, G.; Móczó, J.; Pukánszky, B.: Poly(lactic acid)/lignocellulosic composites: functionalization and modification, BiPoCo2014, August 24-28, 2014, Visegrád, Hungary
21. Kárpáti, Z.; Kun, D.; Faludi, G.; Móczó, J.; Pukánszky, B.: Interfacial interactions in poly(lactic acid)/lignocellulosic composites, Bimate Conference, April 15-17, 2015, Slovenj Gradec, Slovenia
22. Renner, K.; Faludi, G.; Pukánszky, B.: Natural reinforcement: opportunity or necessity, DVSPM, May 11-13, 2015, Gmunden, Austria
23. Renner, K.; Faludi, G.; Móczó, J.; Pukánszky, B.: Factors determining the properties of wood composites, BYPOS 2016, March 14-18, 2016, High Tatras, Slovakia
24. Faludi G.; Kállay-Menyhárd A.; Hegyesi N.; Renner K.; Móczó J.; Pukánszky B.: Improving interfacial adhesion in PLA/wood biocomposites, BiPoCo2016, August 28-September 1, 2016, Szeged, Hungary
25. Renner, K.; Kovács, Á.; Faludi, G.; Pukánszky, B.: Modification of poly(lactic acid) with natural and synthetic fibers, BiPoCo2016, August 28-September 1, 2016, Szeged, Hungary

26. Faludi, G.; Polyák, P.; Kirschweng, B.; Pukánszky, B.: Hydrolytic degradation of poly(3-hydroxybutyrate) films: mechanism and kinetic description of the reaction, 33rd PDDG Conference, September 1-5, 2017, Taormina, Italy
27. Renner, K.; Kovács, Á.; Várdai, R.; Faludi, G.; Pukánszky, B.: Deformation processes and impact resistance in poly(lactic acid) based composites, BioPol 2017, September 11-13, 2017, Mons, Belgium
28. Pregi, E.; Horváth, E.; Faludi, G.; Pukánszky, B.: Development of hybrid PP composites containing lignin and flax, BiPoCo2018, September 2-6, 2018, Balatonfüred, Hungary
29. Kovács, Á., Várdai, R.; Faludi, G.; Renner, K.: Deformation processes and impact resistance in poly(lactic acid) based composites, BiPoCo2018, September 2-6, 2018, Balatonfüred, Hungary
30. Kovács, Á.; Várdai, R.; Faludi, G.; Pukánszky, B.; Renner, K.: Poly(lactic acid) Based Composites: Interfacial Interactions and Impact Resistance ICAMME Conference, September 26-28, 2018, Kuta, Bali, Indonesia
31. Pregi, E.; Faludi, G.; Pukánszky, B.: Development of hybrid PP composites containing lignin and flax, 8th BYPoS, March 25-28, 2018, Oščadnica, Slovakia
32. Várdai, R.; Cui, L.; Bartos, A.; Magyar, N.; Faludi, G.; Móczó, J.; Pukánszky, B.: Physical ageing of poly(lactic acid) based composites, 33rd PDDG Conference, September 1-5, 2019, San Giljan, Malta
33. Pregi, E.; Kovács, N.; Faludi, G.; Pukánszky, B.: Structure and properties of multicomponent films based on lignin, GCNPM Conference, October 9-11, 2019, Riga, Latvia