

I. NATURAL FIBER-POLYOLEFIN COMPOSITES. MINI-REVIEW

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The climate changes and fossil resource depletion have increased environmental awareness, influencing the market of products based on renewable resources. Natural fibers are renewable materials, which are certainly advantageous for sustainability. They have been extensively used as reinforcements within polymeric matrices due to their renewable origin, relatively high strength and modulus, low density and cost. On the other hand, their high moisture absorption capacity, as well as poor wettability and thermal behaviour during processing represent some impediments that limit their applications. By their numerous applications, lignocellulose polyolefin composites are present in everyday life. They combine the best properties of the neat components and can show outstanding performance. Also, the large market for these composites and their performance in terms of durability, maintenance and cost effectiveness make it possible to achieve more sustainable efficient technologies for new materials with improved properties. The paper reviews the recent advances in the development of lignocellulosic polyolefin composite materials.

Keywords: lignocellulosic fibers, polyolefins, composites

INTRODUCTION

The use of composite materials dates back to 3 000 years ago, when straw was reinforced with clay and used to build walls. After centuries, civil engineers use materials comprising natural fibers, which compete with glass fibers, and perhaps more than ever, there is an increasing demand for natural fiber-based products.

The interest in composite materials based on thermoplastic matrix reinforced by natural fibers has increased in the markets of the automotive, building and furnishing industries. One of the main disadvantages of these composites is the difficulty in disposing of the end-of-life products, as the components are well embedded, relatively stable and therefore difficult to separate and recycle. The availability of biopolymer fibers, which are relatively cheaper and occur in abundance in nature, as well as the environment policies to use “greener” technologies and the competitive pressure from the global market, have increased the interest in exploiting these materials. They must be susceptible to microbial and environmental degradation upon disposal, and have no adverse environmental impact. It should be mentioned that the growth of global population, which translated into rapidly rising raw material consumption and energy demand worldwide, calls for renewable resources of energy and consumer products.

It is expected that over the next few years, the demand for biodegradable composites will regis-

ter an explosive growth due to the environment policies that encourage the use of biodegradable polymers for compostable packaging. However, the petroleum crisis has made composites increasingly important and academic and industrial research efforts have been made to create greener and environmentally friendlier chemicals and materials for a variety of applications.

The present minireview is focused on composites prepared from polyolefins and lignocellulosic fibers. Composites are engineered materials comprising two or more constituents with different properties resulting from their components, which remain separate and distinct within the finished structure. With no claim of being exhaustive, the paper highlights some examples of research in the field of thermoplastic lignocellulosic fibers from the standpoint of practical applications. Also, aspects of structure and properties of composite materials are briefly discussed.

Manufacture of composite materials

Lignocellulosic fibers are strong, light in weight, and non-abrasive, and are used as an excellent reinforcing agent for plastics. Besides, fibers originating from biomass processing wastes (such as shell or wood flour) could be used as fillers in polymers. These fibers come from abundant and renewable resources at low cost,

which ensures a continuous fiber supply and significant material cost savings for the plastics industry. In nature, lignocelluloses are present in wood, grass, agricultural residues, forestry wastes and municipal solid wastes. That is why, lignocellulosic fibers have been the object of considerable interest in recent years, regarded as promising alternatives to replace synthetic fibers as reinforcing agents of polymeric matrix composites.

Lignocellulose consists of three types of polymers, cellulose, hemicelluloses and lignin (Table 1), which are strongly interconnected and chemically bonded by non-covalent forces and by covalent crosslinkages.¹⁻² Many microorganisms are able to degrade and use cellulose and hemicelluloses as carbon and energy sources.³⁻⁵ They synthesize two types of extracellular enzymatic systems, one represented by hydrolases and responsible for cellulose and hemicelluloses degradation, and the other represented by the oxidative and extracellular ligninolytic system, which depolymerizes lignin. The presence of lignocellulosic fibers in composite materials makes them environmentally friendly and partly degradable.⁶⁻⁷

Usually, lignocellulosic fibers possess high capillarity and hydrophobicity, which results in the increment of the composites' water absorption capacity, in relation to those reinforced with synthetic fibers. On the other hand, these fibers have good strength and stiffness, which recommend them as reinforcing materials in polymeric matrices. However, the compatibility of the thermoplastic matrix and the lignocellulosic fibers is a significant problem and various methods to improve the adhesion between composite components have been developed. The treatment of composite materials at the end of their lifetime requires less energy and lower managing costs, which reduces their environmental impact.

The main lignocellulosic fibers that can be used to obtain composite materials include bast (stem, soft, or sclerenchyma) fibers, leaf or hard fibers, seed, fruit, wood, cereal straw, and other grass fibers.

Natural fibers can be classified as follows: seed fibers (cotton, kapok); bast fibers (like jute, flax, hemp, kenaf, ramie); leaf fibers (sisal, pineapple); fruit fibers (coconut); wood fibers; grasses and reeds (wheat, oat). Unfortunately, one of the major disadvantages of plant fibers is the poor compatibility exhibited between the fibers and the polymeric matrices, which results in the low ability of the matrix to completely wet the natural fibers, thus restricting the homogenous dispersion of fibers within the matrix, and may lead to poor mechanical, dynamic mechanical, thermal and dielectric properties. The physical, mechanical, structural, thermal, environmental, as well as the cost and the health and safety characteristics of the composite should also be considered in the selection process of fibers. Also, it is well known that the composite manufacturing technique affects the typically achievable fiber volume fraction and porosity.⁸

Polyolefin is a general term used for plastics, including low density polyethylene (LDPE), linear low density polyethylene (LLDPE), high density polyethylene (HDPE) and polypropylene (PP), which are produced from fossil resources.

The manufacture of thermoplastic composites includes extrusion,⁹⁻¹² injection moulding,¹³⁻¹⁷ calendaring,¹⁸ thermoforming and compression moulding.¹⁹⁻²³ Extrusion is the most common processing method for wood-thermoplastic composites, while injection or compression moulding are used especially when a more complicated shape is needed and the total weight is lesser, compared to that produced by extrusion. Also, in these processes fiber orientation is an important parameter that affects composite properties.²⁴⁻²⁶

Table 1
The main components of some lignocellulosic sources

	Cellulose, %	Lignin, %	Hemicelluloses, %
Pine	40.0	27.7	28.5
Spruce	39.5	27.5	30.6
Eucalyptus	45.0	31.3	19.2
Birch	41.0	22.0	32.4
Grass	25.7	36.2	15.8
Wheat straw	33.0	33.0	20.0
Newspaper	45.7	30.2	22.7

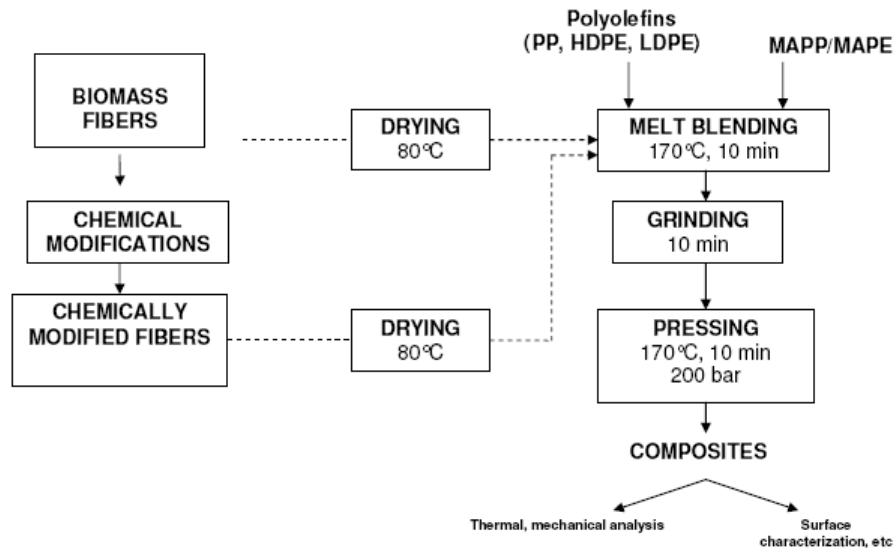


Figure 1: Structure-processing-property relationships of thermoplastic composites with natural fibers

Both compression moulding²⁷⁻²⁸ and extrusion were used to obtain PP composites reinforced with kenaf fibers. A recent study²⁹ focused on the production of PP kenaf composites *via* extrusion and evidenced that this method is most suitable for producing core materials for construction applications. Also, other manufacturing processes, such as pultrusion and, potentially, filament winding, can be used as an alternative to obtain composite materials.³⁰ Some hybrid composites comprising PP and 25% fibers, such as jute, mercerized jute and high tenacity cellulose (Cordenka), were obtained by a pultrusion process and subsequent injection moulding.³¹ Highly filled (up to 60 wt%) bleached cellulose fibers–polypropylene composites were obtained by the pelletization process and extrusion. It was found that increased fiber loading made the composites stiffer, but reduced toughness, while the impact strength of the composites decreased with increased fiber content.³² Another study revealed that PP with higher molecular weight presented stronger interfacial interaction with cellulose in the composites. Also, the composites comprising PP with higher molecular weight exhibited stronger tensile strength at the same cellulose content, by comparison with those comprising PP with lower molecular weight.³³

It has been demonstrated that the interface between natural fibers and the polymeric matrix is

a key issue for the composite properties. It should be controlled to improve the mechanical properties, the behavior to weathering and the composites recyclability.

Methods to improve compatibility

Natural fibers are hydrophilic materials and moisture absorption results in the deterioration of mechanical properties. The poor adhesion between a thermoplastic matrix and fibers also affects the final properties of the composite.

The hydroxyl groups present in the cell wall of natural fibers represent sites for water absorption. Also, they allow chemical modification in order to modify durability, dimensional stability, compatibility with the thermoplastic matrix.

It was found that graft co-polymers of the thermoplastic matrix and the addition of a polar group improved the mechanical properties of cellulose thermoplastic composites.³⁴

The modification of lignocellulosic fibers can be realized by chemical, physical, physical–chemical and mechanical methods.³⁵⁻³⁷

Chemical methods involve treatment with different chemicals in order to modify fiber surface. The selection of the method is influenced by cost, the kind of fibers and matrices and the composite processing conditions.³⁸

They can be classified into two main categories: surface compatibilization (by using a

simple coupling reaction between the surface hydroxyl groups on cellulose fibers and other agents bearing one or more –OH reactive functional groups) and co-polymerization. Silanes are efficient coupling agents extensively used in composites and adhesive formulations.³⁹⁻⁴³ Silane coupling agents may reduce the number of cellulose hydroxyl groups in the fiber-matrix interface and, as a result, the new hydrocarbon chains influence the fiber wettability, thus improving the chemical affinity to the polyolefinic matrix. The increase in the components' interaction, the change in the distribution and orientation of the fibers or reduced water sorption make fibers more hydrophobic.

Mergerization is one of the most widely used chemical methods due to the fact that it removes a significant part of biomass lignin, wax and oils, which cover the external surface of the cell wall. The mergerized fibers present improved surface tension, surface roughness and adhesion characteristics, being appropriate for reinforcing thermoplastics and thermosets.⁴⁴

Acetylation reaction results in the introduction of an acetyl functional group into lignocellulosic fibers, making them more hydrophobic. It was found that the moisture absorption content decreased with the increase of the acetyl content of fibers, due to the reduction of fiber hydrophilicity.⁴⁵ Also, a better interfacial

compatibility between acetylated wood and PP was registered.⁴⁶

Another method consists in chemical treatment with *maleic anhydride* (Figure 2), which is applied not only to modify fiber surface, but also to achieve better interfacial bonding between the fiber and the polymeric matrix and to improve the mechanical properties of the composites.⁴⁷⁻⁴⁸ Also, it was found that succinic anhydride treated jute/PP composites presented higher hardness and lower water absorption values, compared to those of the untreated ones.⁴⁹

Isocyanates are also used as a coupling agent for fiber surface modification. These chemical agents react with the hydroxyl groups of fiber constituents, forming a urethane linkage.⁵⁰⁻⁵⁴

The reaction scheme of wood with poly[methylene(poly(phenyl isocyanate))] is presented in Figure 3.

Benzoilation treatment of fibers results in the decrease of their hydrophilic nature and the improvement of interfacial adhesion, as well as strength and thermal stability,⁵⁵ while another study evidenced the decrease of the water sorption capacity of wood/PP composites.⁵⁶ Moreover, it was found that composites comprising HDPE and benzoilated poplar wood fibers presented an enhancement of tensile strength and toughness, as well as of resistance to fungal decay.⁵⁷

Other agents used to improve wettability and interfacial adhesion characteristics are fatty acid derivatives, such as oleoyl chloride.⁵⁸⁻⁵⁹

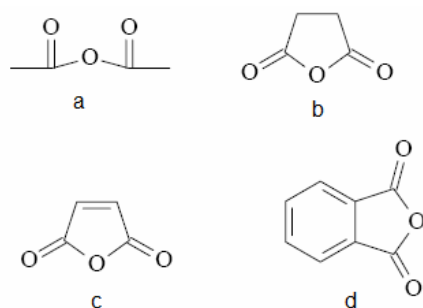


Figure 2: Chemical structure of most frequently used anhydrides to improve fiber compatibility: a – acetic anhydride; b – succinic anhydride; c – maleic anhydride; d – phthalic anhydride

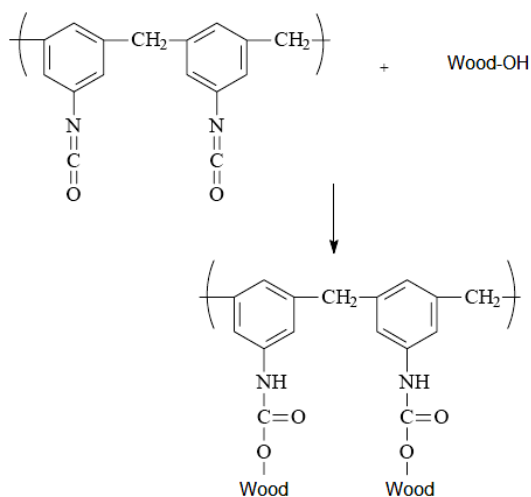


Figure 3: Reaction of wood with poly(methylene(poly(phenyl isocyanate)))

Co-polymerization can be performed by the following methods: the use of planar stiff molecules; grafting with polymerizable molecules; direct activation of the surface to generate an active centre; and reaction with organometallics.⁶⁰⁻⁶² Usually, hydrophobization treatments of fibers reduce moisture sorption, which is very important for composite materials exposed to environmental conditions.⁶³⁻⁶⁴

Many authors used maleic anhydride grafted polypropylene/polyethylene as a coupling agent or compatibilizer to improve the chemical compatibility of the fillers and the matrix⁶⁵⁻⁷⁰ and, consequently the mechanical properties of the composite materials.

Other studies reported that *in situ* grafting of PP/PE blends with maleic anhydride through the extruder resulted in the improvement of the interfacial bonding, as evidenced by the increased mechanical strength.⁷¹⁻⁷² The treatment of natural fibers with MAPP copolymer provides covalent bonds across the interface, which increases fiber wettability and interfacial adhesion between them and the polymeric matrix.

Physical methods involve treatment by plasma,⁷³⁻⁷⁹ corona, laser or γ -ray.⁸⁰⁻⁸² Usually, these treatments improve surface polarity and therefore, the wettability and hydrophilic character of natural fibers are modified. The physical methods are cleaner and simpler than chemical methods and their effects on the composite properties are largely dependent on the treatment durations.

Also, the steam explosion process has been efficient to improving adhesion with polymeric matrices.⁸³ Lignocellulosic fibers are rapidly heated by high-pressure steam (without addition of any chemicals) for a short time and then the product is explosively discharged to atmospheric pressure, which results in a sudden decomposition in order to remove hemicelluloses.⁸⁴⁻⁸⁵ Also, it was found that enzymes offer an inexpensive and environmentally attractive option to improve the surfaces of natural fibers for composite applications.⁸⁶⁻⁸⁹ Enzyme modified abaca fibers (30%) reinforced PP composites presented lower water adsorption properties, as compared to composites comprising unmodified fibers. Also, an increase of tensile strength and flexural strength was evidenced due to fiber modification.⁹⁰ Also, an improvement of the interfacial adhesion and shear stress ability occurs

in the composite comprising PP and 30% enzyme modified palm fibers.⁹¹

Vibratory ball milling and compression milling, which make material handling easier through composite processing, represent mechanical methods that could improve the compatibility between thermoplastic and lignocellulosic fibers.⁹²

Physical-chemical methods involve removing some soluble components of the fibers.⁹³ Due to the great diversity and variability of natural fibers, they exhibit considerable variation in diameter and the length of individual filaments.

Jute, flax and hemp are the most common raw materials in textile markets, but in recent years they have been widely used in the composites area. In terms of specific strength, natural fibers can be compared with well-known glass fibers. In spite of the fact that high tensile strength is attributed to the high cellulose content, it is difficult to correlate fiber strength with the cellulose content. Also, the presence of hemicelluloses and lignin influences the mechanical strength, while the presence of waxes improves fiber wettability and the adhesion between fibers and thermoplastic polymers.⁹⁴

Some studies evidenced that PP-hemp composites⁹⁵⁻⁹⁹ present good properties, which recommend them for numerous applications, including in the automotive and building sectors, where their applications are limited or poorly managed by different modifications of the hemp fibers. Silane and NaOH treatment of hemp improved the thermal stability of hemp-HDPE composites.¹⁰⁰ Flax fibers possess excellent specific mechanical properties and their addition into a thermoplastic matrix increases the mechanical properties of the polymer.¹⁰¹⁻¹⁰²

Due to its rapid growth, bamboo has an important market value for many countries. Bamboo PP composites have high flexural properties, which makes them suitable for replacing glass fiber, which is most commonly used in the automotive industry nowadays.¹⁰³⁻¹⁰⁵

The chemically modified bagasse fibers have a reinforcing effect for a PP matrix, which was confirmed by the increase of the tensile, flexural and impact strengths, as compared to neat PP.¹⁰⁶ An improvement of mechanical properties was reported for composites comprising recycled polyethylene.¹⁰⁷

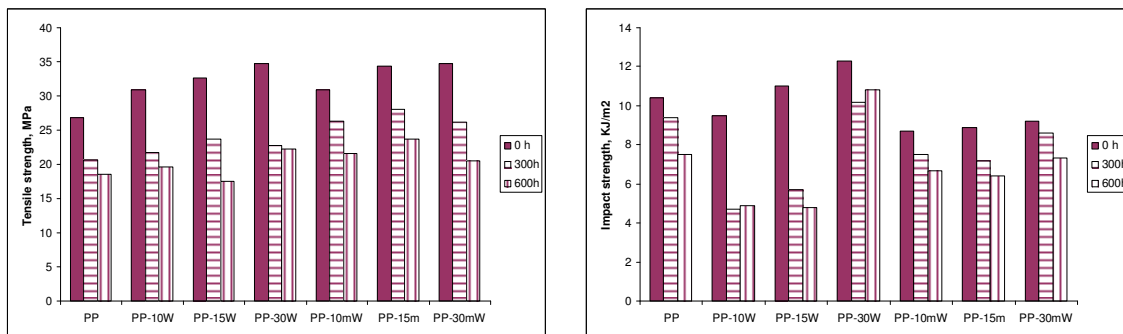


Figure 4: Influence of weathering time on mechanical properties of wood-PP composites

Other agricultural residues, such as stalks of most cereal crops, rice husks, coconut fibers (coir), maize cobs, peanut shells and other agro wastes, are used as fillers for thermoplastic matrices.¹⁰⁸ The addition of natural fiber from hemp bast, flax bast, chemically pulped wood, wood chips, wheat straw, and mechanically pulped triticale to an LDPE matrix improved the stiffness with a corresponding loss of material elongation and impact toughness.¹⁰⁹

The potential of sisal¹¹⁰⁻¹¹¹ or kenaf bast fibers as a reinforcing fibers in thermoplastic composites has been evidenced, due to its superior toughness and high aspect ratio in comparison to other fibers.¹¹²⁻¹¹³ The behavior of composites comprising thermoplastics as matrix and jute,¹¹⁴⁻¹¹⁵ bamboo,¹¹⁶ flax,¹¹⁷ oil palm fibers,¹¹⁸ sisal,¹¹⁹ rice,¹²⁰⁻¹²¹ as well as wood¹²²⁻¹²⁸ has been investigated extensively. Some photo-oxidation processes initiated by UV irradiation in the presence of oxygen and moisture determine the scission of the polymer chain, while others trigger intermolecular crosslinking. The crosslinking and chain scission processes lead to the decrease of mechanical properties. The chemical modification of eucalyptus wood with TDI⁵¹ has positively influenced the mechanical properties of composites comprising wood (5, 10 and 15% respectively) and PP (Figure 4). Also, a higher water absorption capacity after weathering was evidenced.

Fiber source, as well as fiber geometry, influences the density of the composite materials and their end-use applications.¹²⁹

Wood is the most frequently used material for composite applications. The most common thermoplastics are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS), due to the processing temperature (about 200 °C) and to the degradation temperature of lignocellulosic fibers.¹³⁰⁻¹³¹ An

increase of the Young modulus was evidenced with the addition of wood flour to a PP matrix, whereas tensile strength, strain at break and fracture toughness were decreased as fiber content increased. MAPP had a positive effect on the tensile strength and ductility and had no significant effect on fracture toughness.¹³²⁻¹³³

The stiffness and hardness of palm wood/LDPE composites significantly increased with the increase of wood content. The Young modulus of the composite filled with 70 wt% of wood fibers was approximately 13 times greater than that of the LDPE, while the water absorption capacity increased with the filler content.¹³⁴ For composites comprising oak wood (10, 20 and 30%) and LDPE, it was found that the wood loading levels made LDPE behave as a pseudoplastic material.¹³⁵

It is well known that the properties of composites comprising a thermoplastic matrix and wood fibers are strongly influenced by wood composition. Thus, the properties and amount of each lignocellulosic component influence the fiber properties, as well as the composite materials.¹³⁶⁻¹⁴¹ Hemicelluloses removal reduced the water absorption and thickness swelling of the composites,¹⁴² while the removal of lignin resulted in the highest tensile modulus and storage modulus at ambient temperature, but on the other hand, in the decrease of water resistance, thermal stability, and storage modulus of composites at high temperatures. In addition, the removal of both hemicelluloses and lignin resulted in the highest tensile strength, elongation at break, toughness, and impact strength of the composites.

The main component of natural fibers is cellulose. Its hydrophilic character causes the absorption of water in the fiber. This forms a major drawback for the natural fibers, since the interfacial bonding with a thermoplastic matrix weakens, resulting in poor physical and

mechanical properties of the composite materials. The matrix modifications, fiber surface treatments or addition of a compatibilizing agent¹⁴³ contribute to improving the dispersion, adhesion and compatibility between the hydrophilic cellulose and hydrophobic matrix. Different spun cellulose fibers have been used to reinforce thermoplastic polymers, such as polypropylene (PP), polyethylene (PE) and (high impact) polystyrene (HIPS), for injection moulding applications. It was found that cellulose fibers coupled to the above mentioned matrix by adding small amounts of maleic acid anhydride grafted or copolymerized matrix material determined a significant improvement of the mechanical properties, such as strength, stiffness and impact strength.¹⁴⁴ Maleic anhydride moieties from maleic acid anhydride grafted matrix effectively interacted with the free OH group present in the fibers. When cellulose fibers of bagasse were used as reinforcing agent for an LDPE matrix, a significant increase of the mechanical properties of the composites was registered with the addition of cellulose fibers.¹⁴⁵

Composites characterization

The techniques used for characterizing composite materials are strongly dependent on the applications of the end-products. Thus, their manufacture using polyolefin-natural fiber composites requires their melting and flow through the processing equipment, as well as respecting the processing parameters. That is why, knowledge about the melt flow behavior of materials at shear rates encountered in the most relevant polymer processing operations is necessary.

Rheological tests performed in various steady state and dynamic environments are used to determine the sensitivity of a material during processing. Fiber composition has an important effect on melt viscosity and resulting extrusion processing.¹⁴⁶ A recent study on kenaf-HDPE composites¹⁴⁷ evidenced that the products compounded at high processing temperature provided the best performance of rheological, thermo-mechanical and tensile properties, in comparison with the composites compounded at low processing temperature.

The fire resistance of the composite materials is evaluated by **thermogravimetric analysis**. It is well known that polyolefins burn and drip when in contact with fire, thus resulting an integrity loss. The addition of natural fibers leads to the

improvement of thermal properties and integrity of composites.¹⁴⁸⁻¹⁴⁹ On the other hand, the relatively low thermal stability of less common fibers, such as curaua, henequen, fique, buriti, olive husk, and kapok fibers, could be a limitation to their composites.¹⁵⁰

DSC analysis is used to determine the crystallinity of the polymeric matrix in the composite materials.¹¹⁶

The **mechanical properties** are very important for composite applications. Thus, the tensile test evidences the property through the thickness, while the flexural test is influenced especially by the properties of the specimen closest to the top and bottom surfaces. Numerous studies evidenced that the tensile modulus of composites comprising modified fibers are higher, as compared to those of composites with untreated fibers,¹⁵¹⁻¹⁵² which suggests a stress transfer from the matrix to the filler, indicating a better interfacial bonding with a consequent improvement in the mechanical properties of the composites comprising modified fibers.

Flexural stiffness is a function based both on the elastic modulus (stress per unit strain) of the material and the moment of inertia (which is a function of the cross-sectional geometry). The flexural strength of LDPE blended with 70% palm wood as filler powder (DPW) was twice greater than that of the neat LDPE.¹⁵³ The addition of MAPP to natural fiber PP composites improved composite flexural strengths and flexural moduli.¹⁵⁴⁻¹⁵⁷

The impact property is important in engineering applications. With instrumented impact testing, load-displacement curves can be obtained.¹⁵⁸ The impact strength of the cellulose PP composites was significantly lower than that of the neat polypropylene matrix, due to the fact that cellulose fibers act as stress concentrators in the polymer matrix, thus reducing the crack initiation energy, and consequently the impact strength of the composites.³²

Water absorption

The water transport in composite materials could be due to diffusion inside the matrix, imperfections within the matrix or to capillarity along the fiber matrix interface. It was found that LDPE composites containing different lignocellulose content exhibited different water capacity. Thus, a high content of lignocellulose resulted in higher water capacity, due to the increased number of micro voids caused by the larger amount of poorly bonded area between the

hydrophilic filler and the hydrophobic matrix polymer. Besides, water absorption of the lignocellulosic/LDPE composites was higher than that of the lignocellulosic PP composites, which was attributed to the weak interfacial adhesion between the PE chains and the lignocellulosic filler.¹⁵⁹⁻¹⁶⁰

The thickness swelling and water absorption of the composites slightly increased as the filler loading increased, but to a negligible extent, as compared with the wood-based composites (particleboard and fiberboard) and the solid woods (red pine and birch). The mechanical properties of the composites decreased as the filler loading increased, but the composites had an acceptable strength level. It was concluded that these materials are suitable to be used for the interior of bathrooms, wood decks, food packaging, etc.¹⁶¹

Scanning electron microscopy is used to evidence the morphology of composite materials. The images show the interface adhesion between the fiber and the matrix, which allows stress transfer from the matrix to the fiber and accounts for the superior tensile and flexural moduli of the composites. Also, the presence of phase adherence between the fibers and polymeric matrix prevents the propagation of the cracks generated during the impact tests.¹⁶²⁻¹⁶³

Natural fibers exhibit high electrical resistance and it is expected that when these fibers are incorporated in low modulus polymer matrices, they would yield materials with better properties suitable for various applications.

Natural fiber reinforced plastic composites not only act as insulators, but also present good mechanical support for field carrying conductors, serving as terminals, connectors, industrial and household plugs, switches, printed circuit boards, panels. Thus, it was found that the *dielectric constant*, *loss factor* and *conductivity* of polyolefin-natural fiber composites increased with increasing the fiber content, due to the rise in polar groups. Also, the reduction in the hydrophilic nature of jute yarns brought about by chemical treatments resulted in a loss of the dielectric constants and conductivities of the treated composites, as compared to the untreated ones, as a result of reduction in orientational polarisation and moisture absorbance.¹⁶⁴⁻¹⁶⁵

Applications

Presently, it is difficult to imagine our life without plastics. These commodities have

conquered and hold a significant position in various fields over other materials, such as wood, metals, glass, ceramics, due to their low manufacturing costs, excellent mechanical strength, low density, small weight, and thermal and chemical properties. The composite materials are used for wide-ranging applications, in construction, interior finishes, automotives, panels for both indoor and outdoor uses and garden products.¹⁶⁶⁻¹⁶⁹ Particularly, thermoplastic natural fiber composites are used as door and window frames, decking material, railings for the parapet wall systems, furniture sections, and it is expected that their applications will expand significantly in the near future. There is evidence that the economic use of natural fiber composites constitutes a great opportunity for social and economical progress in developing countries.¹⁷⁰ These materials could offer an answer to maintaining the sustainable development of economical and ecological technologies. On the other hand, the demand for lighter and stronger components is increasing in many sectors.

Through life cycle assessment or analysis (LCA), quantitative values of a product's impact on the environment are obtained. The method is based on the calculation of the impacts from the manufacturing, use and scrapping and recycling processes of composite materials. An analysis of the environmental performance for the life cycle of sugarcane bagasse-PP, as compared to talc-PP composites, demonstrated that natural fibers represent an environmentally superior option in automotive applications where weight is particularly relevant.¹⁷¹ As new chemical and physical treatments are efficient in fiber modifications at improving their compatibility with the thermoplastic matrix, it is important to perform the life cycle assessment of these materials in order to identify new applications and enter markets that are unexplored as of yet. A recent LCA study¹⁷² evidenced that hemp mats in glass-fiber reinforced thermosets are more eco-efficient than the conventional glass-fiber alternative, due to the reduction of glass-fiber and resin content, as compared to standard glass-fiber/composites and to the 'green' element represented by the hemp mat. The entire life cycle of the wood-fiber-reinforced PP composite possesses a better environmental standing than that of PP.¹⁷³ Another study revealed that the incorporation of biodegradable waste (rice husks, cotton linter) into composite materials prevents the consumption of natural resources and solves

the problem of waste disposal, and although they cannot be easily recycled, the use of a biodegradable matrix would allow for the valorization of the composite waste as compost.¹⁷⁴ Hence, further studies are needed to unambiguously provide information on the ecological performance of natural fiber thermoplastic composites, as compared to other materials.

The biodegradability of composite materials is a problem, especially when dealing with structural parts of exterior panels for future vehicles.¹⁷⁵⁻¹⁷⁷ There are many aspects to be considered, such as reproducibility of their properties and their long life cycle when used for exterior body parts. The scientific community is still searching for solutions to identify some areas where production performance may be enhanced, according to environmental directives. Also, the identification of new fiber sources for efficient reinforcing agents for thermoplastic matrices to obtain composite materials by economically and environmentally sound technologies represents a challenge for the future.

It is clear that a holistic approach with life cycle assessment will allow increasing the role of polyolefin-natural fibers composites in the future.

CONCLUSION

The need for innovative, sustainable and recyclable materials explains why both natural and synthetic polymers still provide numerous challenges for researchers. Research on polymer composites comprising natural fibers is a significant topic due to the dwindling petroleum resources, low cost of lignocellulosic fibers and impact of human activities on the environment. Also, climate change and resource depletion have increased public environmental awareness and will influence the market of sustainable products based on renewable resources.

This mini review evidences that polyolefin-natural fiber composites have applications in the automotive industry and in some areas such as aircraft components, building industry and rural areas. Also, it is worth mentioning the interest of the market in the development of materials based on lignocellulosic materials that target the biomedical field.

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