

# RECENT DEVELOPMENT IN NATURAL FIBER REINFORCED POLYPROPYLENE COMPOSITES

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**Abstract-** This review article describes the recent developments of natural fiber reinforced polypropylene (PP) composites. Natural fibers are low-cost, recyclable, and eco-friendly materials. Due to eco-friendly and bio-degradability characteristics of these natural fibers, they are considered as strong candidates to replace the conventional glass and carbon fibers. The chemical, mechanical, and physical properties of natural fibers have distinct properties; depending upon the cellulosic content of the fibers which varies from fiber to fiber. The mechanical properties of composites are influenced mainly by the adhesion between matrix and fibers. Chemical and physical modification methods were incorporated to improve the fiber–matrix adhesion resulting in the enhancement of mechanical properties of the composites. The most important natural fibers are jute, flax, and coir and their novel processing technics to develop natural fiber reinforced composites are also described.

**KEYWORDS:** *natural fibers, polypropylene, chemical and physical modifications, processing, properties.*

## I. INTRODUCTION

In past considerable research and development have been expanded in natural fibers as a reinforcement in thermoplastic resin matrix. These reinforced plastics serve as an inexpensive, biodegradable, renewable, and nontoxic alternative to glass or carbon fibers. The various advantages of natural fibers over man-made glass and carbon fibers are low cost, low density, competitive specific mechanical properties, reduced energy consumption and biodegradability. Thermoplastic materials that currently dominate as matrices for natural fibers are polypropylene (PP), polyethylene, and poly(vinyl chloride) while thermosets, such as phenolics and polyesters, are common matrices. With a view to replacing the wooden fittings, fixtures and furniture, organic matrix resin reinforced with natural fibers such as jute, kenaf, sisal, coir, straw, hemp, banana, pineapple, rice husk, bamboo, etc., have been explored in the past two decades.

There is an increasing demand from automotive companies for materials with sound abatement capability as well as reduced weight for fuel efficiency. Natural fibers possess excellent sound absorbing efficiency and are more shatter resistant and have better energy management characteristics than glass fiber reinforced composites. In automotive parts, such composites not only reduce the mass of the component but also lower the energy needed for production by 80%. Eco-friendly composites can be made by replacing glass fibers with various types of ligno-cellulose fibers. However, such composites have a distinct disadvantage of load-bearing capability compared to glass fiber reinforced thermoplastics. The variation in the properties of natural fibers is another important aspect that has to be considered. Demands for natural fibers in plastic composites is forecast to grow at 15–20% annually with a growth rate of 15–20% in automotive applications, and 50% or more in selected building applications. Other emerging markets are industrial and consumer applications such as tiles, flower pots, furniture, and marine piers [1].

Development of new composite products from the easily renewable natural materials has a strong potential to deliver novel biodegradable and/or readily recyclable materials suitable for the automotive and packaging industry, thereby replacing not so easily renewable fossil fuel-based polymers/plastics.

Complete matrix fusion to facilitate thorough fiber impregnation, formation of strong fiber/matrix interfacial bonding, and matrix-to-fiber stress transfer efficiency are vital requirements for the manufacture of reliable, eco-friendly natural composites that can possess better mechanical properties and withstand environmental attacks. Bledzki and Gassan [2] have stated that the quality of the fiber–matrix interface is significant for the application of natural fibers as reinforcement fibers for plastics. Physical

and chemical methods can be used to optimize this interface. These modification methods are of different efficiency for the adhesion between matrix and fiber.

However, the main problem of natural fiber/polymer composites is the incompatibility between the hydrophilic natural fibers and the hydrophobic thermoplastic matrices. It necessitates the use of compatibilizers or coupling agents in order to improve the adhesion between fiber and matrix [3].

Matrix or fiber modification is therefore necessary to improve the compatibility between fiber and matrix. Maleated polyolefins are used to modify the matrix. Such modification of the matrix develops the interactions between the anhydride groups of maleated coupling agents and the hydroxyl groups of natural fibers. For the purpose of making engineering parts with a wide study on the effect of different coupling agents, such as silanes, maleic anhydride grafted polypropylene (PPgMA) or modification by acetylation, has been reported in the literature.

In the present article an attempt has been made to review the state of the art of organocellulosic fiber reinforced polyolefin composites. The various types of natural fibers used for such reinforcement are described. The modifications of fiber and matrix are discussed and the mechanical properties of composites are described also. Performance of the composites is summarized.

## II. LIGNOCELLULOSIC FIBERS OR MATERIALS

Several types of lignocellulosic materials have been investigated in PP [4–12]. Cellulosic fibers from other sources have been reported in polyolefin composites by several authors. A comprehension review of the cellulosic-based materials in composites has appeared recently [2]. The lignocellulosic fibers that have been used in polyolefins include cellulose fiber [4–12], wood fiber [13–22], flax [23–29], *Cannatis sativa* (hemp) [30–35], jute [1,25,33,35–37], pinewood fiber [38–41], sisal [3,33,42–44], rice husk [45–48], saw dust [48–50], lugffa sponge fiber [3,51], wheat straw [52], paper sludge [53], coconut fiber [33,54], kenaf [55,56], kapok/cotton [57], pineapple leaf [58], basalt [59], vetiver [60], bio flour [61], bamboo fiber [62], and date palm fiber [63].

### 2.1 CHEMICAL COMPOSITION OF SOME NATURAL FIBERS

Cotton has the maximum content of cellulose ranging from 85 to 95%. The other fibers have cellulose along with lignin and other components such as hemicellulose, pectin wax, etc., in varying quantities. The various chemical constituents of a specific natural fiber also vary considerably.

## III. MECHANICAL PROPERTIES OF LIGNOCELLULOSIC MATERIALS

The structure, chemical composition, microfibrillar angle, and cell dimension defects are the most important variables that affect the overall properties of the fibers. A fiber is more ductile if the microfibrils have a spiral orientation to the fiber axis. If the microfibrils are oriented parallel to fiber axis then the fibers are inflexible and rigid. The natural fibers exhibit considerable variation in diameter along with the length of individual filaments. In Table 2 the properties of these fibers are summarized.

The properties are affected by the chemical constituents and complex chemical structure of natural fibers. The angle between the axis and the fibril of the fiber (microfibrillar or spiral angle) also affect the strength of the fibers. Mechanical properties are higher if this is smaller. Least tensile strength is shown by coir fibers which may be attributed to low cellulose content and considerably high microfibrillar (41–458 for coir, 208 for sisal, 148 for PALF, and 108 for flax while other fibers such as jute, ramie, and hemp, have values 510). The lignin content of the fibers influence its structure properties and morphology. The waxy substances of the natural fibers affect the fiber wettability and adhesion characteristics. In specific strength the natural fibers can be compared with glass fibers.

### **3.1 SURFACE MODIFICATIONS OF LIGNOCELLULOSIC FIBERS**

#### **3.1.1. Physical Methods**

Reinforcing fibers can be modified by physical and chemical methods. Physical methods, such as stretching, calendering, thermotreatment, and the production of hybrid yarns do not change the chemical composition of the fibers. Physical treatments change structural and surface properties of the fiber and thereby influence the mechanical bondings to polymers.

#### **3.1.2. Chemical Methods**

Strongly polarized cellulose fibers are inherently incompatible with hydrophobic polymers when two materials are incompatible; it is often possible to bring about compatibility by introducing a third material that has properties intermediate between those of the other two. There are several mechanisms of coupling in materials: weak boundary layers – coupling agents eliminate weak boundary layers, . deformable layers – coupling agents produce a tough, flexible layer, restrained layers – coupling agents develop a highly crosslinked interphase region, with a modulus intermediate between that of substrate and of the polymer, wettability – coupling agents improve the wetting between polymer and substrate (critical surface tension factor), chemical bonding – coupling agents form covalent bonds with both materials, and . acid–base effect – coupling agents alter acidity of substrate surface.

The development of a definitive theory for the mechanism of bonding by coupling agents in composites is a complex problem. The main chemical bonding theory alone is not sufficient. So the consideration of other concepts appears to be necessary, which include the morphology of the interphase, the acid–base reactions at the interface, surface energy, and the wetting phenomena.

#### **3.1.3. Change of Surface Tension**

The surface energy of fibers is closely related to the hydrophilicity of the fiber. Some investigations are concerned with methods to decrease hydrophilicity. The modification of wood–cellulose fibers with stearic acid hydrophobizes those fibers and improves their dispersion in PP. A treatment with poly(vinyl acetate) increases the mechanical properties and moisture repellence.

#### **3.1.4. Impregnation of Fibers**

A better combination of fiber and polymer is achieved by impregnation of the reinforcing fabrics with polymer matrices compatible to the polymer. For this purpose polymer solutions or dispersions of low viscosity are used.

#### **3.1.5. Graft Copolymerization of Matrix**

An effective method of chemical modification of matrix and natural fibers is graft copolymerization. This reaction is initiated by free radicals of the cellulose molecule. The cellulose is treated with an aqueous solution with selected ions and is exposed to a high energy radiation. Then the cellulose molecule cracks and radicals are formed. Afterward the radical sites of the cellulose are treated with a suitable solution (compatible with the polymer matrix), for example vinyl monomer (i.e., acrylonitrile, methyl methacrylate, polystyrene, etc.). The resulting co-polymer possesses properties characteristic of both fibrous cellulose and grafted polymer.

For example, the treatment of cellulose fibers with hot polypropylene–maleic anhydride (MAH–PP) copolymers [3] provides covalent bonds across the interface. The mechanism of reaction can be divided into two steps: activation of the copolymer by heating ( $t \frac{1}{4} 170^{\circ}\text{C}$ ) (before fiber treatment), and esterification of cellulose.

After this treatment the surface energy of the fibers is increased to a level much closer to the surface energy of the matrix. Thus, a better wettability and a higher interfacial

#### IV. PROCESSING AND PROPERTIES OF NATURAL FIBER REINFORCED PP COMPOSITES

Khondker et al. [1] have reported that the jute/PP unidirectional composites, specimens with only 20% of jute fiber ( $V_f$ ) content, show remarkable improvement in tensile and bending properties when compared to those of the virgin PP specimens. The improvements in the mechanical properties are broadly related to various factors, such as the wettability of resin melts into fiber bundles, interfacial adhesion, orientation, and uniform distribution of matrix-fibers and the lack of fiber attrition and attenuation during tubular braiding process.

The water absorption properties of coir, sisal fibers reinforced PP composites in water at three different temperatures, 23, 50, and 70°C, were studied. A decrease in tensile properties of the composites was demonstrated, showing a great loss in mechanical properties of the water-saturated samples compared to the dry samples [3].

The hardness and elastic modulus of a cellulose fiber-reinforced PP composite were investigated by nano-indentation with a continuous stiffness technic. A line of indents was produced from the fiber to the matrix. There was a gradient of hardness and modulus across the interphase region. The distinct properties of the transition zone were revealed by 1–4 indents, depending on nano-indentation depth and spacing [4].

The high-tenacity man-made cellulose filament yarn (rayon tyre cord yarn) reinforced PP, polyethylene, high impact polystyrene (HIPS), and poly(lactic acid) (PLA) composites. The pultrusion compounding method developed for highly homogeneous composites. For a fiber load of 30 wt%, typical values for tensile strength, modulus, Charpy unnotched, and notched impact strength are 80 MPa, 3.5 GPa, 85 kJ/m<sup>2</sup>, and 12 kJ/m<sup>2</sup>, respectively. A high impact resistance level is maintained also at low temperatures where the matrix material becomes brittle. For the other matrix materials, similar reinforcing effects are observed, except for the impact behavior of HIPS, where the reinforcing fibers interfere with the impact modification of the matrix polymer. In contrast, the impact characteristics of PLA are drastically improved increasing the unnotched and notched Charpy strengths by 380% and 200%, respectively [5].

Amash and Zugenmaier [6] investigated the thermal, morphological and dynamic mechanical properties of PP–cellulose fiber (CF) composites. The effects of drawing on the structure and physical properties of PP–CF composites were also studied. By increasing draw ratio, the melting peak of PP shifted to higher temperatures suggesting a constrained melting, and the uniaxial elastic modulus was considerably enhanced. The biggest influence was observed for the samples of PP–spun cellulose and the lowest for neat PP. In addition to the fibrillar structure of the oriented PP, the highly CF orientation and the efficient compatibilization in composites are responsible for the effects observed in the drawn samples.

The nanocomposite films of isotactic PP reinforced with cellulose whiskers highly dispersed with surfactant were prepared for the first time and compared with either bare or grafted aggregated whiskers. The nanocomposites obtained with the surfactant-modified whiskers exhibited enhanced ultimate properties when compared to the neat matrix or to the composites containing the other filler types [7].

The tensile and Izod impact strength properties of rice-husk flour and wood flour as the reinforcing filler and different compatibilizing agents, by assessing their mechanical properties and the morphological characteristics of their fracture surfaces. The tensile properties of the composites made with the twin-screw extruding system were better than those of the composites made with the single-screw extruding system, due to the improved dispersion of the filler. The tensile strength and modulus of the lignocellulosic filler–PP composite made with the twin-screw extruding system were improved in the case of the composite made without any compatibilizing agent and significantly improved in the case of the composite made with the compatibilizing agent, as compared with those made with the single-screw extruding system. The Izod impact strengths of the composites made with the two different extruding systems were almost the same, the degree of dispersion of the fillers might influence the notched impact

performance, but the similar impact strength of both samples with different extruding processes might be due to the fact that impact test is not discriminating enough to reveal the difference in dispersion status of the present composites [8].

The mechanical properties of the CFs reinforced polyethylene composites increased with increasing the average fiber length and the composite materials prepared using both matrices and cellulose fibers treated with g-methacryloxypropyltrimethoxy silane (MPS), and g-mercaptoproyltrimethoxy silanes (MRPS) displayed good mechanical performances. On the other hand, with hexadecyltrimethoxy-silanes (HDS) bearing merely aliphatic chain only a modest enhancement on composite properties was observed [9].

Qiu et al. [10] have reported that PP with higher molecular weight revealed stronger interfacial interaction with cellulose fibers in the composites, compared with the lower molecular weight PP; the composites derived from higher molecular weight of PP exhibited stronger tensile strength at the same cellulose content.

Georgopoulos et al. [11] have investigated that the loading of LDPE with natural fibers leads to a decrease in tensile strength of the pure polymer. On the other hand, Young's modulus increased due to the higher stiffness of the fibers. Although the properties of some blends are acceptable for some applications, further improvement is necessary, by optimizing fiber-polymer interface characteristics.

Joly et al. [12] have optimized the fiber treatments for PP/cellulosic-fiber composites. In one method the grafting of a PP chain by an ester bond capable of co-crystallizing or entangling with the fibers was used. While the modification of cellulose fiber was done by isocyanates for improvement in the rupture, initiation strength was observed by PP modification of fibers resulting in the crack propagation strength.

PP The tensile strength of the PP-wood-based composites decreased significantly with increasing wood fiber content and no significant change in modulus of elasticity was found for any weight fraction of wood fiber. Fiber pullout was observed on most of the composite fracture surfaces examined using SEM. These results indicate a lack of adhesion between PP and wood fiber [13].

Tensile and flexural tests of foamed composites were investigated as a dependence of function of density (specific properties) of foamed specimens and compared with nonfoamed composites based on MAH-PP has improved the physico-mechanical properties up to 80%. Chemical foaming agents have also an effect on surface roughness of the composites which decreased surface roughness of the foamed composites compared to the nonfoamed composites. Water absorption and thickness swelling of the composites

were also investigated. The density of microfoamed hard wood fiber-PP composites reduced around 30% and decreased up to  $0.741 \text{ g/cm}^3$  by using an exothermic chemical foaming agent. Optical microscopy showed that the cells are round and cell sizes are affected by chemical foaming agents. Tensile and flexural tests are performed on the foamed composites to investigate the dependence of these properties on the density (specific properties) of foamed specimens and compared with nonfoamed composites and MAH-PP has improved the physico-mechanical properties up to 80% [14].

The impact fracture behavior of PP/wood fiber composites modified with maleated PP as compatibilizer and poly(butadiene styrene) rubber as impact modifier has been studied by a Charpy impact testing method. The neat PP and unmodified PP/wood fiber composite exhibit brittle fracture and nearly elastic behavior while the impact modifier and compatibilizer cause elastic to elastic-plastic transition dominated by unstable crack propagation. Both the impact modifier and the compatibilizer toughen the PP matrix enhance the total fracture energy of the modified composites. While the crack initiation energy is mostly only a little material dependent and reflects the matrix behavior, the crack propagation energy is much more influenced by the morphology [15].

Novel compatibilizer (m-TMI-g-PP) with isocyanate functional group was synthesized by grafting m-isopropenyl-a,a-dimethylbenzyl-isocyanate (m-TMI) onto isotactic PP in a twin-screw extruder. The effect of filler concentration on the mechanical properties of bleached kraft pulp of eucalyptus reinforced

PP composites, prepared by using m-TMI-g-PP as the compatibilizer, was investigated. The addition of the compatibilizer resulted in greater reinforcement of composites, as indicated by the improvement in mechanical properties. Tensile strength of composites so prepared increased by almost 45%, whereas 85% increase in flexural properties was observed [16].

Costa et al. [18] have evaluated the tensile and flexural performance of PP–wood fiber composites. The effect of these variables on tensile strength, Young's modulus, elongation at yield and flexural strength was determined through a  $2^2 3^1$  factorial design (where the variables chosen were: fiber content (F ), percentage of MAPP coating on the wood fiber (C ) and type of matrix (M )). The mechanical properties (responses) selected from tensile data were stress at maximum load (TS), tensile modulus (TM) and elongation at yield (E ). The selected responses of flexural data were stress at maximum load (FS) and flexural modulus (FM). Using standard least-squares parameter estimation procedures and statistical analysis, empirical models were built and parameter values and co-variances were computed. The analysis of variance of the experimental and predicted data shows that the constructed models provide a fair approximation of actual experimental measurements. Finally, experimental details regarding the preparation of optimum composites as predicted by empirical models are discussed.

## V. CONCLUSIONS

The chemical, mechanical, and physical properties of natural fibers have distinct properties; cellulose content of these fibers varies from fiber to fiber. The moisture content lowers the mechanical properties. The mechanical properties of composites are influenced mainly by the adhesion between matrix and fibers. As in the case of glass-fibers, the adhesion properties can be changed by pre-treating the fibers. Novel processing techniques, chemical and physical modification methods are developed to improve the fiber-matrix adhesion resulting in the enhancement of mechanical properties of the composites. Up to now, the most important natural fibers are jute, flax, and coir. Yet the development of novel processing and modification methods is not finished. Further improvements can be expected, so that it might become possible to substitute technical-fibers in composites quite generally. Natural fibers are low-cost, recyclable and eco-friendly materials. Eco-friendliness and bio-degradability of these natural fibers may replace the glass and carbon fibers.

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