

# NATURAL FIBERS PLASTIC COMPOSITES FOR AUTOMOTIVE APPLICATIONS

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## **Abstract**

The use of natural fibers in composite plastics is gaining popularity in many areas, and particularly the automotive industry. The use of natural fibers in polymers can provide many advantages over other filler technologies, and areas of application appear limitless. The automotive industry is currently shifting to a “green” outlook, as consumers are looking for environmentally friendly vehicles. Natural fibers are a renewable natural resource, and are biodegradable, which is an important characteristic for components that must be disposed of at the end of their useful life. They are recyclable and can be easily converted into thermal energy through combustion, without leaving residue. Among the natural fibers with potential application as reinforcement for polymers, curauá fiber is one that recently received special attention from researchers. Curauá is a plant from the Bromeliad family, cultivated in the Brazilian Amazon region. The fiber is extracted from its leaves, providing a high mechanical strength over traditional fibers like sisal, jute and flax.

We have developed thermoplastic composites using either curauá fiber or wood flour. These materials provided a lighter weight product, with good physical properties and unique surface aesthetics. This paper reviews the properties of these bio composites in comparison with glass and mineral filled products. The products were tested in some automotive applications and the results will be discussed.

## **Introduction**

Today nearly 50% of vehicle internals are made of polymeric materials. According to the American Plastics council, the vehicles contain an average of 250 pounds of plastics, which accounts for 12% of their weight [1]. Since the plastic usage in vehicles is on the rise, the use of sustainable plastics is becoming increasingly important. Automakers are now taking initiatives to design parts using bio-based and renewable materials that are healthy and safe for workers, vehicle occupants and the environment. Furthermore, bio-based plastics may provide reduced petroleum use, life-cycle emissions and part weight while increasing recyclability of polymers.

Due to the many benefits that bio-based materials offer, automotive industry is seriously looking into these types of products. In this work we are presenting our efforts to incorporate bio fibers into plastics. Toyota is evaluating “Eco-Plastics” made from sugar cane or corn that is used in the spare tire and floor mats. DaimlerChrysler is looking into use of flax, coconut and abaca fibers in their Mercedes vehicles. Ford has been researching using biomaterials, such as soy-based foam for seating [2]. There are several advantages to using natural fibers in polymers. They come from renewable source that, in principle, is inexhaustible; they are biodegradable, which is an important characteristic for components that must be disposed of at the end of their useful life; they are recyclable and can be easily converted into thermal energy through combustion without leaving residue, causing less pollution and may gain additional carbon credit.

The usage of natural fibers as plastic reinforcement is not new. At the beginning of 20th century, phenyl-formaldehyde and melamine-formaldehyde resins compounded with paper or cotton were used for electric insulation. Applications in the automotive industry could already be found in the 60s, when coconut fibers were used to manufacture car seats, and PP (polypropylene) composites with wood flour, molded by compression, were applied as substrates for car interiors. In the 90s, PE (polyethylene) composites with natural fibers appeared to substitute wood in deck boards, fencing and industrial flooring. More recently, PVC (poly-vinyl chloride) composites with wood flour have been used for window and door frames, because of their balance of thermal resistance and stiffness [3].

Nevertheless, there was a preference for traditional materials such as glass fibers and mineral fillers, because natural fibers are characterized by a large free volume (low density), low thermal stability and high moisture absorption. In addition to this, the processing temperatures, even for plastics with low melting point, were too high to incorporate the natural fibers without degrading them. The high moisture content of natural fibers also caused a great problem for the compounders, used to working with materials of maximum moisture content from 1 to 2%. Even the extruders with degassing were inefficient to remove 5 to 8% moisture from the natural fibers [3].

However, in this decade, the interest for natural fibers as reinforcing material for thermoplastics has increased again. Particularly, in the automotive industry, the applications have grown very fast, not only in structural components but also as finishing parts.

## Natural Fibers

Due to the limited thermal stability of the natural fibers, only thermoplastics that soften at temperatures below 200°C are commonly used in these composites, such as polyolefins and PVC. The base resin and the fiber are chosen in accordance with their typical properties, application requirements, availability and cost. The fibers normally used have a length ranging from 3 to 8 mm and their concentration can vary from 20 to 70% on the composite. The most common are sisal, jute, hemp, coconut, flax, curauá, sugarcane bagasse and banana. The properties of the fibers can vary considerably depending on the part of the plant, which is removed (stem or leaves), where they were grown and the pretreatment fibers. In addition to the fiber and thermoplastic, the composites generally contain process stabilizers, coupling agents, lubricants and ultraviolet stabilizers.

## Characteristics of Natural Fibers

The main component of natural fibers is cellulose. The elementary unit of the cellulose macromolecule (Fig. 1) is anhydro- $\alpha$ -glucose, which contains three hydroxyl groups (-OH). These hydroxyls form hydrogen bonds inside the molecules itself (intramolecular) and between other cellulose molecules (intermolecular), as well as with hydroxyl groups from the air. Therefore, all natural fibers are hydrophilic, that is, they absorb water in the range of 8 to 12.6% [4].

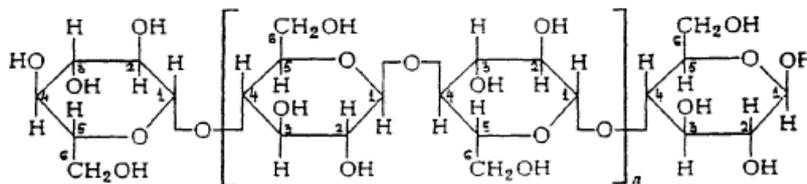


Figure 1: Cellulose macromolecule

Another important characteristic of natural fibers is their degree of polymerization. The molecules of each fiber differ in this aspect and, consequently, the fibers are a complex mixture of the homologous series  $(C_6H_{10}O_5)_n$ . Fibers from the base of the plants normally have a greater degree of polymerization (approximately 10,000). The fibrils of cellulose macromolecules form spirals along the fiber axis. The strength and stiffness of hemp, ramie and jute are correlated with the angle between the axis and the fibril of the fibers. The smaller this angle is, the higher the mechanical properties [4].

Cellulose contains different natural substances, the most important of which are lignin and different waxes. Observations made by scanning electronic microscope (SEM) reveal that the natural fibers are composed by a bundle of simple fibrils covered and bonded together by the lignin (Fig. 2).[5] Thus, lignin influences the structure, properties and morphology of the natural fibers. The waxes are responsible for the wettability and adhesion characteristics of the fibers and they can be removed by extraction with organic solvents [4].

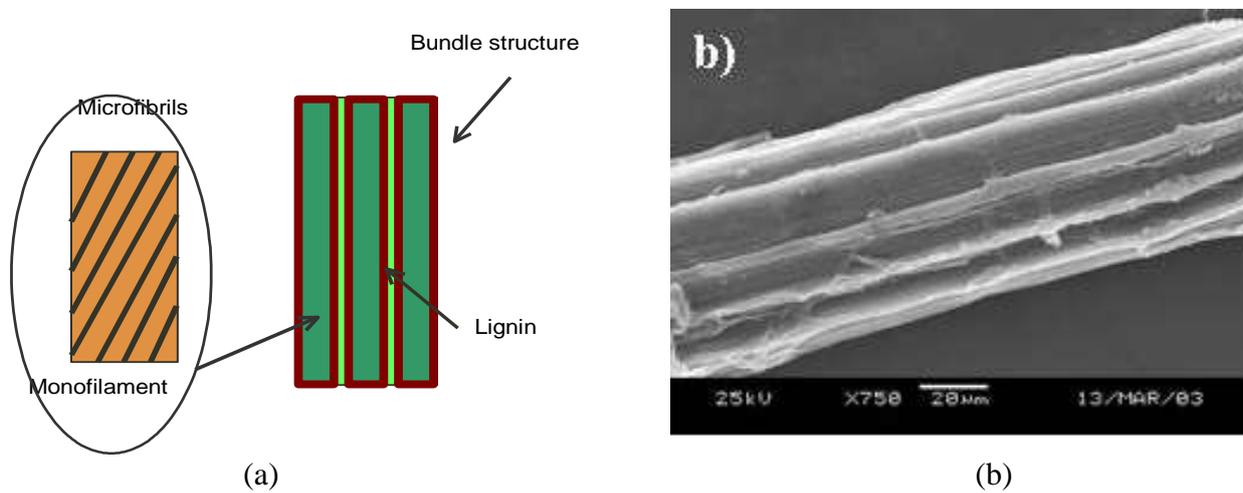


Figure 2: a) Structural model of the natural fiber and b) Micrograph obtained by SEM of the curauá fiber surface

### Curauá Fiber

Among the natural fibers with potential application as polymer reinforcement, curauá is one which has recently received special attention from researchers. Curauá is a plant from Bromeliad family, cultivated in the Brazilian Amazon region. Its leaves that reach up to 1.5 m in length and 4 cm in width are hard, erect and flat (Fig. 3). The fibers extracted from its leaves have high mechanical strength compared to other fibers like sisal, jute and flax. Its composition is shown in Table I [6].

Table I: Composition of Curauá

Composition	(%)
Cellulose	73.6
Hemicellulose	9.9
Holocellulose	83.5
Lignin	7.5
Ash content	0.9
Moisture	7.9



Figure 3: a) Curauá; b) Curauá fiber

Figure 4 shows the tensile modulus of different natural fibers compared to that of glass fiber. If specific modulus (relative to density) is considered, curauá stands out relative to other fibers, which enable composites with curauá fiber up to a 15% weight reduction [7].

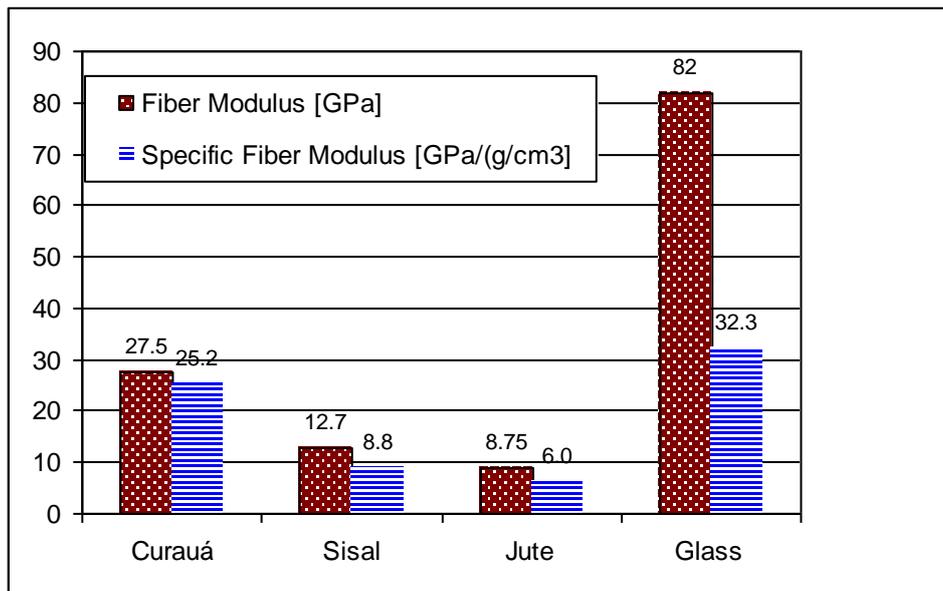


Figure 4: Comparison of the tensile modulus (nominal and specific) of natural fibers with glass fiber

### Fiber / Matrix Compatibility

The properties of the composites depend on those of the individual components and on their interfacial compatibility. The adhesion between fiber and matrix is obtained by mechanical anchoring of the fiber ends into the matrix. In many cases, the absorption of moisture by untreated fibers, poor wettability cause insufficient interfacial adhesion to the polymer matrix. The lack of interfacial interactions leads to internal strains, porosity and environmental degradation. The wettability of the fiber depends on the polymer viscosity and the surface tension of both materials. The surface tension of the polymer must be as low as possible, at least lower than that of the fiber. Therefore, modification of the fiber and/or polymer matrix is a key area for obtaining good composite properties [4].

### Modification of Natural Fibers

There are several different methods for modification of surface energy of the fiber and the polymer. Physical methods like stretching, calendaring and the production of hybrid yarns, change the structural and surface properties of the fiber and thereby influence the mechanical bonding in the matrix. Another physical method used is electric discharge (plasma or corona), which will increase the roughness of the fibers and activate its surface oxidation leading to an increase in aldehyde groups. It can also generate crosslinkings and free radicals [4].

Oxidizing agents such as sodium or calcium hypochlorite and hydrogen peroxides can be used to remove dust and oil from natural fibers. Alkaline treatment with NaOH, known as mercerization, extracts lignin and hemicellulose, increasing the tensile strength and the elongation at break of the composite. In this process the fibers are immersed in a 10 wt% solution of NaOH for 1 or 2 hours, followed by continuous washing and drying at 100°C [5]. Figure 5 shows the structure of treated fibers, seen by SEM.

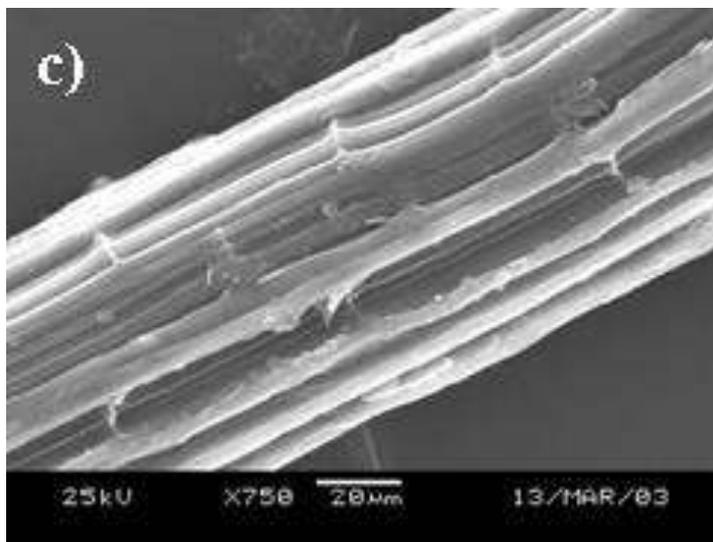


Figure 5: SEM micrograph of curauá fibers surface after alkaline treatment.

It is also possible to improve the compatibility of the fibers with the polymer matrix through chemical methods that introduce a third material (coupling agent), with intermediate properties of the two initial ones. There are several mechanisms of coupling in materials: they can eliminate the weak boundary layers; they can produce a flexible and tough layer; they can form a highly crosslinked interphase region with an intermediate modulus; they can improve wettability between the polymer and the substrate; they can create covalent bonds with both materials; and/or they can alter the acidity of the substrate surface [4].

The surface energy of the fibers is closely related to their hydrophilic character. Thus, modification of natural fibers with stearic acid, make fibers more hydrophobic and improves their dispersion in PP. Silane coupling agents may add hydrophilic properties to the interface, especially when amino-functional silanes are used as primers for reactive polymers. The organofunctional group of the silane reacts with the polymer, through copolymerization and/or the formation of an interpenetrating network. This curing reaction of a silane treated substrate enhances the wetting by the resin [4].

Another effective method of natural fiber modification is graft copolymerization, initiated by free radicals of the cellulose molecule. In this case, 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 suitable solutions like acrylonitrile or methyl methacrylate. The resulting copolymer has the characteristics of both, cellulose fibers and grafted polymer. For example, the treatment of natural fibers with hot PP/maleic anhydride copolymers provides covalent bonds across the interface. After this treatment, the surface energy of the fibers is increased to a level much closer to the surface energy of the matrix. This gives improved wettability and, consequently, higher interfacial adhesion. The PP chain allows segmental crystallization and cohesive coupling between the modified fiber and the PP matrix [4].

### **Processing of Composites with Natural Fibers**

The majority of composites with natural fibers and thermoplastics are produced by extrusion. Some processors use twin screw extruders to produce granulated composites; others use single screw extruders; and others use the latter to directly manufacture final products, like profiles or sheets. The fiber moisture can be removed in a previous process, by drying in a air circulating oven (8 h at 80°C), or in the extruder itself, if there is a vacuum outlet downstream in the barrel.

To produce thermoplastic composites with natural fibers, a fundamental condition is the quality of the fibers in terms of purity and particle size. The fibers should be ground in order to obtain a well defined size and distribution.

During the extrusion, there is a limiting factor that needs to be considered: at temperatures above 160°C and in the presence of oxygen, the fibers can undergo thermal oxidation resulting in darkening and, at higher temperatures, mechanical degradation will occur. From this point of view, the use of twin screw extruders has been preferred, because of the possibility of controlling the energy introduction via screw configuration and processing conditions, in such a way that a lower process temperature can be reliably ensured. The residence time of the fibers at the polymer melting temperature, without the presence of oxygen is short, between 10 and 20 seconds. In addition, excellent mixing and dispersing is achieved, especially when co-rotating, intermeshing screws are used. The best screw configuration is obtained experimentally, but profiles with deep flight depth should be preferred, because of the large volume occupied by the fibers ( bulk density around 0.15 kg/dm<sup>3</sup> ) [8].

The thermoplastic resin is premixed with additives and fed into the first housing of the extruder via gravimetric metering. The fiber is added at the extruder downstream by a lateral feeder directly into the zone in which the polymer is already molten. The air entrained with the fibers can escape via a back venting system in the unit. In this process, the fibers are homogeneously distributed in the molten resin so that each fiber is completely wet by the polymer. Subsequent vacuum venting over a length of 4D ensures that the residual moisture and volatiles are withdrawn. A machine like this, with diameter of 76 mm, can produce up to 1000 kg/h of PP or HDPE composite with a ratio from 40 to 70% of natural fibers [8].

### Properties of the Composites with Natural Fibers

The addition of natural fibers to thermoplastics maintains, at least, the properties of the unfilled polymers. Generally they bring an increase in the rigidity of the composite, which, in turn, makes it more brittle. However, this lower toughness is still higher than composites with mineral fillers, such as talc. UV protection is a cause for concern, because these composites tend to fade with time and need to be well stabilized and pigmented to reduce this effect. In addition to size and content, the type of fiber utilized has a great influence on the properties of the composite. Harder fibers, for example, generally increase rigidity and strength, but cause a darker coloration in the final product [8].

We have developed a product portfolio based on the use of natural fibers in thermoplastic composites. One of these materials, polypropylene (PP) reinforced with 30% of wood flour, MX07442, is aimed at replacing wood because, besides a similar appearance, it is more resistant to fungi and has better dimensional stability, due to slower and lower moisture absorption. Table II shows a comparison of this product with unfilled PP and PP reinforced with traditional fillers. As can be seen, MX07442 shows better flexural strength and tensile modulus compared to unfilled PP and talc filled PP coupled with lower density providing good weight to strength advantage for the wood flour filled product.

Table II: Comparative values of 30% wood powder filled MX07442 with unfilled PP and PP reinforced with 30% of glass fiber (GF) or talc (MF)

Property	PP	MX07442	PP+30%GF	PP+30%MF
Density ( g/cm <sup>3</sup> )	0.91	1.04	1.13	1.15
Tensile strength ( MPa )	19	28	65	25
Tensile Modulus (GPa )	1.4	2.3	4.5	2.2
Elongation at break (%)	50	2.5	3	5
Izod impact strength (KJ/m <sup>2</sup> )	5	9	9.5	8
Flexural strength (Mpa)	50	78	115	65

The Table III shows the property comparison of PA-6 reinforced with 20% of curauá fiber PX07444, with unfilled PA-6 and PA-6 reinforced with traditional fillers. As can be observed, the tensile and flexural properties of PX07444 are better than those of unfilled PA-6, but lower than PA-6 reinforced with glass fiber. In relation with PA-6 reinforced with 20% of talc, PX07444 shows higher tensile strength, lower tensile modulus and equal notched Izod impact strength. Its density is lower than PA-6 reinforced with talc or glass fiber, and its HDT is higher than unfilled PA-6 or PA-6 reinforced with talc, and is comparable to PA-6 reinforced with glass fiber.

Table III: Comparative values of 20% curauá filled PX07444 with unfilled PA-6 and PA-6 reinforced with 20% of glass fiber (GF) or talc (MF)

Property	PA6	PX07444	PA6+20%GF	PA6+20%MF
Density ( g/cm <sup>3</sup> )	1.14	1.18	1.27	1.27
Tensile strength ( MPa )	63	83	101	73
Tensile modulus (GPa )	1.4	5.5	6.5	6.5
Elongation at break(%)	>60	3	3	6
Izod impact strength (KJ/m <sup>2</sup> )	10	9	9	9
Flexural strength (Mpa)	95	115	160	115
HDT @ 1.82 MPa (°C)	57	186	194	110

Figure 6 shows a comparison of specific (divided by density) tensile strength and modulus of each product mentioned above. Taking this factor into consideration, one can note that the results of PX07444 are much closer to those of PA-6 reinforced with glass fiber, enabling it to replace the latter in specific applications, such as automobile interior components or electric tools housing, for example.

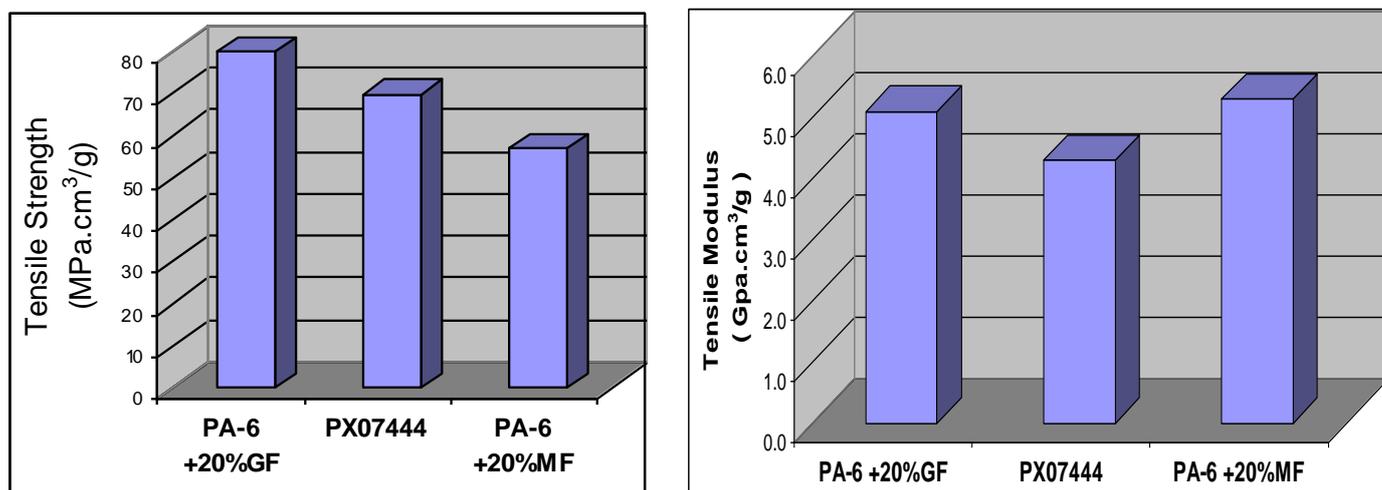


Figure 6: Comparison of the specific tensile strength and modulus (relative to density) of 20% curauá fiber filled PX07444 with 20% of glass fiber (GF) or talc (MF) reinforced PA-6.

### Automotive Applications

The Nylon 6 Curaua fiber filled product that was described above was evaluated in the automotive sun visor brackets and frame parts as shown in Figure 7. The molded parts showed very good surface aesthetics coupled with excellent mechanical properties, when molded using the following parameters:

- Drying conditions: 6h @ 100°C
- Melt temperature: 220 - 245°C
- Mold temperature: 70°C
- Injection pressure: 55 bar
- Holding pressure: 50 bar
- Cycle time: 24 s for the pin and 30 s for the frame



Part: Sun Visor  
Frame  
Mold: 2 cavities  
Part weight: 60g



Part: Sun Visor Hinge Pin  
Mold: 4 cavities  
Part weight: 11g



Assembled Part

Figure 7. Automotive sun visor bracket mounting parts made using 20% Curaua fiber filled Nylon 6 [9]

## Conclusions

Curauá fiber reinforced plastic composites provide excellent weight to strength properties compared to mineral filled products. The product properties track closely with that of glass fiber filled products but more work needed through improvements in fiber-polymer interactions to obtain direct offset products. The wood flour reinforced products show unique surface appearance (“wood look”), which may be of interest to industry where “green” requirements can be coupled with surface aesthetics. Overall these natural fiber plastic composites open up many opportunities not only in automotive but also in many other industrial applications where there is a need for “green” plastic composites.

The development of composites with natural fibers is just beginning. Although the ecological aspects of these fibers have been the primary reason to consider them for industrial applications, including automotive segment current and future research works should focus on how to increase the performance of these composites to reduce the gap in relation to traditional products. Continuous development of fiber treatments, new compounding techniques, optimization of fiber-matrix adhesion and the use of engineering thermoplastics, as base resin will bring significant improvement for the properties of these materials. This improved performance will make the replacement of glass fiber and mineral fillers, much more feasible in a large number of applications.

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