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# **Advancing Sustainable Development through Polymer-Natural Fiber Composites: A Comprehensive Review of Structure, Properties, and Processing Techniques**



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### **Abstract**

Exploitation of natural materials and solid waste in the preparation of polymer-natural fiber composites (PFCs) has been aimed at recent decades. A PFC is composed of a polymeric matrix and natural fiber filler; it is considered environmentally friendly materials as it can consume abundant natural fibers, such as agriculture waste from many crops and wood flour obtained from the furniture industry, in addition to polymer waste. The hybrid formulations have improved performance by enhancing the major characteristics that attain some of the sustainable development goals. This review discusses the structure composition, and types of PFCs. The related properties, additives, and processing techniques are also summarized. The recommended materials can be applied in construction, car accessories, building units, etc. Due to sustainable and environmental concerns, the preparation and development of PFCs are of interest. This work also highlights the deficiencies in conventional ways of manufacturing and provides suggested perspectives on new techniques to maximize the benefits of the related sustainable composites.

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*Keywords: polymers, natural fibers, processing, recycling, composites*



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### **1. Introduction**

The development and production of polymernatural fiber composites (PFCs) have experienced rapid global growth. The technical and commercial importance of these materials has expanded efforts for understanding their chemistry and related structural properties. In addition, exploration of new methodologies for producing these advanced materials has been the subject of research concern [1,2]. The influence of plastic waste on the environment is alarming due to ecological and commercial issues [3]. In addition, thousands of tons of different crops are produced; however, most of their wastes do not have any useful utilization. The abundant agricultural waste can be used to prepare fiber-reinforced polymer composites with highly environmental and commercial characteristics. Developing and analyzing such alternative materials could identify an ecological sound [4,5]. The abundant availability of natural fibers in many countries focuses primarily on the production and development of value-added PFCs, which can be used in many applications. Such natural fiber-plastic composites are well suited as wooden substitutes in many industrial and construction sectors. Reinforcement of different polymers, in virgin or recycled forms, with natural fibers has recently gained traction due to the topics of carbon reduction, lowcost, low-density, good specific properties, ease of processing, enhanced energy recovery, CO2 neutrality, biodegradability, and recyclability [6,7]. Due to the global shortage of fossil and wooden resources, the use of waste fiber-based polymer composites is expected to increase from 18% to 25% by the year 2030 [8]. The aim of this review is to focus on the preparation, properties, and processing of polymer-natural fiber composites based on the plastic matrix and natural waste, and the exploiting of these ingredients for useful products with an environmental impact. The related possible applications are also discussed, along with the key findings and future perspectives.

# **2. Overview on composites 2.1. Definition and need**

Composite is a cohesive material made by the combination of at least two ingredients of different compositions, it is a multifunctional material that provides characteristics not obtained by any discrete component. The process brings its own distinctive

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properties in terms of the ability to enhance some characteristics. The composite is considered a material that consists of two or more phases that are in contact with each other. With another definition, the composite can also be considered a mixture of homogeneous or heterogeneous materials that are combined together in such a way that any portion of the final composite will have the same properties [9]. Modern composites constitute a significant proportion of the materials in the engineering market, ranging from the daily smile products to the advanced industrial applications. While composites have already proven their worth as heavyweight and expensive materials, the current challenge is to enhance their properties and make them cost-effective. The use of composites rather than traditional materials and metals is still the most important need for saving both cost and weight and for achieving the required mechanical properties [10,11]. The reinforcement of polymeric matrices with different natural fibers has had a steady expansion in many uses; this results in an expected reduction in cost. In addition, the high performance of fiber-reinforced polymers can now be found in various applications. The usage of a composite and the selection of its constituents depend on many factors, such as the working environment, lifetime requirements, number of items produced, product shape, and possible savings in total cost and production techniques. These factors help in determining the optimum potential of the prepared composite [12,13]. Techniques of processing offer the desired characteristics that enhance the performance and quality of PFC. Type of fiber and polymer, fabrication process, and dispersion of fiber bundles into polymer are some of the techniques mentioned. Such behavior results in composites that possess better mechanical and physical properties, superior thermal stability, and higher erosion resistance compared with neat polymers [14,15].

#### **2.2. Properties and classifications**

As a composite consists of one or more discontinuous phases embedded in a continuous phase, the discontinuous phase, called "reinforcement" or "filler", is usually harder and stronger than the continuous one, while the continuous bulk phase is termed as "matrix". The properties of a composite strongly depend on the properties, percentages, shapes, sizes, and distribution of filler and matrix.

Furthermore, the interaction between all constituents is a governing factor. The properties of all constituents can interact in a synergistic manner, resulting in improved properties [16]. Many fillers exhibit very good strength properties, and to achieve this, these fillers should be bonded with a suitable matrix. The matrix penetrates the filler, forming a new filler-matrix interface that holds this filler in place. Matrix possesses the ability to transfer the applied load to the filler and distribute stress through the filler-matrix interface. Because of the low interfacial properties between some fillers and matrices, the ability of these fillers to reinforce the matrix can be reduced. Depending on the type of matrix and filler, some additives are considered to optimize the filler-matrix interface. Such additives can introduce an effective interlock between filler and matrix. In this case, matrix and filler are bonded well together after the covering or wetting of all filler particles by matrix. Actually, well-wetted filler can exhibit stronger interfacial adhesion and produce a composite with the desired

properties. Failure at this filler-matrix interface, called "debonding", is undesirable [17]. The composites can be classified into particle-reinforced composites and fiber-reinforced composites. In the particle-reinforced type, the filler has a particulate nature, such as spherical, cubic, tetragonal, platelet, and other regular or irregular shapes. In the polymer-natural fiber composites, the filler has a fibrous nature with a length greater than the cross-sectional dimension. The polymer-natural fiber composite may be single layer or multilayer in continuous and discontinuous forms. The dimensions of the reinforcement filler determine its capability to contribute its properties to the whole composite. Fibers are very effective in improving the fracture resistance of the matrix through their reinforcement effect. The matrix diffuses the bundles of fibers, transfers the external load to the fibers, and then protects them against damage or environmental attack [18]. Scheme 1 summarizes the plain classification of composite materials.



Scheme 1: General classification of composite materials.

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#### **3. Polymer-natural fiber composites**

PFC has been a subject of interest for the past few decades as it is cheaper, lighter, more attractive, and more applicable than other composites. Also, they have a longer life cycle and can be made to have the look of natural wood. Although not as stiff as solid wood, PFCs are stiffer than neat plastics [19,20]. So, there is an excellent opportunity with the fabrication of these composites for a wide group of applications, such as outdoor structures, packaging, automotive accessories, construction, flooring, fuel cylinders, windmill blades, industrial drive shafts, etc. The manufacturing of these composites does not only aim to improve the polymer quality, but also to give the international market cheap and biodegradable alternative composites. However, the main disadvantage of PFC is its relative high moisture absorption. Therefore, some simple chemical treatments may be performed for modification of the fibrous surface. The chemically treated natural fibers can strongly interact with the matrix; this is necessary for the application of PFCs [21,22]. In a detailed study, bagasse and rice straw waste fibers were used as fillers for unsaturated polyester resin in making new composites. The fibers were treated with silane coupling agent and added to the base till 15%. The microstructural date affirmed the treatment process and formation the cellulose-rich phase. It was found that Young`s modulus, flexural strength, and storage modulus were increased with composites that included treated fibers; they also showed stability against water and chemicals [23,24].

# **3.1. Ingredients of polymer-natural fiber composites**

The main materials utilized in PFCs are various types and sizes of lignocellulosic materials as fillers, in addition to various types and grades of polymers as matrices. Mainly, PFCs contain some additives that may be added during processing. Many agricultural wastes, such as bagasse, rice straw, wood flour, hemp, cotton, flax, jute, wheat husk, coir, sisal, ramie, bamboo, shells of various dry fruits, etc., can be applied as lignocellulosic materials that are mixed with different types of polymers [25,26]. As previously mentioned, all these types of fibers contain cellulose, hemicellulose, lignin, pectin, waxes, and other ingredients. The percentage of each constituent may differ from one fiber to another depending on the

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type of fiber and the condition of its growth [27,28]. The natural fibers should be free from dirt or any other foreign materials. In addition, moisture has to be removed from fibers before processing to improve the quality of the final product. The aspect ratio of natural fibers affects the mechanical and physical properties of the prepared PFC. Furthermore, a systematic methodology known as the "optimal formula" can be developed to create the trials and errors in the attempts to manufacture new composites or even modify existing ones [29,30]. Some extraction methods of fibers were performed to waste fiber, getting the cellulose-rich fibrils [31,32]. The polymer may be thermoset, such as polyester and epoxy, or thermoplastic, such as polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC). All these examples of polymers are the most commonly utilized matrices in PFCs. For thermosetting polymers, polyester is largely used more than epoxy because of its low cost. For thermoplastic polymers, PE has the highest usage percentage among all thermoplastics, at 83%. Respectively, PVC, PP, and other thermoplastics contribute by 9, 7, and 1%. Furthermore, any virgin or recycled plastic that can be melted and processed at a temperature lower than the degradation temperature of natural fillers (200°C) can be applied to produce PFCs [33,34].

#### **3.2. Environmental and commercial potentials**

Wastes of natural fibers obtained from different crops, trees, and the residual of furniture manufacturing are generally burned or disposed of, resulting in extra environmental pollution [35]. The same true is for plastics, which have a big environmental problem due to large amounts of their waste, non-biodegradability, and depletion of natural resources. Furthermore, the continuous growth of the worldwide consumption of plastics, reaching about 40% due to their short life cycle of less than one month, gives plastics a great environmental concern compared with other products. Several worldwide attempts, especially in developed countries, depend on taking advantage of these wastes along with the increased demand for alternatives for virgin materials. Therefore, more and more amounts of fibrous and plastic waste are used in the production of green composites [36]. Basically, many trials for the production of PFCs were based on the "cradle-tocradle" approach, where the material is recycled at the

end of its life cycle to produce a new product. So, we can nearly close the environmental loop and simulate the natural ecosystem [37].

In addition to the environmental concern, PFCs have commercial potential, along with the great increase in agricultural waste and plastics. The marketing of these composites is successful due to the manufacturing of new value-added products from large amounts of daily waste. Such cheap products, with their natural look, can achieve the desired properties [38,39]. There is a great opportunity for using agriculture wastes in the large-scale production of PFCs, where these wastes contribute nearly 20% of the total worldwide solid waste. Plastic waste contributes around 60%, of which 22% is recovered and 78% is disposed of. So, there is a main commercial advantage through the utilization of these fibrous and plastic wastes in the PFC industry [40,41]. Besides, the PFCs can achieve the increased demands for advanced products; their market will increase dramatically in the near future and then continue to grow in the long term. Furthermore, depending on the desired features, some synthetic additives can provide additional properties for these composites. More development processes for these new composites can greatly give a benefit to the agriculture waste through increasing of its profitability [42,43].

**3.3. Additives in polymer-natural fiber composites**  According to the nature of additives grafted into polymer-natural fiber composites, we can classify such additives into two groups: coupling agents (CPLs) and mineral additives. The group of CPL is the most commonly used one, which contains several families. Silanes, such as methyltrichlorosilane, methacryloxypropyl trimethoxy silane, diethylenetriaminopropyl trimethoxy silane, and maleated polyolefins, such as maleated polypropylene (MPP) and maleated polyethylene (MPE), are examples of these families [44,45]. For the group of mineral additives, the most commonly used additives are talc, calcium carbonate, kaoline, fiberglass, and wollastonite. Talc is the most commonly used one due to its good water absorption, which minimizes the moisture content of natural fibers. Furthermore, the

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natural similarity to oil makes talc good filler for hydrophobic plastics [46]. Carbon black and graphene oxide may be used. In HDPE-bagasse fiber composites, the effect of graphene oxide nanoparticles and exposure to electron beams was investigated. The composites were suggested for advanced industrial applications due to their enhanced physico-mechanical properties. In another related study, the carbon black and graphene oxide nanoparticles were added during fabrication via the melt-blending technique. The flexural properties, electrical conductivity, and electromagnetic interference shielding efficiency were altered [47,48].

The properties of PFCs are basically dependent on the developed interaction between natural fibers and matrix, which is enhanced in some cases by the addition of CPL in a small quantity, typically up to 5%. Natural fibers show low compatibility with some matrices, with a tendency to form aggregates and absorb much moisture. In this case, the fibrous material should be treated with CPLs that contains some reactive groups that are capable of coupling fibers and polymers together. Thus, CPLs enhance the compatibility between lignocellulosic fibers and matrix through the formation of homogeneous composite phase [49,50]. Coupling agents themselves are classified into organic, inorganic, and organometallic groups. The latter group, such as silanes and titanates, is the most commonly used because it contains hybrid compounds in its structures, this type is between organic and inorganic CPLs [51,52]. The group of organic CPLs, such as anhydrides, isocyanates, chlorotriazines, and some organic acids, is commonly used in PFCs. Organic CPLs have multifunctional groups in their molecular structures, which can interact with the hydroxyl groups of the cellulosic material through hydrogen bonding. The group of inorganic CPLs such as silicates is shortly applied, compared with the other two groups. They act as dispersing agents and counteract the polarity of the fiber surface, resulting in an improvement in the compatibility between fibers and polymers [53,54]. Table 2 summarizes the commercial additive materials used in PFCs and their effects.

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<b>Table 2:</b> Commercial additive materials used in PFCs, and their effect. <b>Materials</b>	<b>Main effect</b>	Data source
PVC/low-cost olive pits flour/calcium carbonate	• Generation of hydrogen bonding between cellulose and PVC • Increasing of tensile modulus and hardness at 10% • Increasing of water absorption and thickness swelling	Ali et al. [55]
Recycled expanded polystyrene/rice husk-coir biomass	• Increasing of mechanical properties with 3% biomass • Decreasing of water absorption with more content of coir	Bollakayala et al. [56]
Waste thermoplastic food pails/decorative high-pressure laminate sanding dust 60%/MPP 7%	• Maximizing the utilization of waste resources • Improved impact, tensile, and flexural strength • Low formaldehyde emissions and improved flame-retardancy	Huang et al. $[57]$
Low-cost and high-strength ultra-high-filled wood fiber/PP composites based on recycled PP and 60-85% recycled wood fiber from waste sawdust	• Robust mechanical properties with better creep resistance at 80% • Sustainable strategy for full- component utilization of the industrial residue	Tang et al. $[58]$
Waste expanded polystyrene/sawdust biomass using compression molding process	• Increasing of physico-mechanical properties with the increase in raw material quantity, molding pressure and polymer loading • Reducing of water absorption with the increase in compression pressure and polymer loading	Bollakayala et al. [59]
PP/HDPE/coupling agent/plain woven jute fabric/short bamboo fiber using co-rotating extrusion process	• Improving the mechanical properties and water absorption of the blend with increased proportion of fibers to 30% • SEM confirmed a physical bonding between matrix and fiber in the form of mechanical interlocking	Arya et al. $[60]$
Wood fibre 50% (hard wood, soft wood, long wood and wood chips)/PP/MPP	• Increasing the interfacial adhesion and mechanical strength with 5% <b>MPP</b> • Fully dispersed fillers • Decreasing of water sorption	Bledzki and Faruk [61]
Plywood boards based on virgin and recycled HDPE/wood veneer/MPE	• Chemical interaction between coupling of poplar fibers and polymer • Improving the mechanical and physical properties of the boards with 11% MPE	Ashori et al. $[62]$

**Table 2:** Commercial additive materials used in PFCs, and their effective

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**Table 2:** Continued….



Other methods of treatment could play the same role as direct additives, such as chemical and physical pretreatments, to achieve the compatible composite. The chemical pretreatment is more preferred than the physical pretreatment. There are many kinds of chemical pretreatments, such as alkaline treatment, acetylation, oxidative bleaching, nono-treatment etc, depending on the type, temperature, and concentration of the chemical reagent [69-71]. The physical

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pretreatments are shortly applied; some types, such as radiation, stretching, weaving, thermal, and steam, can already be performed on different types of fibers. The purpose of these physical methods is to change the properties of the fiber surface. One of these physical treatments is the electric discharge method, which activates the surface energy. The biological treatment using enzymes is considered an environmentally friendly process [72-75].

# **4. Applications of polymer-natural fiber composites**

Different PFCs can be manufactured using several processing techniques with various shapes and angles to obtain a suitable design for the selected application. These composites can be treated in the same manner as conventional wood, using the same cutting and sawing equipment. Various products of PFCs are available in the market as outdoor products [76,77]. Having higher mechanical behavior and thermal stability compared with neat plastics, PFCs can be used in many structural and building applications. There is a growing demand for the production of composites with high performance, low maintenance, and low cost. To meet this demand, PFCs are applied to produce products such as landscape timbers, fencing, railing, decking, window and door elements, molding, floors, roofing, siding, skylights, and indoor furniture. PFCs are extensively used in the production of strips that are applied in furniture industry; these strips contain fibrous layers that tighten together in the same direction. These composites are also used in producing panels that are manufactured by mixing natural fibers with definite polymers, giving composites similar to 100% plastic-based products. Hence, applicable PFC became a major concern in the market [78,79]. Furthermore, natural fiber-polymer composites are applied in the industry of automotive accessories because of their low density and environmental advantages over conventional composites. Actually, famous automotive companies such as Daimler, Opel, and Volkswagen apply PFCs in their car series, especially in the underbody panels and interior components such as door trim panels, rear shelves, and seat squabs [80].

In other works, polymer-natural fiber composites based on HDPE, industrial residues of pine sawdust, and MPE were designed for the production of an innovative shading system to apply in the forefront of buildings by means of shutter units through sequential extrusion. In another field of applications, it was reported that some fiber-reinforced thermoplastic composites are used in the drilling, refining, and production pipelines of oil [81,82]. Figure 1 illustrates some applicable PFC products.



**Figure 1:** Some applicable product of PFCs.

# **5. Manufacturing of polymer-natural fiber composites**

### **5.1. Conventional manufacturing**

The traditional techniques of manufacturing fiberbased polymer composites depend on the mixing and further compounding stages. The related techniques are selected depending on the type of filler and matrix used in addition to the desired shape. For the fiber-

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based thermosetting composites, the fibrous filler is attached to the thermoset, e.g., polyester, epoxy, or phenolic resins. Basically, these thermosetting polymers include some reactive groups that increase the attachment at the fiber-matrix interface. The curing parameters, such as operation time, curing temperature, and processing technique, can affect the properties of the composites. Actually, the wooden*\_*

thermosetting composites are commonly formulated through simple processing techniques such as hand layup, filament winding, pultrusion, spraying, resin transfer, and injection. Other techniques such as compression, centrifugal casting, cold press molding, continuous laminating, rotational molding, pressure bag molding, and vacuum forming may be applied, but with some difficulties because of the hard processing with natural and other fibers. By means of these different techniques, the fibers may be applied in the form of yarns, mats, or unidirectional tapes. More than one of these types may be impleaded during the same process. These types of fibers are saturated and impregnated within the thermosetting matrix that contains crosslinking agents, and then exposed to high temperatures for the completion of curing reaction. Hand layup, filament winding, and pultrusion formulations are the three most common processing techniques for the fiber-thermosetting composites [83,84].

The manufacturing of wooden-thermoplastic composites is more complicated than woodenthermosetting composites because the thermoplastic polymer itself has to be melted completely during mixing with natural fibers, and then the overall composite will be formed. Generally, the processing of wooden-thermoplastic composites is performed through a two-step process. The first step is "compounding", which is performed by feeding and dispersing fibrous material and other additives into the molten polymer through extrusion techniques. This step produces compounded composites in the form of pellets. The operation conditions of compounding directly affect the properties of the resultant composite. Suitable extrusion time and temperature with a moderate intensity of mixing can improve the quality of the compounded pellets and final composite. The second step is "shaping" or "formation", which converts the compounded pellets into the desired endshape. The shaping process is generally performed by injection molding or compression molding, in addition to other techniques. The combination between the two steps of compounding and formation in a single processing step, which is called "in-line processing", may be performed through the direct conversion of raw materials into an end-product [85,86]. The commonly used processing techniques for fiber-based thermoplastic composites are extrusion, injection molding, and compression molding [87,88].

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The types of extrusion, as the main applied processing technique, are single extrusion and twin extrusion. The first is the simplest and fastest extrusion type; however, it requires a further separate formulation process to give the pre-compounded material its final shape. The twin extrusion is the best type regarding the perfect profile of the product and gathering the mixing and formulation stages in one process. It may be processed by a co-rotating or counter-rotating twin extruder. All the types mentioned contain feeding, melting, metering, and die zones. In more details, the extrusion process is performed by forcing the molten polymer, including additives, through a die, and then the extruded composite is exposed to cooling using water or air. Finally, the obtained extruded composite can be cut into small pellets, which may be exposed to another extrusion process or another molding process, such as injection molding and compression molding, to get the desired end product. The injection molding technique is processed by forcing the molten composite into a cold mold that has the desired shape. However, compression molding is performed by pressing the molten composite between two mold halves close to each other for a definite time to get the end product with the shape of the cavity between these two halves. With those techniques, the final product is well exposed to water cooling before demolding [89,90].

# **5.2. Additive manufacturing**

Additive manufacturing (AM) is a modern technique for the manufacturing of polymer composites. It produces highly uniform and accurate objects, compared with conventional techniques, through the three-dimensional computerized model called "computer-aided design". The program is able to build up the polymer composite electronically by using molten, powdered, or liquid starting materials, layer-by-layer, reaching the final desired formulated geometry. Thus, AM can be called "rapid prototyping", "three-dimensional printing", and "solid freeform fabrication" [91,92]. Thermoplastic polymers, e.g. PE and PP, in addition to a few thermosets, are available to be used as hosting matrices in the AM technique; however, thermoplastics are more common. "Selective laser sintering", "stereolithography", "material extrusion", "powder fusion", and "binder jetting" are methods for formulations based on AM [93,94]. The advantages of AM can be summarized as low cost, time-saving, ease

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of processing, uniform production, and high-quality composite. Furthermore, challenges in conventional manufacturing techniques, such as viscosity and temperature change, filler consistency, filler-matrix interface, and aggregation, can be addressed and controlled by adjusting the inputs of the computed model [95]. Since the composite processing using AM is very limited to some accessories, it is needed to increase and develop this technique with broader applications to catch its outstanding properties and uniformity.

### **6. Conclusion and perspective**

- The waste of polymers and fibers can be utilized for preparing useful polymer composites to achieve environmental and economic impact.
- The successful grafting of fiber into polymers can promote the composite interface, resulting in improved mechanical, thermal, surface, and physical properties.
- The PFC can be improved via the treatment of fibers or polymers due to strong interfacial adhesion.
- The related PFCs can serve in different advanced industrial applications such as construction, car accessories, and buildings.
- The properties of PFCs depend on the types of matrix and filler, treatment process, and formulation technique.
- The common conventional techniques have some drawbacks, such as viscosity and temperature change, filler consistency, nonuniformity, and aggregation.
- The additive manufacturing processing technique and surface treatment are perspective points for future work. Such techniques are recommended for the development of polymernatural fiber composites with broader applications to achieve outstanding properties and uniformity.

### **7. Conflicts of interest**

There are no conflicts to declare.

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