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Green composites: A review of materials and perception

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Abstract

The purpose of manufacturing a composite material is to take advantage of the recognized properties of each component in order to build one that will be better suited to certain applications. A composite is a material that tries to combine the properties of its components (generally a polymer matrix and reinforcement) to improve certain characteristics such as breaking strength, or rigidity, for composites for mechanical uses.

Previously, composite materials were most often thermoplastic or thermosetting matrices reinforced with carbon fibers, glass ... However, in recent years, composite materials reinforced with natural fibers have attracted the attention of researchers because they could be an effective solution to solve the problems of environment, biodegradability and oil depletion. This review paper is mostly an introduction to the description of composite materials with vegetable reinforcements.

Keywords: *Biocomposite, Biodegradables, Biopolymers, Interface treatments, Lignocellulosic biomass,*

I. Introduction

Every second on earth, an area of forest equivalent to 19 tennis courts disappears; the desert increases by 78 m^2 in China, 1600 t of ice melts in Greenland and 720t of CO_2 are emitted from oil into the atmosphere [1]. These figures show that it is crucial to take into account the effects of human activities on the environment. It is in this context that eco-materials are being developed and that the term sustainable development is increasingly mentioned. This notion of sustainable development requires consideration of environmental issues.

Biodegradable polymers (biopolymers), which have been the subject of much research, appear to be an increasingly credible alternative to address environmental concerns. Biopolymers are divided into two classes: agro-polymers, such as starch, proteins, etc., and biopolyesters (PHA, PLA, etc.). However, while bioplastics have seen extensive growth in applications such as disposable packaging films, they still lack the mechanical properties to be used in applications such as those found in the automotive industry [2].

The current obstacles that face the use of PLA are its properties which are still considered too weak for them to be able to definitively replace petroleum-based polymers and which are mainly their resilience, their water resistance as well as the cost. Often considered too high. In order to overcome these shortcomings, it is therefore necessary to seek solutions which make it possible to improve the various properties of these biopolymers, either by incorporating natural reinforcement, thus allowing the development of composites. [3].

These composites are characterized by two main aspects, economic and technical. Composites are materials with high technical performance with interesting physical and mechanical properties for relatively easy manufacturing and implementation. Over the past decade, the vision of industrial development has changed direction by integrating the environmental aspect into the search for innovative materials. The objective of technical performance having been achieved and in view of the question rose today concerning the environment and the impact of the evolution of man on his environment; a new problem arises, namely, can man continue to evolve while being in adequacy with the environment in which he lives? To answer this problem, researchers and industrialists have sought to use plant resources for the production of new materials. Today, agro-materials or bio-sourced materials partly meet this need [4].

For the reinforcement of polymers, plant fibers have important specific mechanical properties (at least for some of them) and many advantages if we are interested in environmental impacts. These are renewable resources, naturally biodegradable, neutral in terms of CO_2 emissions into the atmosphere and requiring little energy to produce.

These fibers can replace glass fibers in many areas or make it possible to reach new markets [5]. Currently, food waste is a major concern for businesses, governments and consumers. One of the largest sources of food waste occurs during industrial processing, where substantial by-products are generated. Fruit processing creates many of these by-products, from undesirables to fruit skins, seeds, and fleshy parts. These by-products make up to 30% of the initial mass of processed fruit. Millions of tons of fruit waste are generated worldwide from spoilage and industrial by-products, so finding alternative uses for fruit waste is essential to increasing their values. This goal can be achieved by transforming fruit waste into reinforcement and integrating it into polymeric materials [6].

Many research works have been carried out to modify and improve the properties of polymers, by adding reinforcements of plant origin to form composites [7].

This present review towards a description of biocomposite materials with vegetable reinforcements, and situate the context of development and use of these products. Knowledge of the structure and chemical composition of each of the constituents is necessary to understand the study of the interactions between the reinforcing fibers and the matrices.

II. Biocomposites

Biocomposite is the general term for composite materials that are entirely or to a large extent produced from biomass: natural fibers are combined with petrochemical or biological polymers to achieve very good, often well-matched, mechanical properties.

Other, bio-composites is the combination of bio-sourced or petro-sourced matrix with natural fibers. The most common types are wood-plastic composites and natural fiber composites [6].

III. Biopolymers matrix

Biodegradable polymers are sustainable polymers synthesized from renewable resources such as biomass instead of the conventional fossil resources such as petroleum oil and natural gas, preferably based on biological and biochemical processes.

They are characterized by the nature of carbon neutral or carbon offset in which the atmospheric CO_2 concentration does not increase even after their incineration [6,7].

III.1. Classification of biodegradable polymers

Biodegradable polymers and biopolymers can be produced from renewable and fossil resources. Briefly, there are 3 main types of materials, which are called bio-polymers: those of natural origin, artificial ones and their composites [8,9].

- Materials of natural origin are those synthesized by living beings: animals, plants and micro-organisms. We find:

The family of polysaccharides (carbohydrates) such as starch, cellulose, lignin, chitin, the family of proteins such as gluten, casein, collagen and gelatin, and the family of lipids; rapeseed, soybean, sunflower oils To this family, we could include carbonaceous elastomers such as natural rubber [10,11].

Polymers of bacterial origin resulting from the fermentation of sugars and starch by bacteria or produced by genetically modified micro-organisms (biotechnology). Depending on the bacteria, various polymers are obtained such as PHA, [10,11].

Biosynthetic polymers, whose monomer from biomass are obtained by fermentation. The polycondensation of these bio-monomers gives polyesters; the best known is poly (lactic) acid PLA [10,11].

A new generation of polymers from renewable resources but this time non-biodegradable; traditional bio-based polymers such as PE, PET, PA, [10,11].

- Biodegradable materials from petroleum resources obtained by industrial synthesis processes. The best known are PBS (poly butylene succinate), PBAT (poly butylene adipate terephthalate), and PCL (poly caprolactone) [10,11].

On the other hand, there are traditional polymers of fossil origin such as PE or PET to which an additive is added which promotes its biodegradability. The end of life of these materials results in a biodegradation of the additives and in a physical (visual) degradation without molecular disintegration of the synthetic elements [10,11].

Table 1: General classification of polymers [10-12].

	Not Bio-sourced	**Bio-sourced**
Biodegradable	**Bio-polyesters** from conventional chemical synthesis: Polycaprolactone PCL, Polyester amide PEA, co-polyester: PBSA, PBAT, etc…	**Agro-polymers** polysaccharides: starch, cellulose; gelatine, casein, gluten… **Bio-polyesters from microorganisms:** PHAs, **Bio-polyesters synthesized from renewable monomers**:

		PLA…
Not Biodegradable	**Traditional polymers** PE, PET, …	**Traditional polymers** from synthase of renewable origin, PE …

IV. Lignocellulosic biomass reinforcement

Lignocellulosic biomass is the most abundantly available form of terrestrial biomass on Earth, with an estimated 181.5 billion tonnes generated annually, among which about 7 billion tonnes are produced from dedicated agricultural, grass and forest land and another 1.2 billion tonnes stem from agricultural residues [13].

Thus, lignocellulosic biomass represents an immense, inexpensive and renewable resource for the production of biofuels, bio-based chemicals and materials. It includes mainly crop residues (cane bagasse, rice straw, rice hulls, corn Stover, corn stalk, wheat straw, barley straw and pulp), organic wastes (waste newsprint, recycled paper, pulp/paper mill sludge), woody biomass (e.g., aspen and poplar, pine and spruce and forestry residues), herbaceous biomass (coastal Bermuda grass, switchgrass, reed canary grass and timothy grass) and organic fraction of municipal solid wastes [30].

Interest in lignocellulosic biomass reinforced polymer composite materials is rapidly increasing both in terms of industrial applications and basic research. Their availability, renewability, inexpensively, biodegradable, low density and as well as satisfactory mechanical properties make them an attractive ecological alternative to glass, carbon and synthetic fibers used for the manufacture of composites [14].

IV.1. Chemical composition of lignocellulosic biomass

Lignocellulosic biomass is composed of three main biopolymers: cellulose, hemicellulose and lignin [15-17].

In a much lower proportion they also contain extractables, proteins and certain inorganic compounds [18].

➢ Cellulose

Cellulose is the most abundantly synthesized biopolymer by the plant kingdom on the planet. This parietal polysaccharide represents 40% of all terrestrial organic matter. Its molecular structure, widely studied, allows its supramolecular organization, based on the formation of hydrogen bonds in large numbers. This supramolecular organization defines crystalline zones and amorphous zones in the assembly, in variable proportions depending on the origin and the treatment imposed on the raw material. It nevertheless makes it possible to consider cellulose as a semi-crystalline biopolymer [19].

Cellulose is made up of a sequence of gluçopyranose cycles with a glycosidic bond of the β 1-4 type. The degree of polymerization (DP) varies according to the plant species. It is for example 10000 for cotton, 9000 for hemp, flax and ramie. However, the glass transition temperature of amorphous cellulose, which would be above 200°C, cannot be assessed with precision because cellulose degrades thermally from 180°C.

The melting temperature of the crystalline zones is not accessible because it is located beyond the degradation temperature [20, 22].

➢ Hemicelluloses

Hemicelluloses are by definition polysaccharides that are soluble in water and can be extracted from the wall of plant cells by alkaline solutions. They are amorphous polysaccharides, with a molecular mass lower than that of cellulose. They are composed of neutral sugars: xylose, arabinose, galactose, glucose, mannose, and uronic acids. In their natural state, they have a degree of polymerization between 200 and 300 and their structure depends on the plant species [23].

➢ Lignin

Lignin is after cellulose the most abundant renewable organic material on the surface of the earth. Lignin is three-dimensional polymers resulting from the radical polymerization of three phenylpropenic alcohols whose structure depends on the plant species: coumaric alcohol, coniferyl alcohol and sinapyl alcohol [24].

IV.2. Advantages of using natural fibers

In general, the use of natural fibers as reinforcements for composite materials is justified for:

- Develop materials and technologies that take into account the impact on the environment. Natural fibers are biodegradable and renewable materials.
- Promote a local resource in industrialized or less industrialized countries. Natural fiber composites open up new outlets for agricultural products.

The choice of fibers coming from a natural environment and presenting interesting mechanical performances is guided by:

- Their origin and function.
- Their availability: a natural fiber is considered available if the volume of fibers present on the market is sufficient to produce industrial parts. Its production is linked to outlets.

- Their often complex composition and structure. Although composite materials are often presented as original and innovative, the natural environment already offers a wide range of them. A multitude of natural materials, some rigid but light, such as walnut shells, and others flexible but resistant, such as wood, owe their mechanical properties to their composite structure [25,26].

V. Surface modification

The hydrophilic nature of natural fibers comes from lignocellulose which contains many polar hydroxyl groups. These fibers are therefore intrinsically incompatible with hydrophobic thermoplastics.

The main limits of use of these fibers, such as Reinforcement in such matrices leads to poor fiber-matrix interfacial adhesion, and the difficulty in mixing is mainly due to the low wettability of the fibers with the matrix which would lead to low interface composites [27, 28-31].

According to the literature, many attempts have been made in modifying the fiber-matrix interfacial characteristic, due to incompatibility between the two phases [28-31]. Among these various methods of modification, chemical treatment is mentioned.

These chemical modifications call upon interface agents. Indeed, the interface plays a preponderant role in the behavior of the composite when it is subjected to constraints. The interface agent ensures reinforcement/matrix compatibility by transmitting the stresses on either side of the interface, without relative displacement. Several essential roles are assigned to the interfaces for a good behavior of the composite:

- Ensure physical continuity from one component to another through all materials, preventing, for example, the formation of porosity.
- Transmit effort. Indeed, the matrix distributes and transmits the forces to the fibers via the interfaces [32].

The most widely used technique is alkaline treatment [33-36]. The alkalization process affects the natural fibers and brings a clear performance with respect to the quality of fiber-matrix adhesion. This treatment increases the fiber/resin contact surface. Some authors advocate that this improvement is due to the elimination of non-cellulose compounds (cuticle layers oils, oils and dirt) from the surface. Thus, it leads to an improvement of the interfacial bond between the fiber and the matrix [37,38]. Alkaline treatment is often practiced to extract residual lignin, hemicellulose, and to remove natural and artificial impurities. It also promotes fibrillation of the bundles [39,40]. The concentration of the alkali, the temperature and the treatment time of the fibers are essential parameters of the treatment process. The optimization of these parameters contributes to the improvement of the mechanical properties of the composite material [41].

Zafar et al. [42] reported jute fiber and poly lactic acid biocomposites obtained by treated as well as in treated NaOH solution using a twin-screw extruder and found different effects of treatment of jute fiber at FTIR and stated the formation of hydrogen bonds. Polarized light optical microscopy pictures clearly intimated there is an improvement in trans crystallinity at the matrix fiber interface. In the scanning electron microscopy the images showed the matrix covered fiber surfaces when composites combined with surface-treated jute fibers. They mentioned that untreated jute fiber in PLA Tensile and modulus strength has been improved.

Jayabal et al. [43] reported the tensile, flexural and impact properties of a woven coir polyester composites when the surface is treated by the alkali solution. He mentioned with the help of scanning electron microscopy (SEM) images that the coir fiber treated with the alkali solution have improved the tensile, flexural and impact properties when compared with the raw and untreated coir fiber.

Siregar et al. [44] have studied the impact of alkali treatment on the mechanical properties of short Pineapple Leaf Fiber (PALF) reinforced High Impact phenyl ethylene (HIPS) composites. Where NaOH is employed as an alkali. The fibers were treated with completely different concentration of NaOH (0%, 2% and 4%) resolution. It was determined that adhesion of fiber/matrix is improved by treating the short PALF fibers with an alkali resolution before production of a composite.

VI. State of the art on composite materials

Natural fiber reinforced composites have received considerable attention in the research community. We can cite as examples the work carried out by certain researchers.

Kimura et al. [45] They examined compression-molded PLA composites reinforced with ramie fibres, using non-twisted commingled yarn made of ramie and PLA fibres as raw material. The best mechanical values, i.e. tensile strength, bending strength and stiffness, were obtained at ramie fiber volume content between 45% and 65%. Large proportions of ramie also increased the notched impact strength considerably.

A study of injection molded flax and man-made Cordenka fiber reinforced PLA was carried out by Bax and Müssig [46] The composites' mechanical qualities improved with a rising fiber mass content of 10 up to 30%. Flax/PLA and Cordenka/PLA composites clearly differed in their impact strength characteristics: While the impact strength of pure PLA could be multiplied by adding Cordenka, the values of flax/PLA composites were inferior to the pure matrix.

David et al [47] determined the mechanical properties of the PLA/jute fiber composites and showed that the tensile strength of composites was significantly higher than that of PLA. But the elongation at break of the composites is still very low as about 2%.

Ngo et al [48] focused on tensile and flexural properties of varied volume fraction of kenaf fiber; Vf (20, 40 and 60%) reinforced polymer composites. It is obtained, when increase in kenaf fiber, the tensile and flexural strength increased which is up to the maximum value and the properties were decreased at Vf of 40%. Further increase of fiber Vf at 60%, deteriorate occurs. The fibres tend to aggregate which weakens the interfacial area and debonding tends to take place between the fibres and matrix that at Vf greater than 50%.

Ochi et al. [49] studied the properties of kenaf fiber /PLA composites fabricated at a molding temperature of 160°C. The result showed that the flexural strength of the kenaf fiber reinforced composites increased linearly up to a fiber content of 50%. The biodegradability of kenaf/PLA composites was examined for four weeks using a garbage processing machine. The results have shown that the weight of composites decreased 38% after four weeks of fabrication.

Hammiche et al. [50] assessed the effects of the Alfa fiber content and the fiber surface treatment on the mechanical properties as well as the degradation of composites based on poly (hydroxybutyrate‐co‐valerate) (PHBV) in an aqueous medium. The idea behind is to develop eco-friendly composites by exploiting the use of natural fibers and PHBV biopolymer. Based on the obtained results, the main findings are summarized below: Morphological changes occurred on the fiber surface after chemical surface treatment.

Improvement of mechanical performance of composites reinforced with lignocellulosic fibers needs strong adhesion. Alkali treatment of alfa surface fibers enhances both of the tensile strength (by 4–16% according the percentage of alfa fibers) and the rigidity (by 7–25%) of PHBV/alfa composites. SEM micrographs highlighted differences in microstructure of untreated and treated composites confirming the enhancement of the quality of the fiber-matrix interface owing fiber surface-treatment. The treated composites were highly biodegradable compared to both neat PHBV and untreated composites. All elaborated composites undergo degradation more pronounced into seawater (compared to distilled water) that is wealthy in microbial populations indicating their environmentally friendly character.

Klaai et al. [51] reported the hydrolytic aging of biocomposites which were composed of Alfa fiber and Polylactic acid (PLA) prepared by extrusion. To improve the fiber/matrix compatibility, the Alfa fiber has been treated with a dispersing agent which is BYK W-980. Spectroscopic (FTIR), thermal and water absorption tests before and after hydrolytic aging of biocomposites have been studied and compared. The results revealed that PLA/Alfa biocomposites in the presence of BYK W-980 are more resistant to hydrolytic aging compared to untreated composites.

Klaai et al. [52] focused on tensile and thermal properties of prickly pear seed fibres as reinforcement in polylactic acid biocomposites.

The results show that the mechanical and the thermal properties of the biocomposites with treated fiber are better than that of neat PLA and the biocomposites with untreated fiber.

VII. Conclusions

This review has provided a concise summary of the major material attributes of green composites; description, Knowledge of the structure and chemical composition of each of the constituents.

Undoubtedly, environment-friendly, fully biodegradable reinforced plastics or ‘green’ composite materials will play a major role in greening the products of the future.

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