Leaf based natural biofibers and biocomposites: An overview

[1]Darsan. R.S, [2]Stanly Jones Retnam .B, [3]M Sivapragash

[1,2]Dept. of Automobile Engg, Noorul Islam Center for Higher Education, Kumaracoil, Kanyakumari, Tamil Nadu, 629180 India
[3]Dept. of Mechanical Engineering, M G college of Engineering, Thiruvallam, Thiruvananthapuram, Kerala, 695017 India
Email: ndmresearch11@gmail.com, er.stanleybjones@yahoo.co.in; mspragash@rediffmail.com

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Abstract—Natural biofibers are extracted from the plants and animals. Applications based on them go back to ages. As the civilization grows the reinvention of biofibers into more specific uses has taken place. Usage goes to various fields such as medical, automobile, food packing industries, textiles etc. Natural biofibers have the capacity to replace the synthetic fibers in terms of characteristics. Nonrenewable based artificial fibers reinforced composites have biodegradability issues, thus making them less environment friendly. This paved the path for the development of biocomposites. And they are biodegradable, bioactive, bio compatible and eco-friendly. Production of more volumes of biofibers will lead to extensive cultivation of plants, which will help in reducing the carbon dioxide emission, thus can help in controlling the global warming rates. Surface modification of fibers enhances the characteristics of biocomposites produced.

Keywords— Natural leaf biofibers; chemical and physical characteristics; extraction; surface modification; biocomposites.

I. INTRODUCTION

Natural biofibers (NBF) are extensively used by researchers, over synthetic fibers which are made from nonrenewable resources. They are environmental friendly, biodegradable, cheaper, low density and bio compatible as they are formed naturally[1]. The current trend in scientific research is to extend the application of natural biofibers in almost all fields to make cent percent green materials using biocomposites. Biodegradable composites are made from biopolymers which degrade without causing harmful effects to the surrounding [2,3]. The basic content in natural biofibers is cellulose /lignocellulose. They are obtained from leaf, seed, wood, stalk and grass /reed [3-5].

Manmade fibers and polymers from petrochemical are currently being widely used. These artificial materials have good chemical, physical and thermal properties, which suits for different types of applications. In addition to the fact that they are biodegradable, they also release toxic chemicals to environment during recycling process[6]. Present circumstances lead to the research growth and production of biocomposites[7]. Biopolymers are used along with NBFs for production of biocomposites[8]. Poly(lctic acid) (PLA) belonging to the aliphatic polyesters family are biodegradable. PLAs are produced using renewable sources like corn sugar, potato and cane sugar [9].

There are different classifications of natural biofibers on the basis of the source from which they are extracted. Natural biofibers are found in plants, animals and minerals. Natural biofibers founds in plants are extracted from different parts of the plants, from materials such as soft wood, grass and reed. The fiber from the stalk is rice ; sisal is an example of fiber from leaf; jute is an example for blast(stem), cotton fiber is from seed; the fiber from fruit is coir. Some of the NBFs, from plants such as cotton and jute are used as fabrics in textiles over a long period of time. Bamboo and coir fibers are used in construction industry from time immemorial. Thus, it is evident that use of NBFs are more relevant in fields such as construction, automobile, aerospace and medical field etc.

NBFs are best alternates for the conventional fibers made from nonrenewable resources. Depleting nonrenewable resources and the potential hazard to the environment are greater concerns regarding the use of conventional fibers such as fiber glass, carbon, aramid etc. NBFs are biodegradable fibers and are agro- based, which are from renewable sources . Mass production of NBFs require larger scale agriculture turnovers. This will lead to utilization of waste land which otherwise is underutilized, causing the reduction of carbon foot prints for the developing and developed countries.

Biocomposites made out of NBFs are in the form of short length randomly oriented fibers and long length continuous fibers [6]. They are obtained from wood or structural members from the parts of the plant structure. Biodegradability of both NBFs and biopolymers have some challenges too[10] . Weak adhesion between fiber and polymer are caused by the hydrophobic nature. Uneven thermal growth takes place due to the differences in thermal expansions of fiber and matrix separately. Treatment of the fibers and enhancement of the polymer strength by addition are some of the solutions to the problem[11].

This paper generally focuses on different types of natural biofibers based on origin and their characteristics. Leaf based natural biofibers are currently left out for degradation once harvesting of crops has taken place. Also the paper discusses different methods of extraction for most commonly used NBFs. There is tremendous scope for research on NBFs and biopolymers, especially poly(lactic acid)(PLA).

II. LEAF BASED BIOFIBERS

The manmade fibers such as carbon, E-glass, aramids etc. give exact range at which it shows it specific characteristics [2]. The characteristics shown in the case of NBFs vary depending on the part from which the fiber is being extracted. Fiber can be extracted from wood, bast, stem, leaf, etc. Natural biofibers can be subdivided on the basis of the part of plant from which the fiber is extracted [6].

(a) Stem or bast are termed as soft. They are in the form of groups of several clusters of large quantities of cell entangled spirally to form strands. Each of these are glued to the nearby strands. They can resist bleaching and other treatments .

(b) Leafs are hard structural fibers formed and can undergo chemical treatments.

(c) The remaining fibers from fruits, seeds, etc. act as a central bone.

A. Chemical composition of leaf natural biofibers

Main content of plant fibers is lignocellulose [2]. Cellulose, hemicellulose and lignin together form the structure of natural biofibers. Pectin, wax micro fibrillars and moisture are the remaining elements in natural biofibers. The Chemical composition of the leaf natural biofibers is shown in table 1. Cellulose in NBFs consists of repeating units of anhydroglucose and β-1,4-glycosidic, in which there are three hydroxyl groups[11]. This makes them hydrophilic in nature. Hemicellulose consist of polysaccharides in a combination of 5 – and 6-carbon ring sugars[3,10]. They form support structures for the cellulose microfibrils. Lignin, a complex structure formed by aliphatic and aromatic polymer structures, insoluble in almost all solvents and cannot form monomeric group on synthesis. Structure, property, and morphology depend on lignin content. The other name of pectin is hetro polysaccharides. Remaining elements are wax, microfibrillar angle and moisture content.

Table 1

Chemical composition of leaf natural biofibers [1-8, 10 - 14]							
Type of fiber	**Cellulose**	**Lignin**	**Hemicellulose**	**Pectin**	**Wax**	**Micro Fibrillar**	**Moisture content**
	%wt	**%wt**	**%wt**	**%wt**	**%wt**	**Deg.**	**%wt**
Sisal	67-78	8-11	10-14	10.0	2.0	10-22	11
Abaca	56-63	12-13	9-13	1.0	0.2	N.S	1.6
Pineapple	80-83	8-12	15-20	2-4	4-7	8-15	1-3
Henequen	60-77	8-13	4-8	N.S	0.5	N.S	N.S
Oil Palm	65	19	0-22	N.S	0.5	N.S	N.S
Banana	63-64	5	5-10	N.S	N.S	14	10-22

N.S – Not Specified

B. Physical & Mechanical properties of leaf natural biofibers

Physical characteristic of the fiber filament is an important factor which influences the mechanical characteristics of the fiber. The modulus of fiber increases with decrease in diameter of the fiber. Internal structure will influence tensile strength, density, modulus etc. The physical and mechanical characteristics of leaf natural biofibers are given in Table 2. Tensile modulus of oil palm is less compared to others. Bigger diameter of the oil fiber attributes to the lesser value of tensile strength and modulus. It seems that the tensile strength and young's modulus of the pineapple shows a wide range. This is because the diameter of the fiber lies over a wide range. Starting range of tensile strength of banana fiber is due to the lager value of percentage of elongation.

Table 2

Physical & Mechanical Characteristics of leaf natural biofibers						
Type of fiber	**Density g/cm3**	**Diameter µm**	**Tensile Strength MPa**	**Young's modulus GPa**	**Elongation at break %**	**References**
Sisal	1.2-1.45	50-200	468-855	9.0-22	2-7	2, 3,4, 11,12
Abaca	1.5	10-30	400-813	31.1-33.6		3, 10
Pineapple	1.5	20-80	170-1627	287-1130	1.6	2,3,4, 10, 11
Henequen			430-570	10.1-16.3	3.7-5.9	10,13
Oil Palm	0.7-1.55	150-500	248	2.5-3.2	0.5 - 3.2	4, 10,
Banana	1.35	12-30	529-914	27-32	3	3,4, 10, 12,13,14

III. EXTRACTION OF PINEAPPLE, SISAL AND BANANA LEAF BIOFIBERS

NBFs are extracted from cellulose/lignocellulosic by bacteria and fungi, mechanical and chemical methods. Retting the most common method of extraction of fibers using bacteria and fungi has deployed around the raw material to remove lignin, pectin and other elements. They attack the cellulose surrounding the fiber and separate the fiber from the parent structure. Good quality fibers are produced in atmospheric retting and prolong period can cause defect to the fiber itself[13].

The chemical retting method obtains the fibers by employing alkalis, mild acids and enzymes. Alkali used in this case is sodium hydroxide. Sulphuric acid and oxalic acid are added with some quantity of detergents. The acidic agent is the one used in extraction of fibers from the source. Enzymes like pectinases, hemicelluloses and celluloses are usually used before or after chemical treatments. Factors affecting the chemical treatments are concentration of chemicals, temperature and treatment time .

Mechanical methods include the peeling of fibers from the composite fiber/cellulose structure [15]. Peeled fibers are like ribbons and can be done either manually or by using machines. Process of peeling is thus simply known as ribboning [14]. Manual ribboning may take place with the help of locally made devices. NBFs are peeled using powered decorticator strikers. Initially the devices compresse the soaked (semi retted) raw materials followed by striking, will lead to separation of NBFs from the primary source into separated fiber volumes[15,16].

C. Sisal leaf biofibers

Sisal leaf fibers (SLFs) are produced from Agave sisilana, grown in tropical and sub-tropical regions around the world . 200-250 leaves are produced in a plant, with 1000 -1200 fiber bundles per leaf out of which 4% are fibers and remaining largely constitute water. Three different types of fibers are produced from sisal leafs namely mechanical, ribbon and xylem. Fibers obtained from the periphery are mechanical fibers and are in the shape of thick horse shoe. Median line of the sisal leaf produces ribbon fibers and have high mechanical properties. Xylem fibers are Opposite to median fibers, which are thin walled and are irregular in shape . These fibers are obtained from sisal leaf either by retting or mechanical decorticators. After extraction they are washed in water to remove the solvents during retting and mechanical extraction processes .

D. Pineapple leaf biofibers

Pineapple leaf biofibers (PALFs) are produced from pineapple plant belonging to Anannis comosus variety of Bromeliaceae plant family found in topical parts of American, African and Asian countries [17]. PALFs are not directly used in any engineering application because of its smaller diameter [18]. They are reinforced with matrix to produce composites which have wide range of applications. Hand separation method is employed to produce PALFs, because power machine will damage the fibers during extraction. Two different fibers are produced during extraction processs. Large vascular bundles is of about 75wt% from the top lamina and fine fiber about is 25wt% from the bottom lamina. Another method is abrasive combing using fibers being drawn between abrasive sandpapers.

E. Banana leaf biofibers

Most edible and dominating banana variety is formed by combination of two species, M. acuminate Colla and M. balbisiana Colla[19]. Banana plants are 2-7m tall herbaceous plants, having around 30 leaves (30-60cm wide and 2 m long)[20]. The natural biofibers can be extracted from banana leaves and banana stems. NBFs are extracted by chemical, mechanical and biological methods. Use of chemical methods is not environmental friendly and mechanical methods find it difficult to remove the gum from the raw material [21]. They are more ecofriendly and give higher yield from biological process. Degumming process should be employed after extraction of fiber before usage. Microbial enzymes such as pectinases which use Solid-state fermentation process will yield good amount of NBFs during extraction [21].

IV. SURAFACE MODIFICATION METHODS

NBFs are ecofriendly and biodegradable and their application to various other fields are restricted due to some problems. The NBFs are hydrophilic, when used with hydrophobic matrix with higher water absorption which will lead to reduction in mechanical characteristics. Hydrophilic NBFs and hydrophobic matrix lead to poor interfacial adhesion, this being the major concern with NBFs/biopolymer matrix. Wax, lignin and pectin can be removed by using sodium lauryl sulphate or potassium permanganate which reduce the hydrogen bonding, and increase surface roughness. In some fibers like banana biofibers, degumming and bleaching are required to remove the glue material from the fiber [21, 22].

F. Silane treatment

Silane (3-APS) with a fixed wt % (0.6 wt% for banana natural fiber and 5wt% for pineapple leaf fiber) with respect to the fiber weight is mixed with ethanol/water at 60:40 ratio and stirred for some time to get a correct mixture.The pH adjustment to 4 can be done by adding acetic acid and mechanical stirring for 1 hour. The fibers are dipped in the solution for 3 hours, subsequently water washed and dried outside for 3 days [23]. They are dried again in a vacuum oven at 80^0C for one day. Here the moisture combines with hydrolysable alkoxy to form silane. Silane treatment will enhance the mechanical and thermal stability of NBFs/biopolymer composite .

G. Alkali treatment

The most used chemical treatment for NBFs is alkali (mercerization) treatment . Alkoxides are formed from the ionization of hydroxyl group on addition of acqueous sodium hydroxide (NaOH). Treatment increases the amorphous cellulose on consuming crystalline cellulose . Sisal and pineapple fiber are dipped in 5% aqueous NaOH solution for 2 to 72 hours under room temperature . For banana fiber same process under 1% aqueous NaOH is carried out. Treated NBFs are washed with water and added with few drops of acetic acid and continued washing with water till NaOH is removed from fiber. Wax, oil or lignin etc. from the external surfaces are removed. The structure is depolymerized, leading to exposure of long fibers. Degumming operation is done by 2% alkali at 200^0C for 90 seconds under 1.5MPa .

H. Alkali and silane treatment

NBF bundles after alkali treatment are dried and immersed in silane solution for 1 hour. After this process, the NBF bundles are dried outside for a day and vacuum dried at 80^0C for 12 hours [22, 23].

I. Graft copolymerization

Chemical coupling method is used to enhance the interfacial adhesion between the NBFs and polymer matrix. For specific application to fix vinyl monomer to NBFs, a special agent Redox ($CuSO_4$-$NaIO_4$) is used. In this process acids are not used as an agent. Result of graft copolymerization gives an optimized orderly arrangement of specific agent on the NBFs structure. Graft copolymerization of the pineapple leaf fiber results in increase in thermal characteristics. For sisal/ polypropylene composite, there is an improvement in interfacial adhesion [16].

J. Benzoylation treatment

Benzoylation treatment decreases the hydrophilicity of NBFs and enhances the thermal characteristics . On Benzyl chloride treatment, it forms a link between the hydrophilic NBFs and the hydrophobic polymer. This will aide better interaction between the NBFs and polymers, because of this there is betterment in mechanical and thermal characteristics of the composites.

V. BIOCPMPOSITES

Composites made of biocompatible and biodegradable materials are ecofriendly. The degraded product will not cause toxic waste to the surrounding . Such types of composites are made with NBFs and biopolymers. There are different biopolymers made from the natural route, such as starch, cellulose, poly(lactic acids) (PLA) etc. [13] . Final degradation will produce carbon dioxide and water. Different co-polymers of PLAs are biocompatible and biodegradable. PLAs have very good Physical characteristics given in the Table 3.

K. Sisal natural biofibers, biocomposites

Sisal NBF and PLA biocomposites are made with different fiber volume fractions. The natural biofibers are surface treated one time or more, before they are fabricated. The resultant composite is mechanically tested and compared with pure and untreated NBFs biocomposites. Double surface treated sisal natural biofibers(SNBFs)/poly(lactic acid) at 20% fiber volume fraction show 30% higher tensile strength than untreated fiber biocomposites [26]. Alkali and alkylation treated SNBFs and PLA composite show a greater effect on the fracture behavior [27]. Different surface treated SNBFs with PLA composite were studied. Alkali treated and alkali and silane treated fiber show higher tensile strength [28].

L. Pineapple natural biofibers, biocomposites

Pineapple natural biofibers(PNBFs) have smaller diameter and less amount of elongation, hence the tensile strength and modulus are higher compared to others natural fibers. Silane and alkali treated PNBFs are used in preparation of PLA biocomposite. Different mechanical characterizations are performed on them. The properties of the treated fiber composites are much superior to others. Result show that treated PNBFs is found to the best suited fiber for replacing manmade fibers [23]. Short length PNBFs are extruded in a twin screw extruder, with different PNBFs weight percentages. Results show that the properties depend on the fiber weight percentage [29]. A comparative study conducted with PNBFs and thermoplastic starch fiber as reinforcing agents with PLA matrix element. It is found that thermoplastic starch with PLA shows best result [18].

M. Banana natural biofibers, biocomposites

Green nanocomposites were prepared using melt blending and subsequently by injection moulding . Silane treated banana natural biofibers (BNBFs) show improvement in tensile and flexural properties . Addition of nanoclay to the silane treated BNBFs with PLA shows increase in melting point. Surface treated BNBFs and originally modified nanoclay along with PLA prepared by melt mixing method [30]. Newly prepared composite is used as cutlery.

Table 3

Properties of Biodegradable Aliphatic Polyester [2, 3, 4, 7, 24,25]				
Polymer	**Compressive /tensile strength (MPa)**	**Young's Modulus (GPa)**	**Tensile Modulus (GPa)**	**Elongation (%)**
Poly(L-lactic acid) PLLA	28- 2300	4.8	1.5-2.7	5-10
Poly(D,L-lactic acid) PDLLA	29-150	1.9	1.9	3–10
Poly(Glycolic acid) PGA	350-920	12.5	5-7	15-20
Poly(D,L-lactic-co-glycolic acid (85/15)	41.4-55.2	2.0	1.4-2.8	3-10
Poly(D,L-lactic-co- glycolic acid (75/25)	41.4-55.2	2.0	-	3-10
Poly(D,L-lactic-co-glycolic acid) (50/50)	41.4-55.2	2.0	1.4-2.8	3-10

VI. CONCLUSION

The characteristics of natural biofibers show that they are found to be the best replacement for manmade fibers in terms of physical, mechanical and biodegradation property. Sisal, pineapple and banana are found to be most used natural biofibers. This is due to large cellulose content and smaller diameter of the fibers, which will lead to better mechanical characteristics. Extraction methods adopted vary for different NBFs. Out of these, biological method is found to be the best, as yield and quality of fiber is high. NBFs have some of the properties which are superior to manmade fibers, but require fiber surface modification to enhance the properties. Biocomposites made of treated and untreated fiber show variation in properties and improved compared to pure PLA matrix. Treated NBFs found to have optimum properties compared to untreated biocomposites. Out of three NBFs, sisal, pineapple and banana, pineapple is found to be the best replacement for manmade composites.

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