A Comprehensive Review on Mechanical Properties of Natural Cellulosic Fiber Reinforced PLA Composites

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Abstract Due to the alarming rise in environmental regulations and ecological threats, research on natural fiber-based biodegradable polymer composites has burgeoned. One such polymer which has received a plethora of attention in recent years is Poly-Lactic Acid (PLA). Natural fibers possess distinct physical, chemical, and mechanical properties. In addition, it is recyclable, less expensive, and easily available. However, natural fibers have some drawbacks like moisture absorption, subsequent swelling, poor chemical resistance, and poor interfacial interactions with polymer matrices. These shortcomings can be overcome through surface modification techniques which include the usage of plasma technology and chemical agents such as sodium chloride, silane, stearic acid, etc. This paper presents a review of the mechanical properties of PLA based natural fiber composites. This review also demonstrates the influence of fiber surface treatment and manufacturing methods on the properties of PLA composites.

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1 Introduction

Poly-lactic acid, most commonly known as (PLA) is a thermoplastic polymer obtained from natural resources like maize or sugarcane which can be an efficient substitute to polymers produced from petroleum reserves. PLA is one of the front-runners in the bio-polymer market due to its connatural properties and it also has a production capacity of 140,000 tonnes per annum. This is also the second-largest production volume of any bioplastic [1]. Furthermore, it can be manufactured using existing equipment that is designed and currently used to produce petroleum-based plastics. Thus, it is reasonably cost-efficient to produce PLA [2].

PLA is exceptionally adaptable and can be extruded into sheet or film, injection molded into plastic parts, thermoformed into packaging items, or spun into fibers for non-woven and textiles [3]. The peculiar properties for end applications can be offered by blending PLA with other polymers. These blends are investigated for applications like sutures, implants, drug delivery systems, and tissue engineering in the medical field. In addition, PLA is commonly used in plastic films, bottles, and biomedical prototypes [4]. Also, PLA constricts while heating thereby making it suitable to employ it as a shrink wrap material and in some 3-D printing applications (lost PLA casting) [5].

The end-of-life options for PLA include reusing, renewable energy recovery (incineration), biodegradation, and feed-stock recovery. The production of PLA utilizes 65% less energy than the production of conventional plastics. Therefore, consumers and manufacturers can use resources more efficiently by reducing waste. Polymers that can degrade naturally have led researchers with a panacea to waste disposal problems by obtaining the same level of utility. It produces non-toxic compounds when burned, unlike many plastics, and also it doesn't break down quickly on land or in the ocean [6–8]. On the other hand, the major short-comings in its commercial usage is its high price, low impact resistance, hydrophobicity, and inability to withstand increased temperature. These issues are solved by using natural fibers as reinforcements, which enables us to obtain specific desirable properties of the composites for specific end-user applications. By considering the increasing environmental concerns, the interest of the public for biopolymers and natural fiber-reinforced polymers has proliferated. The sisal, banana, flax, coir, jute, etc. are some of the naturally occurring fibers that can be reinforced with PLA [9]. The fiber content as well as the processing parameters will have a significant impact on the characteristics of natural fiber-reinforced composites. Therefore, reasonable processing methods like direct injection, extrusion injection, extrusion compression moulding, etc. and the corresponding parameters should be conscientiously chosen to obtain an ideal end product. This paper will act as an effective source to academicians, industry

personnel, scientists, and researchers about the mechanical properties of untreated and treated natural fibers based PLA composites [10].

2 Natural Fibers Reinforced in PLA

Natural fibers are generally extracted from plant sources as well as animal sources and some of these include banana, sisal, flax, jute, coir, silk, wool, etc., [11, 12]. In particular, the plant-based natural fibers that are reinforced in PLA to improve the properties of the resulting composite are discussed below.

2.1 Banana Fiber

Banana fiber (*botanical name: Musa ulugurensiswarb*) is popularly called Musa fiber. This fiber is lightweight, has good strength with adequate elongation, and is biodegradable. The fiber is extracted from the banana stem, these stems are thereafter dried in sun for 7–8 h and then in a hot oven for 1 day at 75–80 °C, thereby removing the water absorbed in the banana fiber [13]. Banana fiber reinforced PLA composites show higher flexural and tensile properties on comparing to neat PLA. These composites can be employed in areas where lightweight is of prime importance and strength is not an issue [2].

2.2 Sisal Fiber

Sisal fiber (*botanical name: Agave sisalana*) is the most prominently used natural fiber and it is cultivated widely across the globe. Some of its useful properties include good toughness, low cost, high durability, minimum wear, and recyclability. They were extracted from the sisal plant from the outer skin of the sisal leaf and kept under the sun and dried. This resulted in obtaining good moisture resistance and sufficient load absorbing properties. Table 1 enlists the different mechanical and chemical properties of sisal [14]. Sisal fiber-reinforced in PLA provides moderate tensile, superior impact, and flexural properties [15].

2.3 Flax Fiber

Flax (*botanical name: Linumusitatissimum*) is a cellulose fiber, crystalline in the structure which absorbs and releases water quickly and is stiffer in handling. Flax is extracted by soaking it for 7–10 days and drying it in the atmosphere. In doing so, the

Table 1 Mechanical and chemical properties of natural fibers

Fibers	Banana [13]	Sisal [14]	Flax [16]	Jute [18]	Coir [20]
Mechanical properties					
Density (g/cm^3)	750–950	1.45	1.5	1.4	1.4
Tensile strength (MPa)	392–677	350–360	600–1100	400–800	17–20
Young's modulus (GPa)	27–32	3.8	60–80	30	3.79
Poisson ratio	0.28	0.4	0.24	0.27	0.3
Chemical properties					
Cellulose (%)	60–65	65–70	64.1	50–57	43–44
Hemi cellulose (%)	6–19	14	16.7	20–24	25
Lignin (%)	5–10	9.9	2.0	8–10	45–84
Moisture content (%)	–	10	10	65	5–20

fiber is broken down, brushed, and finally it is crimped. The compressive strength is about 80% of tensile strength [16]. Flax fiber with PLA matrices produces promising mechanical properties. However, PLA/flax composites are not widely used as the bonding between PLA and flax is poor [17].

2.4 Jute Fiber

Jute fiber (*botanical name: Corchorusolitorius*) is widely used cellulose and a bast fiber for various industrial applications. Initially, it is dried in the sunlight, and then it is soaked in water, simultaneously breaking and crimping is done and jute fiber is extracted [18]. Jute reinforced with PLA has shown better fiber/matrix bonding, higher production efficacy, and considerably good mechanical properties. In addition to these, jute fibers also enhance the Izod strength and tensile modulus of the polymer [19].

2.5 Coir Fiber

Coir fiber (*botanical name: Cocos nucifera*) are available in India in large amounts, especially in the southern states of India. Coir is extracted from the coconut husk with the help of a disintegrator machine. The husk is then beaten and combined using a revolver screener, and it is kept in the sun and dried [20]. Coir fibers show greater elongation at break (15–40%) in contrast to sisal, jute, etc. which has low elongation. Therefore ductility of PLA can be improved when coir is reinforced in PLA [21].

3 Surface Modification of Natural Fibers

Due to environmental issues, several studies are taken up by the researchers to find a substitute for synthetic fibers. Natural fibers are biodegradable, less expensive, abundant, and have low density. But they have some downsides such as moisture absorption, swelling property, and poor compatibility [22, 23]. To overcome these drawbacks, physical (plasma treatment) and chemical (alkali, silane) treatments are made on the surface of the natural fibers. Physical techniques like plasma treatment will introduce different functional groups on natural fiber surfaces and will form strong covalent bonds with the matrix. This leads to strong adhesion and enhancement in properties. Chemical techniques can be done by introducing chemical agents like peroxides, alkali, water-repelling agents, etc. [24]. It has been demonstrated that chemical treatments can remarkably enhance the properties of natural fibers by changing their crystalline structure and eliminating weak constituents like hemicelluloses and lignin from the fiber structure [25].

3.1 Surface Modified Banana Fiber Reinforced PLA

Banana fibers were taken and cut to the required length and soaked in detergent to remove the impurities. The fibers were then dried in sunlight for minimum 2 days. Mercerization is then carried out by dipping the fiber into sodium hydroxide solution (NaOH) 1 N for one hour at ambient temperature. Finally, the fiber was taken out and dried at ambient temperature for 24 h and in Oven at 80 °C for 12 h. The composite material was produced by the twin-screw extrusion process (acceleration 50 rpm) and then it was subjected to the injection process. Finally, a composite was obtained. Then Mechanical, Thermal analysis, Heat deflection test was conducted using the sample. The result obtained is shown in Table 2. From the above result, it was clear that surface-modified banana fiber shows good bonding with PLA and the composite obtained shows a considerable rise in the tensile modulus, tensile strength, and impact strength [26].

3.2 Surface Modified Sisal Fiber Reinforced PLA

Sisal fibers were taken and cut into the required amount and soaked into di-oxane solution of MPS-g-PLA at 1 wt% for 48 h. Acetic acid was used to control the pH. After this, the fibers were taken out and dried at ambient temperature for 48 h. Then the fibers were allowed to react with MPS-g-PLA at 120 °C for 2 h. The composite material was prepared by twin-screw extrusion process at 75 rpm and 190 °C for 10 min. After that samples were prepared by injection moulding. Then tests like heat deflection, Mechanical, Thermal analysis, were conducted using the sample. The

Table 2 Properties of natural fibers before and after surface modifications

Fibers	Properties	Tensile strength (MPa)	Flexural strength (MPa)	Impact strength (kJ/m^2)
Banana [26]	Untreated	14.61	–	19.1
	Treated	16.01	–	19.69
	Percent increase (%)	9.58	–	3.09
Sisal [27]	Untreated	56.68	97	3.25
	Treated	59.71	104	3.1
	Percent increase (%)	5.35	7.22	−4.84
Jute [28]	Untreated	55	67	12.98
	Treated	62	78	14.25
	Percent increase (%)	12.73	16.42	9.78
Coir [21]	Untreated	4	25	–
	Treated	7	35	–
	Percent increase (%)	75	40	–
Flax [29]	Untreated	55	–	–
	Treated	63.4	–	–
	Percent increase (%)	11.23	–	–

result obtained is shown in Table 2, from the above result, it was clear that surface-modified sisal fiber showed good bonding with PLA and the composite obtained shows a significant improvement in properties like impact strength, tensile strength, and tensile modulus [27].

3.3 Surface Modified Jute Fiber Reinforced PLA

Jute fiber is obtained from the bark of the jute plant. It has 3 major chemical compounds cellulose (58–63%), Hemicellulose (20–24%), lignin (12–15%). First, the jute fibers were extracted by the retting process. After the retting process, the fiber was soaked in water for one hour. Then it is taken out and dried at 50 °C. After this, the Mercerization process was done by immersing the fiber in the required percentage of NaOH for 6 h at 70 °C with stirring and shaking. Acetic acid was used as the neutralizing agent to absorb excess alkali. Then the alkali-treated jute fiber was passed for the bleaching process. Here the fiber was added with hydrogen peroxide with occasional stirring for 45 min. Finally, the fiber was taken out and dried in the Oven at 50 °C until it reaches constant weight. The composite material

was prepared by a vertical injection moulding process with the required mixture of treated jute fiber. Then Mechanical, Thermal analysis, heat deflection tests were conducted using the samples obtained. The result obtained is shown in Table 2. We can conclude from the result that surface-modified Jute fiber shows good bonding with PLA and the composite obtained shows a considerable increase in mechanical properties [28].

3.4 Surface Modified Coir Fiber Reinforced PLA

Coir fibers are extracted from the husk of coconut It has three main chemical compounds cellulose (36–43%), Hemicellulose (0.2%), lignin (41–45%). Coir fibers were taken and cut to the required length and it is washed well with distilled water. The prepared fiber was immersed in the NaOH solution for 1 h at 70 °C; it was then washed again to remove the excess NaOH. In order to remove the absorbed alkali, Acetic acid was used as the neutralizing agent. After this, the fiber was dried in the oven at 60 °C for one day. The composite sample was prepared by compression moulding process. Then Mechanical, Thermal analysis, Heat Deflection tests were conducted using the sample. The result obtained is shown in Table 2. It is understood from the obtained results that surface-modified coir fiber shows good bonding with PLA and the composite obtained shows a considerable increase in mechanical properties [21].

3.5 Surface Modified Flax Fiber Reinforced PLA

A flax fiber comprised of a middle lamella region, which is composed of pectin and a small quantity of lignin. Flax fibers were initially soaked in NaOH (5%) solution for 20 min at 23 °C. The fibers were removed, filtered, and thoroughly washed in distilled water. It was then rinsed with a diluted solution of HCl (neutralizing agent) to remove excessive NaOH and it was washed again with distilled water. Then it was dried in a vacuum at 65 °C for 3 h. The hot pressing method was used to manufacture the composite. It was then subjected to a tensile test in a universal testing machine. The results obtained were Tensile strength (MPa) untreated Flax fiber = 55, treated = 63.4. From the above result, it is clear that surface-modified flax fiber removed certain weakly adhering amorphous polysaccharides, most importantly pectins and hemicelluloses from the middle lamellae. Other noticeable treatments show that NaOH reaches the secondary layer and attacks the surrounding polymers and decreases the strength of the fiber thereby affecting the mechanical properties of a composite. By adding the proper amount of NaOH (%) and proper drying we can avoid the intrusion of NaOH into the inner layers [29].

4 Manufacturing Methods

Natural fiber-based PLA composites can be produced by a wide range of processes. Some of the most common manufacturing methods are discussed below. Figure 1 depicts the classification of the common processing techniques used to fabricate PLA-based composites.

4.1 Direct Injection Moulding (DIM)

In direct injection moulding, the PLA and natural fibers were thoroughly mixed and were fed into the direct injection moulding machine. Various parts in DIM are hopper, nozzle, heaters, driving unit, and the mould assembly. The temperature of the barrel can be set according to our requirements like 160 °C, 170 °C, etc. The temperature from hopper to the nozzle will remain almost the same. The pressure can also be set according to our requirements. The screw is provided in the injection moulding machine which will be rotated using the driving unit. This screw is useful for mixing PLA and natural fiber. PLA and natural fiber which is fed in the hopper reaches the barrel and both these get mixed and heated. During heating, PLA gets converted into liquid. This mixed reinforced composite enters into the nozzle and from the nozzle, it enters into the mould-assembly. These composites are held and cooled for a particular time. Then it is removed from the mould assembly and can be cut into a number of pieces to prepare samples [30].

4.2 Extrusion Injection Moulding (EIM)

In this type of moulding, PLA and fiber were fed into the hopper. The parts present in this moulding are hoppers, heaters, nozzle, cooling tank, guide rollers, and pelletizer. From the hopper, PLA and fiber enter into the barrel and it is compounded at a required temperature. Then these reinforced composites enter into the cooling tank where it

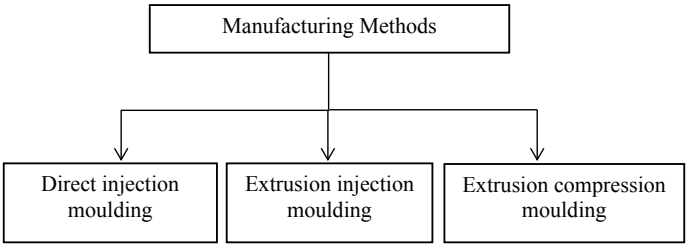

Fig. 1 Classification of manufacturing methods for PLA based composites

is cooled and it further enters into pelletizer. Pelletizer is used to cut the composite pellets into a number of pieces. Now the composite pellets coming out of pelletizer were fed into the injection moulding machine and it is fabricated [30].

4.3 Extrusion Compression Moulding (ECM)

The compounding in this process is similar to extrusion injection moulding. The various parts are also similar to injection moulding. The melted PLA and natural fiber were collected in the preheated mould. From the preheated mould, it is transferred to the compression moulding machine where it is pressed under the required temperature and pressure. Then it is taken from the mould and was cut into various pieces [2].

5 Conclusion

There has been a tremendous increase in the demand for newer materials that are environmentally safe and also reduces the burden on petroleum-based products. One such material that has gained a lot of attention is natural fiber-reinforced bio-polymer composites. Researchers are working on identifying different natural fibers that can act as a replacement for synthetic fibers like carbon, glass and Kevlar. The bio-polymer composites fabricated by using the natural fibers have properties comparable to the synthetic fiber-based composites. Furthermore, recent research works are concentrating on the surface treatment of natural fibers to enhance the interfacial bonding between the matrix and the fiber thereby good quality composites can be obtained. The cost associated with using bio-polymers as matrix material is considerably high and this makes it difficult for the commercialization of green composites. Therefore, further research should be explored to develop low-cost manufacturing techniques for fabricating green composites. This will ensure the reduction in the usage of synthetic fiber-based non-biodegradable composites that contribute to a sustainable environment.

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