Mechanical properties of natural fiber reinforced polylactide composites: A review

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Abstract. Nowadays, there is an augmented concern for the development of not only well-designed but also fully sustainable materials. The extraction of renewable resources and disposal processes that would not harm our eco-system is associated with material sustainability. In this respect, biodegradable polymers and natural fibers are suitable alternatives to many non-degradable composites which one lightweight. Many researchers are trying to fabricate composites by adding these two through various means and characterizing their mechanical properties to find the suitability of these for different applications. The present article reviews the literature on the fabrication of polylactide composite reinforced with natural fibers, both of which are biodegradable fully; further, the article reviews the methods used by the researcher for characterizing mechanical properties of these composites.

1. Introduction
Nowadays, biodegradable polymers are gaining popularity because of awareness of environmental pollution caused by conventional polymers. The development of biodegradable polymers promotes the use of alternative materials that are environmentally friendly and use them in composite making for various engineering applications. Agricultural crop residues such as rice husk, coconut, flax, oil palm, rice straw, pineapple leaf, banana, jute, bamboo, and sugar palm are produced in very large quantities across the globe[1,2]. They are low cost, easily degradable, and available in abundance. Among these huge quantities of residues, only a little quantity of these are used as domestic fuel or fertilizer, and the maximum portion of these residues are burnt in fields, resulting in Air pollution[3]. These can be overcome by utilizing suitable agricultural residues as reinforcement in the fabrication of natural fiber reinforced polymer composites[4–8]
Many researchers have successfully used these natural fibers in thermoplastics as reinforcement; the advantage being that these composites are recyclable. Most polymers themself do not possess load-bearing capacity as they don't possess the required strength, stiffness, stability. But fibers do have good stiffness and strength but difficult to be used in load-bearing applications because their structure is typically fibrous like say in order to exploit the advantages of these to compensate for the disadvantages these materials are put together in the form of composites[9].

However, research on biodegradable polymers as composite matrices are limited in comparison to conventional polymers (both thermoplastic and thermoset) since they are not readily available and are priced more [10]. The existing literature generally dealt with the study on the effect of adding the natural fiber in conventional polymers on mechanical properties only; the influence of adding natural fibers in the biodegradable matrix-like PLA is hardly dealt with. Adding natural fiber with PLA results in completely biodegradable composites which can be treated to be sustainable development, taking into consideration, both economic and ecological aspects [11]. The classifications of Natural fiber-reinforced bio-composites are shown in figure 1.

![Figure 1. Classifications of bio-composites (adapted from Mohanty et al. [8]).](image)

1.1 Polylactide (PLA)
Polylactide is a biodegradable thermoplastic aliphatic polymer based on lactic acid which can be derived from 100% renewable resources like corn starch, sugar cane, cassava roots, or chips. The molecular Structure of polylactide is shown in figure 2.
Polylactide is a biodegradable polymer and has similar characteristics as polystyrene (PS), polyethylene (PE), or polypropylene (PP). It can be manufactured by the same equipment used to produce petrochemical based industry plastic[12]. This method of fabrication is relatively cost-efficient. Among a variety of bioplastics, Polylactide has a second huge quantity of bioplastic production. PLA can be soluble in hot benzene, chlorinated solvents, dioxane, and tetrahydron[13,14]. The properties of polylactide are presented in table 1.

Table 1. Polylactide’s properties.

| Property                  | Value                        |
|---------------------------|------------------------------|
| Melting temperature       | 157ºC to 170ºC               |
| Injection molding temperature | 178ºC to 240ºC            |
| Heat deflection temperature | 49ºC to 52ºC at 4.6 MPa     |
| Shrink rate               | 0.37 to 0.41%               |
| Flexural strength         | 48 to 110 MPa               |
| Tensile Strength          | 61 to 66 MPa                |

2. Fabrication of fiber-reinforced plastic composites (FRPCs)
FRPCs are mainly fabricated using the production process of extrusion, injection, thermoforming process, or by compression molding methods, and the details are given below.
2.1 Extrusion method: Two types of extrusion methods available; single screw, and twin screw. The schematic set-up of the Polymer Extrusion device is shown in figure 3.
For pure polymers, single screw extrusion is mostly used. Twin-screw extrusion is used if it is made of distinct materials. As part of a large extrusion system, single screw extruders can be used or used as a stand-alone extrusion machine. Twin-screw extruders are suitable for mixing, compounding, and processing viscous materials. Twin-screw extrusion has consistency in production and control of product quality as the process ensures uniform mixing.
In an extruder, a fiber and plastic mixture is fed into the hopper, then they pass through the heating barrel pushed by the rotating screw; during heating, the barrel softens and melts the plastic; the screw supplies the mixture continuously. Melted plastic goes through the die, take the intended shape. The product is then cooled down and drawn out in the form of a pellet, thin sheets, or granules for further molding. The process of extrusion is suitable to produce composite filaments or pellets for further processing[15].
2.2 Injection Molding: In injection molding, the schematic set-up of which is shown in figure 4, the molding compound is poured in a hopper and the same is fed to an extruder. The plastic is pushed by the extruder screw into the heating chamber in which the molding compound is heated, changing it into a liquid state; then, at high pressure, it is injected into the mold of the desired shape. Once the mold is filled, the part is allowed to cool; the mold opens and the final object comes out. Injection molding is a
suitable manufacturing process for mass production in large volumes, but it is confined to short fibers[16,17].

2.3 Compression molding: in this process pressure and heat are applied in molding the material to the desired shape, the schematic set-up of which is shown in figure 5. The molds are heated to a preset heat-pressure cycle; a preform/charge is placed in the cavity of the matched mold in the open position; then by bringing two halves of the mold together, the pressure is applied in making the resin fill the mold cavity; heated under a pressure, mold that cures the resin, the composite component is then ejected from the mold[18].

2.4 Thermoforming: Thermoforming is the process where the thermoplastic sheet is heated and deforms into the desired shape, the schematic set-up of which is shown in figure 6.

Process:
- Heating plastic sheet to the temperature wherein softens
- Forming or converting soft sheet into desired die shape by either air pressure, vacuum, or mechanical power
- Cooling the finished part and trimming excess plastic[18].

Eco-composites are the one which has environmental and ecological advantages in comparison with traditional composites. In eco-composites, natural fibers are reinforced in the biodegradable polymer matrix. A variety of biodegradable polymers are available these days[19]. The objective of the present article is to review the literature on natural fiber reinforced polylactide polymers and their properties.
3. Mechanical properties of FRPCs
The properties of natural fiber reinforced polylactide composites (NFRPCs) are depending on several parameters such as types of fiber used, fiber orientation, fiber volume fraction, fabrication process, processing of polylactide polymer, fiber structure, source of fiber, moisture conditions, fiber aspect ratio, treatment of fiber and bonding.

The mechanical properties of the polylactide matrix can be improved by adding fibers since fibers have higher strength and stiffness values. The mechanical properties of a few selected fibers are mentioned in Table 2.

Several researchers experimented on the mechanical properties of FRPCs. Yu et al., carried out experiments on PLA/ramie fiber and PLA/jute fiber composites [21], Oksaman et al., on PLA/flax fiber composites [22], Plackett et. al, on PLA/jute fiber composites [23], Hu and Lim on PLA/Hemp fiber composites[24], Rajesh Gunti et al, on PLA/jute fiber, PLA/sisal fiber and PLA/elephant grass fiber composites[25], Bax and Mussig reviewed the impact and tensile properties of PLA/Cordenka and PLA/flax composites[26], Ochi, 2008 worked on PLA/kenaf fiber composites[27], Nam et al., on Coir fiber reinforced PLA composites[29].

| Fibers              | Specific mass in g/cm³ | Tensile Strength in MPa | Modulus of elasticity in GPa | Elongation at Break Percentage in % |
|---------------------|------------------------|--------------------------|-------------------------------|------------------------------------|
| Ramie               | 1.5                    | 500                      | 44                            | 2                                  |
| Jute                | 1.46                   | 400 to 800               | 10 to 30                      | 1.8                                |
| Flax                | 1.4                    | 88 to 1500               | 60 to 80                      | 1.2 to 1.6                         |
| Hemp                | 1.48                   | 550 to 900               | 70                            | 1.6                                |
| Sisal               | 1.33                   | 600 to 700               | 38                            | 2.3                                |
| Elephant grass      | 0.8175                 | 185 to 327               | 7.4                           | 3.23                               |
| Kenaf (Bast)        | 1.2                    | 295                      | -                             | 2.7 to 6.9                         |
| Coir                | 1.25                   | 220                      | 6                             | 15 to 25                           |

3.1 Tensile strength of fiber-reinforced PLA composites
The tensile strength of pure PLA is less compared to PLA composites as shown in figure 7 [21]. With the inclusion of either ramie fiber or jute fiber in different weight percentages to the PLA polymer, an increment in tensile strength was observed. But the tensile strength of composites decreased less than that of pure PLA when the addition of fiber is more than 30wt percent. This is due to poor fiber dissipation in the PLA polymer. The tensile strength of ramie fiber reinforced PLA composite is higher than that of jute fiber-reinforced composite since the strength of ramie fiber is more than jute fiber[21]. These results are in agreement with the findings of Rajesh et. al[25].

Further, efforts were made by Plackett et. al[23], to find the suitable heating temperature of molding of jute fiber-reinforced composites for getting maximum tensile strength. Under a heating temperature of 2100°C in a hot press, the tensile strength was increased more than two times that of the polylactide. When the temperature in the heating stage was increased to 2200°C, which is far above the recommended processing temperature for the polylactide, a statistically equivalent result was obtained.
Figure 7. Tensile strength of PLA/fiber composites [21].

The inclusion of flax fibers of 30 weight percent improved the tensile strength marginally; thereafter strength decreased. This was attributed to the poor adhesion of flax fiber with PLA polymer. For composites of PLA/40 weight percent flax fiber treated with triacetin, the tensile strength was reduced with the inclusion of triacetin content from 5 to 15 weight percent. This was anticipated due to the softening effect. The greater triacetin inclusion (15 percent) showed an adverse effect in flax fiber reinforced PLA composites [22,26].

Figure 8 shows the tensile strength of pure PLA which is 50 MPa as found by Oksman et al [22]. The inclusion of flax fibers of 30 weight percent improved the tensile strength marginally; thereafter strength decreased. This was attributed to the poor adhesion of flax fiber with PLA polymer. For composites of PLA/40 weight percent flax fiber treated with triacetin, the tensile strength was reduced with the inclusion of triacetin content from 5 to 15 weight percent. This was anticipated due to the softening effect. The greater triacetin inclusion (15 percent) showed an adverse effect in flax fiber reinforced PLA composites [22,26].
Figure 9 shows the tensile strength of pure PLA is lower than Hemp fiber reinforced PLA composites. The addition of hemp fiber to PLA helps in transferring the load from the matrix to the fiber resulting in increased tensile strength. When the volume percent of hemp fiber in PLA polymer composite is 40 percent, higher tensile strength was observed. But in PLA with 50 volume percent hemp fiber, the tensile strength slightly decreased.

PLA with 40 volume percent hemp fiber treated by 6 percent alkali, showed a greater tensile strength of 54.6 MPa compared to PLA with 50 volume percent hemp fiber treated with the same percent of alkali. This is possibly due to inadequate fiber wetting as the fiber volume fraction increases[24].

Figure 10 shows the tensile strength of PLA/ sisal fiber composites and PLA/ elephant grass fiber composites. It was observed that the strength value of both the composites is more than pure PLA. The tensile strength showed an increasing trend up to 20 percentage by wt of fiber addition; thereafter a declining trend was observed. The PLA/elephant grass composite showed superior strength than that of PLA/ jute fiber and PLA/ sisal fiber composites.

Treating all the types of fibers mentioned above with 5 percent alkaline and reinforcing the same with PLA increased the tensile strength further. The PLA/ 5 percent alkaline treated elephant grass fiber composite showed a superior tensile strength than other types of composites[25].
Treating the fiber with alkaline would interrupt the existing hydrogen bonding in the structure, resulting in roughening the surface; further, a small amount of lignin, wax, and oils covering the outer surface of the fiber is removed; all these put together results in enhanced interlocking between the matrix and the fiber and effective stress transfer. As a consequence, treated fiber-reinforced composites showed improved strength[30].

Bleaching with H2O2 removed the lignin without fiber oxidative damage. This treatment ensured compact, clean, and smooth fiber surfaces. Increased NaOH concentration during treatment resulted in the excessive deletion of non-cellulosic material, thus weakening the fiber. This resulted in decreased fiber strength. Further, it was suggested that the lignin amount was not significantly affected by strong NaOH concentrations, but it could contribute to the degradation of the extracted fibers.

\[\text{Figure 1.}\]

Figure 11. Tensile strength of untreated and 5 percent alkali (NaOH solution) treated coir/PLA composites[31].

Nam T H et. al[31]. Experimented assess the influence of adding coir fibers to PLA matrix. Increase presence of fiber improves the tensile strength up to 20 wt percent of fiber content; further addition of fiber did not improve the strength. 50 wt percent of coir presence deteriorated the situation. From figure 11 it is to be noted that the strength values of all the composite with different wt percent of fiber is less than pure PLA.

In order to improve the strength of composites in comparison with pure PLA authors treated the coir fiber with 5 percent alkali (NaOH solution) and then reinforced the same with PLA matrix. A marginal improvement in strength was observed over pure PLA up to 20 wt percent of fiber content, further, addition resulted in decreasing the strength of composite and presence of 50 percent by weight of coir fiber saw a steep decrease in strength.

Using kenaf fiber bundles, Kenaf / PLA biodegradable composites have been produced. Figure 12 reveals that tensile strengths increased linearly with an increase in fiber reinforcement up to 50 percent by weight of fiber content[27]. The tensile strength of PLA with 70% by wt of fiber content was observed to be 223 MPa. The tensile strength of kenaf fiber cultivated at the mean temperature of 30ºC was higher than that of fiber cultivated at the mean temperature of 22ºC. The kenaf cultivated at a mean temperature of 22ºC grew up to about 2000 mm, while that cultivated at a mean temperature of 30ºC by about 3650 mm; this means that the kenaf fibers which grew over longer length showed improved strength.
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Figure 12. Tensile strength of PLA/ kenaf fiber composites[27].

The addition of fiber in PLA showed a definite increase in tensile strength irrespective of the type of fiber used. The increase in strength was dependent on the type of fiber used as a reinforcement. The addition of elephant grass fiber improved the tensile strength of composite compared to all other types of fibers. Reinforcing PLA with kenaf fiber increased the strength substantially compared to all other types of fiber addition. The presence of 70 percent by wt of kenaf fiber in the composite increases the tensile strength up to 225Mpa which is more than the strength of many of the metals and their alloys.

3.2 Flexural strength of fiber-reinforced PLA composites

Figure 13. Flexural strength of PLA/fiber[25].

Figure 13 shows the flexural strength of pure PLA and PLA-based composites. From the results, it is observed that the flexural strength of PLA/ ramie fiber and PLA/ jute fiber composites is marginally high with pure PLA up to 30 wt percent of fiber loading; the strength falls with further increase in fiber content; in many cases, the strength decreased lower than that of pure PLA. This is attributed to the poor dispersion of fiber in PLA polymer[25,30].

The flexural strength of PLA/ sisal and PLA/ elephant grass reinforced composites increased with the fiber loading initially and then decreased when the fiber loading is more than 20 wt percentage [30]. Flexural strength was found to be maximum for PLA/ elephant grass composites with 20 wt percentage
reinforcement, both for treated and untreated fibers. The flexural strength of the PLA/ sisal and PLA/ elephant grass reinforced composites was higher in comparison with other types of composites.

Figure 14. Flexural strength of PLA/hemp fiber composites [24].

The flexural strength of PLA/ hemp fiber composites at different fiber volume fractions is shown in Figure 14 [24]. Flexural strength for PLA/untreated hemp fiber-reinforced composite does not improve with increasing fiber loading. This is due to the lack of fiber surface wetting by the PLA, resulting in poor bonding between the fiber and the matrix.

According to the rule of mixtures, the strength and modulus should increase with the increase of fiber volume fraction but results showed that this tendency is not followed. Flexural strength increased noticeably with alkali treating of fiber in composites. The maximum flexural strength was found to be 112.7 MPa obtained for alkali-treated hemp fiber/PLA composite with 40 percent by wt. of fiber presence; this value was comparably larger than that for PLA with 30 wt percent of chopped glass fiber composite and much larger than that of polypropylene reinforced with 30 wt percent of chopped glass fiber composite.

Figure 15 shows that flexural strength rises directly with increasing fiber loading in PLA/kenaf fiber composites up to 50 percent by weight [27]. With further increase in fiber content to 70 percent by weight showed only a marginal improvement in strength; anyway, the strength was found to be 254 MPa for this composite which is much higher compared to any other natural fiber reinforced PLA composites.

Figure 15. Flexural strength of PLA/kenaf fiber composites [27].
4. Conclusions
Considerable research is being carried out on FRPCs over the last few years because of their excellent properties. PLA is a fully biodegradable thermoplastic polymer, which has certain excellent properties. Several different types of fibers are employed as reinforcement with PLA to fabricate a variety of composites. Kenaf fibers are very strong amongst all the natural fibers; thus kenaf fiber reinforced PLA polymer composites showed the best mechanical properties.

There are certain drawbacks observed in natural fibers while used in PLA for making composites like poor interfacial bonding with polymer, absorbing moisture, low processing temperature of about 200°C, and inadequate dimensional stability. Surface treatment of fiber using NaOH removed the impurities from the surface of fiber making the surface rougher which helps in getting better mechanical interlocking with the matrix resulting in improvement in properties, but there is an increase in the final product cost.

FRPCs have a bright future, as PLA is a fully biodegradable thermoplastic polymer. Future researchers could look for enhancing the mechanical properties of FRPCs. Also in view of the environmental consideration, the emphasis should be on replacing synthetic fibers with natural fibers in PLA composites.

The objective of the present article is to throw light on whether PLA can be used as a matrix in which bio-fibers can be reinforced to fabricate the composite structure. Many of the works as briefed above suggest that composites with renewable fiber reinforcement show qualities comparable to those of synthetic fiber

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