Recent Researches In Polylactic Acid Reinforced With Natural Fiber Composites– A Literature Review

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Abstract. Growing interest in the development of natural fiber composites have been witnessed over a period of last decade which is attributed to low cost, availability, eco friendliness. The large scale production and use of natural fiber reinforced composites is limited due to degradation by micro organisms, water absorption, low strength. These issues are addressed by using various fiber modification techniques such as biological, physical or chemical modifications. This paper addresses the recent developments in surface modifications of the NFRP composites (Natural Fiber Reinforced Polymer). The various effects of such modifications have also been discussed.

Keywords: Natural fiber composites, Surface treatment, Mercerization, Silane treatment, Corona treatment

1. Introduction
Environmental consciousness and community interests have become the focus point of the researchers for over a decade and this trend is an attribute to the survival of the human species [1]. New regulations by the governments have forced the development of bio-composite materials in the markets. A bio-composite material can be established as a material in which one of the constituents is derived from a natural resource. When bio-composites are reinforced with a natural fiber it endows the material with an inclusive benefit of biological, mechanical and physical properties [2]. Synthetic fibers such as Kevlar, glass, carbon etc are widely used as the fibers for reinforcements in composite with petroleum based matrices owing to its excellent physical, mechanical and corrosion properties[3]. Even though the synthetic fibers have better performance in terms of physical and mechanical properties it has an inherent disadvantage of non- renewability, non-recyclability has prodded the scientific community to search for better alternatives. Recent researches suggest the use of natural fibers as a suitable substitute to the synthetic fibers. A recent study indicated an energy saving of around 50,000 MJ for a substitution 65% of hemp fibers for the composite made with 30% glass fiber [4]. This has led to the development of natural fiber reinforced composites for structural and engineering applications.

Natural fiber reinforced polymer composites (NFPC) are an emerging field of engineering which emphasises impact of petroleum based materials on the environment.
NFPCs with resins from renewable agricultural products reinforced with biofibers can be considered as fully green composites[6]. The bio fibers can be sub classified into non-wood fibers and wood fibers based on their origins [7]. Due to the increased cellulose content, crystalinity and lesser weight the non-wood fibers (natural fibers) are superior in physical and mechanical properties. Natural fibers are derived from plants such as oil palm, sisal, jute, flax, hemp etc. or from animals such as sheep, human hair, feather etc [8]–[10]. Even though the natural fibers are bestowed with advantages such as low self-weight, resistance to corrosion, fatigue, low cost, high specific strength and modulus the natural fiber has few drawbacks such as high moisture, high anisotropy, low compatibility with the matrix [11]. Fiber modification methods such as physical, chemical, biological treatments are a few solutions which can be used to address the above mentioned issues of natural fiber composites[12]. Much research has been delved into the significant progress in the laboratory scale modification and production of natural fibers. The latest environmental issues have pushed our requirements to find better alternatives which can replace the conventional natural fiber reinforced composites. Figure 1 shows the world production of natural fibers in terms of $10^3$Tonnes[5].

1.1. Natural fibers
The interest in industrial composites began to rise in the early 1960s during which glass fibers were used as reinforcements for tough rigid resin matrices[13]. The major advantage of this method was that these composites could easily be incorporated in to a scalable production. But for past two decades the industries have given a major interest in developing natural fiber composites due to its lower weight, comparatively low price, ecological motivations, requirement of renewable resources as an alternative for glass fibers[14]. Natural fibers can be classified into animal fibers and Plant fiber. The animal fibers can be further classified into avian fiber, silk fiber and animal hair. The avian fiber is obtained from bird feathers whereas the silk fibers are obtained from the saliva of bugs/cocoons of insects[15]. The plant fibers can be classified into bast fibers, leaf fibers, seed fiber, grass fiber, core fiber, wood pulp fiber, root fiber and fruit fibers as shown in figure 2. The plant fibers are hydrophilic in nature due to which the composites show an increased amount of water absorption leading to creation of interfacial voids between matrix and the fibers[16]. This may lead to critical failure of the composite prematurely, but it can be overcome with proper pre-treatment of fibers. The structural organisation of a natural fiber is shown in figure 2 [17].
Extraction and separation of fiber are fundamental properties that affect chemical composition, structure, yield and quality of the fibers. Many different approaches are adopted for the extraction and separation of fibers from their individual sources such as dew retting, water retting, Mechanical retting, Enzymatic retting, Chemical retting etc. [18]. Optimum results in all perspectives are not achievable by a single method. The natural fibers are attached to each other through the hemicellulose and pectin, which are non-fibrous materials and individual fibers can be released by decomposing these non-fibrous materials. In dew retting process takes 2-10 weeks for completion, it is done by cutting the plant stems and distributing them in the field thus exposing it to the microorganisms that consume pectin surrounding it [19][20]. A drawback of this method is this method is depended on weather conditions, soil contaminations also adversely affects strength, consistency and quality of the fibers [20]. Water retting method takes a duration of 7-14 days for completion during which the fibers are submerged inside water [21]. While the fibers are submerged in water the anaerobic bacteria decomposes the pectin holding the fibers together. Even though it is capable of producing high quality fibers it requires large amount of water and high labour cost are some of the shortcomings of this method [22]. Mechanical Retting process uses a decorticator for the extraction of fibers. It takes 2-3 days for the fiber production yet the fast production rate is affected by high cost and low quality of the fibers. Enzymatic retting is the best method which provides best result with a very high production rate of 8-24 hours. This method can ensure high quality fibers at fast rates however the only short coming is the high cost of production [23], [24], [25].
2. Fiber content in fibers and its effect on tensile strength

Single fiber properties are dependent on the crystalline content, orientation, thickness of the cell wall, size, shape etc [26]. Plant fibers, unlike synthetic fibers, are normally rigid and are resistant to fracture during processing. Some plant fibers have specific strength and stiffness similar to that of a glass fiber[27]. The characteristic properties of natural fibers are shown in table 1.

**Table 1. Characteristics of natural fibers**

| Pros                  | Cons                                      |
|-----------------------|-------------------------------------------|
| Good thermal insulation | Low impact resistance                     |
| Less weight           | High moisture absorption                   |
| Biodegradable         | Large variations in the properties of same fiber due different sourcing and processing methods. |
| Low energy consumption | Poor microbial resistance                 |

In the figure we can analyse that cellulose content plays an important role in the tensile strength of the fiber. A specimen of sisal fiber was treated with alkaline treatment for various durations and different levels of cellulose, hemicellulose and lignin was compared. Out of the tested samples the maximum tensile strength was obtained when lignin content and hemicellulose was minimal and cellulose content increased[28].
Figure 4. Corelation between composition of a fiber and tensile strength

The effect of fiber composition with varying amounts of lignin and cellulose is briefly discussed in table 3. As described in the table, when the lignin content in the fiber is higher, the fiber becomes more flexible and has an increased shear stress resistance. But in fibers with higher cellulose content, the fiber exhibits superior tensile strength and hardness. The chemical compositions of various fibers most recently used in the composite industries are shown in table 4. Hydrophilic nature is an inherent property of natural fibers at normal environmental conditions; the amount of moisture content present in a fiber adversely affects the properties of the fiber[26]. Figure 5 illustrates the moisture content in widely used fibers at 65% relative humidity and temperature of 21°C[29],[30]. Natural fiber itself can be considered as a polymeric composite with constituents of cellulose, lignin, and hemi-cellulose. Cellulose is a major building block in wood as well as in plants as it makes up to 40% to 50% of the dry matter. The size of the cellulose molecule is approximately around 5μm which is larger than that of the areas of crystallinity. This shows that the cellulose molecule can pass through areas of crystalline as well as amorphous regions. Cellulose I is a naturally occurring molecule but cellulose II is more stable thermodynamically[19], [31]. Cellulose II can be produced from cellulose I by mercerisation process (Alkaline treatment). Cellulose III and IIII can be produced by treatment of fibers with ammonia [32].

Table 2. Comparison of mechanical properties of different fibers[2],[5],[33],[34]

| Natural fiber          | Density (kg/m³) | Tensile strength (MPa) | Elongation at break (%) | Youngs modulus(GPa) |
|------------------------|-----------------|------------------------|-------------------------|--------------------|
| Jute(Corchorus capsularis) | 1440-1520       | 393.0–773.0            | 1.5–1.8                 | 13.0–26.5          |
| Flax(Linum usitatissimum L.) | 1420-1520       | 345.0–1100.0           | 1.3–10.0                | 27.6               |
| Hemp(Cannabis sativa)     | 1470-1520       | 550.0–900.0            | 1.6                     | 30.0–70.0          |
| Kenaf(Hibiscus cannabinus) | 1435-1500       | 295.0–1191.0           | 3.5                     | 53.0               |
| Ramie(Boehmeria nivea)    | 1450-1550       | 348.0–938.0            | 1.2–8.0                 | 44.0–128.0         |
| Cotton(Gossypium sp.)     | 1520-1560       | 264.0–800.0            | 7.0–8.0                 | 5.5–12.6           |
| Milkweed (Calotropis gigantea) | 1480           | 381.0                  | 2.1                     | 8.2                |
| Coir (Cocos nucifera)     | 1150-1220       | 131.0–175.0            | 15.0–25.0               | 4.0–6.0            |
| Kapal (Ceiba pentandra)   | 1320            | 90.0–95.0              | 1.8–4.2                 | 4.0                |
| Sisal (Agave sisalana)    | 1400-1450       | 500.0–800.0            | 2.0–25.0                | 9.4–22.0           |
| Pineapple (Ananas comosus) | 1440           | 170.0–1627.0           | 2.4                     | 60.0–82.0          |
Agave (*Agave americana* L.) 1360 430.0–580.0 3.0–4.7 13.2

Banana (*Musa sepientum*) 1350 529.0–914.0 3.0 27.0–32.0

Sugarcane Bagasse (*Saccharum officinarum*) 1250 20.0–290.0 1.1 17.0

Bamboo (*Bambusoideae*) 600-1100 140.0–230.0 - 11.0–17.0

Snake grass fiber (*Sansevieria cylindrica*) 887 287-545 2.87 2.5-7.5

Silk (*Bombyx mori*) 1300 650.0–750.0 18.0–20.0 16

Wool (*Ovis aries*) 1300 120.0–174.0 25.0–35.0 2.3-3.4

Hemicellulose even though is the second most abundant polymer only next to cellulose is entirely different from cellulose and it takes up to 25-40% of the dry mass. [35]. The main content of hemicellulose is known as aldoses and it is linked in linear or branched arrangement. Lignin consists of a large number of phenyl propane units forming a highly complex amorphous molecule.

**Table 3. Effect of fiber composition on the natural fibers**

| Content | Effect |
|---------|--------|
| Lignin rich fiber (eg coir-45% lignin, kenaf 12% lignin) | Increased flexibility and higher maximum deformation. |
| Cellulose rich fiber (eg- cotton 90% cellulose, pineapple 70% cellulose) | Increased hardness of the fiber, higher elastic modulus (E), superior tensile strength. |

**Table 4. Chemical composition of most widely used plant fibers**

| Natural fiber | Cellulose (wt %) | Hemicellulose (wt %) | Lignin (wt %) | Wax (wt %) |
|---------------|------------------|----------------------|---------------|------------|
| Jute          | 71.5             | 20.4                 | 13            | 0.5        |
| Flax          | 71               | 20.6                 | 2.2           | 1.7        |
| Hemp          | 71.4             | 22.4                 | 5.7           | 0.8        |
| Kenaf         | 57               | 21.5                 | 19            | -          |
| Ramie         | 76.2             | 16.7                 | 0.7           | 0.3        |
| Cotton        | 91               | 5.7                  | -             | 0.6        |
| Milkweed      | 55               | 24                   | 18            | 2          |
| Coir          | 43               | 0.3                  | 45            | -          |
| Kapok         | 35               | 32                   | 21            | -          |
| Sisal         | 78               | 14.2                 | 11            | 2          |
| Pineapple     | 82               | -                    | 12            | -          |
| Agave         | 68.4             | 4.9                  | 4.9           | 0.3        |
| Banana        | 64               | 6                    | 5             | -          |
| Sugarcane Bagasse | 55.2     | 16.8                | 25.3          | -          |
| Bamboo        | 43               | 30                   | 31            | -          |

Lignin has a 3D polymeric structure unlike cellulose and is not affected by hydrolysis. Therefore it is speculated that lignin is hard to be disintegrated into finer molecules. Hence extraction of lignin is done by Alcel process in which the fiber is treated at high temperature and pressure with aqueous
ethanol[36]. For increased yield of lignin treatment with alkali and subsequent enzymatic hydrolysis has been adopted[37].

3. Effect of fiber treatments- Physical, chemical, Biological Treatment

Natural fibers are inherently hydrophilic and polar in nature due to which it has very high moisture absorption rate. Environmental conditions like rain, soil conditions, sun, and amount of water are also factors which govern the quality of fibers[26] as a result of which the performance and properties vary in different harvesting seasons. Low thermal stability of natural fibers is also another factor that hinders its use in structural applications[30]. These shortcomings of natural fibers can be overcome by pre-treatment methods such as physical, chemical or biological treatment.

![Figure 5. Illustrations of moisture content in fibers][29],[30]

3.1. Physical modifications

Some of the inherent properties of the natural fibers such as poor compatibility with matrix, moisture absorption leading to reduced physical properties are disadvantageous prospects for engineering composite. Thus pre-treatment strategies have come into play to alleviate these issues. Researches have indicated that the mechanical adhesion between the fiber and the matrix can be enhanced with the help of physical modification without changing the chemical properties. The various physical modifications done on natural fibers are discussed in table 5.

3.2. Chemical Modifications

In order to augment the adhesion between the reinforcements and the matrix treatment of fibers with chemicals has shown definite results. The effect of chemical treatment on the Natural Fiber Reinforced Composites (NFPCs) has been extensively studied. Modifications in natural fibers can improve its adhesion with various matrices. It can also lead to producing very good strength and stiffness with a very strong interface.

It also allows better efficiency in transferring stress between the fiber and matrix. Extensive study showed that the researchers have reported that chemical treatment of fibers are more desirable than physical treatments such as corona treatment, plasma treatment etc. [38]. The research reports have shown that chemical treatments have increased adhesion between matrix and fiber meanwhile decreasing the hydrophilic nature of the fibers[5]. Alkaline (Mercerization), silane, acetylation, bezoylation, peroxide, maleated coupling, sodium chlorite, acetylation, acrylonitrile grafting, isocynate, steric acid, trizane, oleoyl chloride are the various chemical treatments applied on natural fibers.
Inclusion of various coupling agents also has shown significant improvement to the mechanical properties as it opens up more anhydride functional groups which enable more sites for chemical links. Owing to the addition of coupling agents in the matrix the water absorption rate also has been reported to rapidly decrease [39], [40], [41].

Table 5. Physical modifications and its effects on natural fiber.

| Sl no. | Treatment          | Modifications                                                                                     | References          |
|--------|--------------------|--------------------------------------------------------------------------------------------------|---------------------|
| 1.     | Plasma treatment   | It is one of the most commonly used method, it increases the surface roughness                      | [42]                |
| 2.     | Corona discharge   | It changes the surface energy of the fiber, increased compatibility with hydrophobic matrix, treatment on jute fiber increased mechanical properties, hemp fibers exhibited better tensile strength | [43], [44]         |
| 3.     | Steam explosion    | This method produces relatively short fibers and reduces the fiber strength                        | [45]                |
| 4.     | Ultra sound        | Effective in rinsing pollutantants and substances, it can increase the crystallinity index of the fibers. | [46]                |
| 5.     | UV light           | Wave length of UV used is 10 to 400 nm, it increases the polarity of the fibers, increases the wettability of fibers, increased strength of composite owing to the adhesion between matrix and the reinforcements. | [47], [48]         |
| 6.     | Ozone treatment    | It degrades lignin and solubilises hemicellulose content; it also increases the size and degree of crystallinity. The physical and chemical properties of the fiber decreased with respect to time. | [49], [50]         |
| 7.     | \(\gamma\) irradiation | It has been reported increase in the tensile strength, bending strength, tensile modulus, bending modulus of the jute/ PP composites and reduced water uptake than that of untreated fibers. | [51], [52]         |
| 8.     | Laser treatment    | The weight of the cellulose was unaffected but the lignin content in the bagasse fibers was reduced by 10% due to evaporation. The chemical modifications on the surface were similar to thermal degradation of fibers. | [53]                |
| 9.     | Fiber beating treatment | Increased surface area of the fibers, mechanical interlocking between the fibers is noted along with defibrillation. This method has reported 10% increase in strength of the fiber. | [54]                |
| 10.    | Heat treatment     | The fibers are treated to a temperature close to fiber degradation. Increase in crystalinity of the fibers was reported but for bamboo and hemp fibers the upper limit temperature was 140°C and 160°C above which the mechanical properties tend to decrease. | [55], [56]         |

Weaker components such as hemicellulose, pectin, lignin, wax and other oils which decrease the mechanical strength of the composite is also removed from the fiber surface. Treatment of fibers with silane coupling agents have shown to augment the fiber matrix interactions due to the formation of
strong chemical bonding as a result of which the mechanical properties of the composite is substantially increased [41]. The various chemical treatments are mentioned as following.

3.2.1 Alkaline treatment (Mercerization)

Alkaline treatment is considered as most economical and effective method that can modify the surface and alter the morphological properties of the fiber. This treatment initially helps in breaking down the fiber bundles and releases individual fibers. With further alkaline treatment the fiber fragmentation and disaggregation rate is increased [57]. As depicted in figure 6 fiber cell-O-Na structure is formed by breaking alkali sensitive OH groups from the fiber [39], [42] Due to this amount of OH group becomes exposed rendering the fiber to become more hydrophobic in nature [26]. Mercerization also decreases the amount of weaker compounds such as hemicellulose, pectin, and lignin and increases the amount of cellulose in the fiber due to which the crystalinity of the cellulose also increases as a result of which the stress transfer capacity of the fiber also increases [58]. H2O2 or NaOCl2 can be used as bleaching agents to purify the cellulose content and remove the organic waste from the fibers. It is important to note that optimum level of mercerization should be maintained to reduce the weakening and damage to the fibers, it also ensures that there is considerable reduction in diameter of the fiber leading to better adhesion with the matrix. [59] states that the optimum level of alkalization is 10%-30% concentration, higher concentrations of alkali leads to reduction in cellulose content which further declines the strength of the fiber [58],[59]. The comparison on mechanical properties of kenaf and pineapple fibers are shown in figure 7 and 8 [46].

\[
\text{Fiber-OH} + \text{NaOH} \rightarrow \text{Fiber-O'Na}^+ + \text{H}_2\text{O} \hspace{1cm} (1)
\]

![Figure 6. Mechanism of mercerization [59]](image)

| Comparison of fiber properties wrt treatments for Pineapple fiber |
|---------------------------------------------------------------|
| IFSS (MPa) | Diameter (μm) | tensile strength (MPa) |
|-------------|----------------|----------------------|
| untreated   | 1.7 290.61     | 78.8 47.8            |
| NaOH        | 432.01         | 1.93 50.6            |
| NaOH- silane| 424.63         | 2.35 42.4            |
| Silane      | 629.9          |                      |

![Figure 7. Comparison of fiber properties wrt treatments for Pineapple fiber](image)

3.2.2 Silane Treatment

Silane, represented as SiH4, is mostly used as a stabilization agent in composites [60]. Silane treatment is carried out using reagents such as Triethoxy silane [61],[62], Triethoxy octyl silane[63], pheniltrimethoxy silane[30], A174 (γ-methacryl oxypropyl trimethoxy silane)[64], A151 (vinyl...
triethoxysilane)[64]. The mechanism of silane coupling is by forming a chemical bond between matrix and fiber through a siloxane bridge which is created by the hydrolysis of alkoxy groups in the presence of moisture[46].

Figure 8. Comparison of fiber properties wrt treatments for Kenaf fiber

The effectiveness of silane treatment can be increased with prior alkali treatment as the number of reactive sites in the molecule will be increased drastically. In figure 7&8 we can clearly see that silane treatment has better mechanical properties as compared to that of other chemical treatment strategies. This is mainly due to the silanol bonding between the fiber and the matrix. Silane treatment has also augmented the resistance towards fiber swelling into the matrix as it is restricted by the molecular continuity formed across the interface of the hydrocarbon chain and the composite.[8] Discussed that in oil palm fiber composites that fibers when treated with silanes (pre-treated with NaOH) has better thermal conductivity than the acetylation treatment.[27] reported that the silane treated pineapple fiber had highest tensile and impact strength.[66] found that the silane treated ramie fiber had good tensile and impact strength when compared to other treatments.[67] silane treatment of abacca fibers showed improvement in storage modulus.

3.2.3. Acetylation treatment

A major advantage of acetylation is improved dimensional stability, durability and increased hydrophobic properties [68]. This treatment is carried out by using reagents like isopropenyl acetate [69], vinyl acetate [70], ketene gas [71], di-ketene liquid [72], cyclic/non cyclic anhydrides, acid chlorides, isocyanates, aldehydes, acetic anhydrides [73]. Of these reagents the most promising reagent is acetic anhydride. In acetylation treatment the cellular structure of the fibers are attached with acetyl groups with the help of acid catalysts. This is done by the preliminary soaking of the fibers in acetic acid and the secondary treatment is done with acetic anhydride for a period of 1-3 hours at elevated temperatures which increases the rate of chemical reaction [30]. During this reaction it should be noted that the simultaneous reaction of acetic acid and acetic anhydride does not occur. Acetylation is also known as esterification [74] due to the reaction of OH group and carboxylic groups with the fiber cellulose this reaction is represented by the equation 3 [75]. Many researches have reported that when the alkaline pre-treatment of the fibers have augmented physical and mechanical properties [76]. SEM analyses of the treated flax fibers have shown smoother surface
morphology and eradication of wax and cuticles from the surfaces [7]. Surface morphology of the treated fiber is altered by replacing polar hydroxyl groups present in the fiber with less polar acetyl groups due to this there is better compatibility with non-polar matrix and more hydrophobic. Acetylation treatment is effective in diminishing the rate of moisture absorption in natural fibers due to which there is an increase in dimensional stability and mechanical properties such as interfacial shear stress, tensile strength and flexural properties. Coir and oil palm reinforcement in polyester resin with 50% acetylation treatment showed 700% increase in tensile strength and 35% increase in tensile and flexural strength was shown in treated flax fiber/ polypropylene composite[77]. Heat insulation properties of oil palm/ phenolic composite have shown to improve in comparison with alkali and silane treatment.

\[
\text{Fiber- OH} + (\text{H}_2\text{C-CO})_2\text{O} \rightarrow \text{Fiber-O-CO-CH}_3+\text{CH}_3\text{COOH} \quad (3)
\]

3.2.4. Benzoylation

The principal purpose of this chemical treatment is to diminish the treated fibers’ hydrophilic property. Initially the fiber made to undergo a pre-treatment procedure during which the fiber is treated with alkali thus removing lignin, wax and covering oil [78]. Pretreatment with alkali activates the hydroxyl groups of fiber; the reagent used for benzoylation is benzoyl chloride which reacts with the cellulosic hydroxyl group and forms C6H5CO-(a functional group of benzoyl) in the fiber which improves the adhesion to the matrix. This chemical reaction is shown in equation 4[79]. Benzoylation treatment can increase the strength and thermal stability of the reinforcement fibers[12]. Treatment of sugar palm fiber with benzoyl chloride indicated an increase in tensile strength to 173.99 Mpa and a substantial decrease in the diameter of the fiber from 344.35um (untreated) to 480.33um (treated) was confirmed with SEM [80].

\[
\text{Fiber –O-Na} +\text{C}_6\text{H}_5\text{CH}_2\text{Cl} \rightarrow \text{Fiber –O- C}_6\text{H}_5\text{CH}_2, \text{NaCl} \quad (4)
\]

3.2.5. Peroxide treatment

Benzoyl peroxide, Methyl ethyl ketone peroxide (MEKP) [81], dicumyl peroxide[57] are the common reagents used for peroxide treatment. Researchers have reported improved adhesion with the matrix at the interface, reduced moisture absorption in fibers and better thermal stability in fibers. Pre-treatment with alkali reagent is a prerequisite for peroxide treatment in order to remove wax, hemicellulose, lignin components present in the reinforcement fiber[39]. The mechanism is initiated with the disintegration of organic peroxides into peroxide free radicals[57]. The peroxide free radicals thus produced reacts with the hydroxyl group of the reinforcement and matrix respectively. The presumed chemical reaction is described in equation 5 [57].

\[
\text{RȮ + cellulose-H} \rightarrow \text{R-OH- Cellulose} \quad (5)
\]

3.2.6. Maleated coupling treatment (MCT)

The main reagent used in this treatment is maleic anhydride and maleated polyethylene which modifies the surface morphology and polymer matrix [2]. Maleic coupling agents can help in enabling good compatibility between the matrix and fiber reinforcements. The hydrogen bonds or covalent bonds of cellulose-OH groups becomes more activated to react and thus enabling Maleic anhydride to be grafted on to the polymer. MCT is mainly used to increase the strength of the composite which is achieved by modifying the fiber morphology as well as polymer morphology of the matrix unlike in other surface treatment methods thus achieving better interfacial bonding [46]. The chemical reaction is represented as in equation 6. As depicted in the equation 6 [46] the MCT forms a coating on the surface of the fiber and decreases the hydrophilic tendency of the fibers due to exclusion of OH groups on the surface. Maleic anhydride (MA) is capable of C-C covalent bond with the polyer matrix and also of forming bonds with hydroxyl groups of cellulose fiber thus forming an effective and efficient bridge interface. MA treated fiber indicated better fiber resin adhesion by the failure analysis of the treated fibers which showed fracture by fiber tearing rather than the failure at the interface. Jute fiber/ polypropylene composite were prepared by compression moulding and it was reported that
dynamic storage modulus and young’s modulus improved with MCT [82]. Kenaf and polypropylene composite prepared by injection moulding reported decrease in the melting point in DSC test [83].

\[
\text{Fiber-OH} + C_4H_2O_3 + (C_3H_6)_n \rightarrow \text{Fiber-OH-C}_4\text{H}_2\text{O}_3-(C_3\text{H}_6)_n \quad (6)
\]

3.2.7. Sodium chlorite treatment

Bleaching of natural fibers can be subdivided into two types such as 1) Reduction bleaching 2) oxidation bleaching. Reductions bleaching are performed with reagents such as SO2, Na2S2O4, and are usually carried out on protein fibers[84]. Oxidation bleaching is done with reagents such as sodium hypochlorite, H2O2, NaClO2 (sodium chlorite) on cellulosic fibers. NaClO2 reduces moisture content and hydrophilic and increases the flexibility nature of the fibers[5]. The mechanism of NaClO2 can be explained with equation 7[84]; with the acidification of reagent it releases chloreric acid which liberates chlorine dioxide. The chlorine dioxide reduces the amount of lignin present in the fiber by creating compounds of lignin thus improving the adhesiveness of the fiber to matrix[85]. Random oriented flax fiber/ polyester composite produced by injection moulding reported increase in tensile strength with the sodium chlorite treatment. Sisal fiber/ polyester composite prepared by hand layup method reported increase in tensile, flexural, impact properties on treatment with sodium chlorite.

\[
\text{RCH}_2\text{OH} \xrightarrow{\text{Na}\text{ClO}_2^-,-\text{H}^+} \text{R} - \text{CH} = \text{O} \quad (7)
\]

3.2.8. Isocyanate treatment

Diphenyl methane diisocyanate, polymeric Methane diphenyl isocyanate, lysine based isocyanate are common reagents used for isoycante treatment. This treatment forms a strong covalent bond due to the reaction of its functional groups (-N=\text{C}=\text{O}) with OH groups present in the reinforcing fibers, thus the free OH groups become depleted as a result of which the hydrophobic property of the fiber increases[5]. The chemical reaction indicating the reaction is shown in equation 8 [57]. [86] reported better thermal stability with lysine based diisocyanate for bamboo fiber and PLA composite. Isocyanate treatment performed on sisal fiber/polyethylene composite produced by injection moulding has reported better tensile strength [86].

3.3. Biological modifications

Although chemical modifications being effective in increasing the mechanical and morphological properties of the natural fibers it has an inherent disadvantage of producing remnant materials which are harmful to the nature [87]. Therefore recent researchers have delved into biological modification methods to compensate for these short comings. Based on the mode of treatment the biological modifications can be classified into enzymatic treatment and fungal treatment [46].

3.3.1. Enzymatic treatment

It is a very expensive method for fiber treatment and it is utilized in pilot scales for the treatment of natural fibers and many researches have reported an increase in the interface strength of the treated fibers. It has the ability to selectively remove hemicellulose, pectin, lignin which are all hydrophilic in nature which in turn reduces the moisture absorption characteristics for the fibers [88]. Many researches have claimed that enzymatic treatment of reinforcing fibers increased the fiber matrix adhesion and also improved the mechanical properties in the final composite [46]. It utilises enzymes such as hydrolases and oxidoreductases for the specific removal of impurities. The major difference between chemical catalysis and enzymatic catalysis is the high specificity and high velocity of the reaction for efficient catalytic activity the temperature must be below 100oC, pressure of 1 bar, pH 4-8 should be maintained [46]. Amylase, cellulose, protease, pectinas, lipase/ esterase come under the classification of hydrolases and tyrosinase, laccase, peroxidase etc. are classified under oxidoreductases [89]. The common pectinas which are applied on the lignocellulosic fibers are commercial mixtures of different enzymes such as viscozyme, flaxzyzze, pectinex AR [90]. After the treatment of the fiber with enzymes it becomes soft to touch, improved wettability, dyeability, surface texture improvement without deterioration in the fiber.
Laccase is an oxidoreductase enzyme which has been reported to show best results in removing lignin from natural fibers without much damage to the fibers [88]. [91] claimed that Laccase is capable of reducing the lignin content in the cellulosic fibers form 35% to 24% when used in conjunction with natural phenols like acetosyringor, syringe aldehyde has indicated increase in antimicrobial properties. Enzymatic treatment on jute fiber indicated deterioration in toughness and flexural strength of the fiber [46]. Even though the enzymatic treatment tends to be efficient in removing lignin without any damage to the fibers a major drawback is its cost of operation, which can also be reduced with recycling the enzymes after each treatment[92].

3.3.2. Fungal treatment
Natural fiber treatment with fungal treatment has paved an efficient and innovative strategy to reduce the effect of chemical treatment on the environment. It enhances the properties of the natural fibers by the reaction of specific enzymes with non cellulosic components and lignin to form a soluble substrate which is washed away by an aqueous medium. The absence of non cellulosic components increase the hydrophobic characteristics of the fiber and reducing the moisture absorption rate in the composites. It is capable of removing unwanted cellulosic components such as lignin, pectin, hemicellulose, wax, etc. schizophyllum commune commonly known as white rot fungi can remove lignin and increase the solubility of hemicellulose by releasing extracellular oxidases onto the surface of natural fibers. Fungal treatment is capable of roughening surface morphology by producing a hyphae which absorbs the non cellulosic components as a result of which fine holes are produced on the surface of the fibers and thus the interlocking between the fiber and matrix is increased.

Fungal treatment performed on kenaf fiber reinforced composites showed 91.3% decrease in the amount of pectin. [93] reported that fungal treatment of on hemp fiber/ polypropylene composite prepared by injection moulding increased tensile strength by 22% abd 32% increase with additional alkaline treatment. 22% increase in the strength of composite was reported by [94] using fungal treatment on natural fibers. [95] reported that hemp fiber treated with ophiostoma ulmi fungi species contained enhanced fiber constituents in comparison to the untreated fibers and the byproducts of fiber treatment are water soluble.

4. Conclusion
Natural fiber composites have become a lucrative opportunity for several industries due to the associated monetary benefits of using natural fiber reinforced composites as opposed to the synthetic fiber reinforced composites. Enventhough with many merits of natural fibers one of the impending issues of the NFRPs is its low compatibility with the matrix. Hence various surface modification techniques have come into play to take advantage of the matrix and fiber interfacial adhesion. The many methods described here such as biological treatment, physical treatment, chemical treatment are employed in augmenting the overall performance of the composites. The future of sustainable development in the engineering field relies on the inclusive and extensive usage of natural fibers in engineering composites. Thus polymer composites with surface modified natural fibers are a viable solution to ameliorate the effects of non biodegradable composites. The upcoming research pool suggest that biological and nano treatment of natural fibers may soon be beneficial in producing composites that are bio friendly and efficient in engineering structures.

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