

Manufacturing problems and Solutions of Natural Fiber Reinforced Composites

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Abstract-In the modern world, natural fiber reinforced polymer composites (NFRP) are gaining too much attention due to the lower manufacturing cost and favorable effects on the environment after use. Besides this, the processing problems of such materials always remain a challenge. The bio/natural fiber has some protective layers and coatings causing hindrance for good interactions with polymer for good performance characteristics. Although numerous processes are already developed for overcoming the processing parameters but the desired goals are still not achieved. This short review focuses on some useful properties and pre-treatment of fibers used for the manufacturing of NFRP along with problems associated with effective remedial solutions. The viewers of this paper will get the basic knowledge and may select the area to study further in this field.

Keywords: Natural fiber, composites, treatment of fiber, hybridization of composites.

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I. INTRODUCTION

Talking about composites, NFRP are considered to be most influential material in the modern world as compared to synthetic materials due to their lower cost and improved sustainability [1]. This implies the significant use of such materials for the manufacturing of biodegradable material. On the other hand, the use of biomaterial is not new to mankind; numbers of historical archives confirmed their existence. The bricks used in Great Wall of China were made strong by mixing clay, reeds, twigs, branched willow and sands (biocomposite). Likewise, same is employed for natural polymers as paper and silk are known to humans since early history.

Since the development of synthetic materials in the 19th century, the application of synthetic material enhanced. However, the issues related to disposal of the end product, costly dumping or environmental concerns on the incineration reduced the feasibility [2]. The importance of natural fiber materials in the field of high-performance polymeric composites was realized since the start of 21st century. Furthermore, plastic production is based on utilizing oil-based substances which are nonrenewable.

For the application of composites, not requiring high properties, an alternative method can be employed to fulfill the needs by using such materials in the composites having minimum impact on the discussed issues. Material scientists used the term "eco-composites" or "green composites". These materials are based on the fact that many of the typical applications do not require excellent mechanical behavior; while fulfill the needs for some

secondary applications like panels, packaging, cases, gardening items [3, 4].

Based on the above statements, it is necessary to discuss here some materials. The first used material in this concern was recyclable polymers (polyolefin) with the addition of organic fillers. The use of such materials replaced many nonbiodegradable or nonrenewable polymers. Polyolefin polymers like polypropylene (PP) and polyethylene (PE) are most widely used among others. These fall under the category of thermoplastics. Besides this, another class of polymer matrices is also important, especially when concerning mechanical performances. Polyesters, epoxies, and phenol-based polymers fall into the category of thermosets. Thermosets have higher mechanical properties than the thermoplastics [5]. Organic fillers are preferred over inorganic fillers. Fillers from the wastes reduce the overall cost of the composite on one hand and have comparatively less negative effects on health issues of the workers, are easy to be burned/damped or incinerated and also have lower specific weights, allowing them to be effectively incorporated for good thermal and sound insulations [6-11].

Wood is the well-known material for filler application. It can easily be obtained from any saw dust mill. It can easily be crushed into required size depending upon intended application by simple grinding procedures. Besides, there is a variety of fibers that can be employed as filler. High mechanical performance is usually directly proportional with cellulose content and also on the alignment of microfibrils in length. Most of them fall into the category of stem fibers which include

cellulose-based fibers such as jute, sisal, flax, kenaf, ramie, hemp, etc.[9, 12]

This paper provides a brief overview of natural fiber based polymer composites along with problems in the processing of the fibers. The role of most important cellulose-based fibers like jute, kenaf, sisal, hemp as a filler in polymer matrices is discussed briefly to provide basic knowledge to the beginners in the field of NFRP.

II. DISCUSSION

Problems of NFRP composites processability

A. Rheological problems

The flow of resin is primarily important for commercial applications. As a general rule, increasing the fiber content in the resin increases the viscosity of polymer, leading towards the processability problems [13-16]. This problem is often observed in the processing of short fibers composites as compared to the sheet of fibers. By using a bit longer fibers, this issue can be resolved to some extent.

Rheology studies of the PE and PP composites with wood flour were investigated. Researchers found that viscosity is related to the filler content percentage and not on type of fiber being used [16]. However, it cannot be neglected that interaction of fibers with polymers has a strong relation with the viscosity of matrix. For instance, gelation of matrix material is also effected especially when dealing with thermosets. Addition of chemically modified fiber affects the gelation by catalyzing or retarding the reaction of reactive components during polymerization reaction or network formation [17]. For fiber based composites, gelation is a key factor to control the properties which is usually accompanied by good wetting of fibers before starting of gelation.

Addition of maleicanhydride adhesion promoter can significantly alter the rheological behavior of fibers with the matrix by forming filler-polymer bonds [18]. The addition of short fiber to a matrix at a certain level reaches to form clumps and balls usually called as neps. These entanglements limit the addition of fibers to cementitious composites [19]. It was also noted that the fibers aspect ratio is responsible for this [20] and the level of filler addition is dependent on it.

B. Fiber surface problems

All natural fibers are hygroscopic in nature i.e. water loving. At room temperature, natural fibers tend to absorb/adsorb a certain quantity of moisture from the environment. This moisture can negatively affect the interaction of fiber with the matrix. The equilibrium moisture contents of the natural fibers are shown in Table 1 [21]. The overall performance of fiber can be increased by making the hydrophobic surface. The hydrophobicity of fiber can be by certain treatments. The treatment with silane coupling is the most suitable option. This only changes the water absorption rate while moisture regain of the fiber does not change because the cell walls do not absorb these compounds. On the other hand, coating treatment changes the pore size thereby covering the hydroxyl groups thus decreasing water sorption[22]. Water contact angle of fibers is increased by these treatments which can be determined by dynamic contact angle test [23]. Polar component of the fiber decreases

while nonpolar component increases [24, 25]. Treated fibers show smooth surface pattern than untreated fibers[26]. Polar amino groups only slightly alter the moisture content because of highly hydrophilic properties of amino end group [24].

In order to overcome the moisture uptake of natural fiber, material scientists have tried to convert these hydrophilic bonds with some reactive agents known as Silane coupling agents [27]. Fig.1 shows a basic mechanism that how these compounds work.

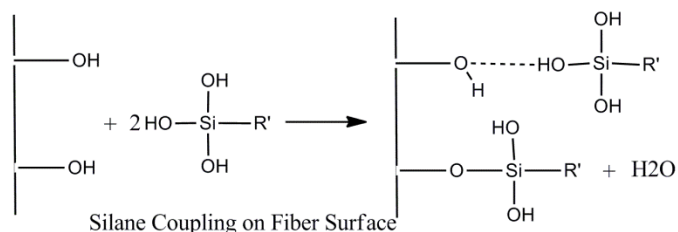


Fig.1. The reaction of silane coupling agents with Natural fiber [28].

Besides this, there are many techniques available to overcome the issues of hydrophilicity of fibers like acetylation, stearic acid treatment, benzoylation, TDI, Peroxide treatment, Anhydride treatment, permanganate treatment, Isocyanate treatment, Plasma treatment, gamma radiation [29]. Among the present methods, TDI, peroxide treatment and silane treatments give best properties related to mechanical performances while alkali and acetylation improve the thermal and dimension stability. The most important of all the available techniques will be discussed further in later sections.

C. Thermal stability problems

Some specific applications require processing of polymer at high temperatures. A good thermal stability is also desired to withstand the process parameters during flowing through the extruder at high temperatures. This is related more to thermosets. For example, curing of epoxy using aromatic or anhydride hardeners requires high heat treatment of composites to accomplish curing of networks. It is desirable for NFRP to have a curing degree above 90% to satisfy the application needs. For instance, the use of silane coupling agents is a suitable option available which alters the thermal response of fiber [30]. Nano particles addition in NFRP is a recent field of interest. Studies suggest that nano particles lower the interfacial thermal resistance and provide higher thermal conductivity [31].

D. Fiber strength problems

Fiber hydrolysis is the key factor responsible for the loss of strength. Highly acidic and basic treatment can distort the structure of the fiber. On the other hand, high heat treatment can result in depolymerization, hydrolysis, oxidation, dehydration, decarboxylation, and re-crystallization [32]. It is recommended to process or treat fibers in milder conditions. The tensile strength varies greatly when surface treatments are done therefore care must be taken to avoid the problem of loss of

strength which implies that optimization of every surface treatment/process uses tensile strength as the main feature. As a general rule strong conditions are susceptible for short period of times. Silane treatment causes a little effect on fiber tensile strength as reported [24]. Fiber tenacity loss was also reported by such treatment.

E. Fiber composition

Cellulose is the major component present in natural fiber and is considered to be most important when processing natural fiber. However, some special applications consider lignin also, especially when designing temperature resistance composites. Almost all stem based natural fibers are cellulose-based with small proportions of others substances. Table 1 shows material contents of some important stem fibers:

It can be seen from the components of fibers, the amount of proportions of cellulose, hemicelluloses, lignin and waxes for natural fibers varies greatly. This depends on the climatic conditions of the origin from where the fiber is grown. The geography is directly related to the composition and properties of the fiber [33]. This difference in properties will lead to different extent of treatments for removal of unwanted materials to improve its compatibility with matrices.

In this context, many researchers use Sodium hydroxide (NaOH) to make the fiber rough thereby reducing these unwanted contents and increase hydrogen bonding from the native cellulose structure [34, 35]. The later is of importance in the sense of hygroscopic properties of the fiber. Acrylic acid is also reported to be effective in this concern[35]. High-pressure steam is also used by many scientists to alter the surface characteristics by removing the unwanted materials and making it more favorable to polymer matrices[36]. These treatments are quite effective in reducing the amounts but exact treatment conditions cannot be made possible in the first attempt because of the variation in composition.

Table 1: Chemical Composition of some common natural fibers [5].

Fiber	Cellulose (%)	Hemicellulose (wt%)	Lignin (wt%)	Waxes (wt%)	Equilibrium moisture content (%)
Baggase	55.2	16.8	25.3	-	8.8
Curaua	73.6	9.9	7.5	-	-
Hemp	68	15	10	0.8	9
Jute	61-71	14-20	12-13	0.5	12
Rice Husk	35-45	19-25	20	14-17	-
Rice Straw	41-57	33	8-19	8-38	-
Ramie	68.6-76.2	13-16	0.6-0.7	0.3	9
Abaca	56-63	20-25	7-9	3	15
Coir	32-43	0.15-0.25	40-45	-	10
Oil Palm	65	-	29	-	-

Pineapple	81	-	12.7	-	13
Flax	71	18.6-20.6	2.2	1.5	7
Bamboo	26-43	30	21-31	-	8.9
Kenaf	72	20.3	9	-	-

F. Fiber properties

NFRP properties depend on the properties of fiber; therefore, it is important to know individual fiber properties before using it in composites. The properties of fiber vary and it depends on its chemical composition, dimensions of cells, alignment of fibrils along the length, cellulosic structure and amount, physical attributes and mechanical properties. Table 3 shows the physical and mechanical attributes of some important natural fibers.

Fiber strength can be taken as an important property for use in an application. Usually, the high ratio of length to width is desired which is directly related to the maturity of the fiber. A mature fiber has a perfect ratio with fewer defects on its surface. The elastic behavior of fiber is related to the angle of microfibril along the axis of the fiber. The elastic modulus decreases when this angle is increased. It is also interesting to note that crystallinity of plant fiber is not constant. The crystallinity varies from plant to plant and within the same plant from root to stalk core.

These variations in characteristics of fiber may point out the users for good mixing of fibers before using it in composites.

Table 3 Physical and mechanical properties of natural fibers [5, 37, 38].

Fiber	Tensile Strength (MPa)	Youngs Modulus (GPa)	Elongation at break (%)	Density (g/cm ²)
Coir	175	4-6	30	1.2
Pineapple	400-627	1.44	14.5	0.8-1.6
Bamboo	140-230	11-17	-	0.6-1.1
Flax	345-1035	27.6	2.7-3.2	1.5
Jute	393-773	26.5	1.5-1.8	1.3
Kenaf	930	53	1.6	-
Sisal	511-635	9.4-22	2.0-2.5	1.5
Oil Palm	248	3.2	25	0.7-1.55
Ramie	560	24.5	2.5	1.5
Bagasse	290	17	-	1.25
Abaca	400	12	3-10	1.5
Curaua	500-1150	11.8	3.7-4.3	1.4
Oil Palm	248	3.2	25	0.7-1.55
Elephant grass	185	7.40	2.5	0.81

III. DETAILS OF EFFECTIVE TREATMENTS OF FIBER FOR COMPOSITES

The methods can be broadly classified as two different techniques. Physical methods include heat and without heat extension, corona treatment, plasma treatment and gamma radiation. These treatments do not change the chemical composition of fiber while changes the surface characteristics to a greater extent to enable mechanical bonding. Secondly, more versatile techniques are chemical modifications including

silane treatment, alkali treatment, acetylation, maleated coupling, enzyme treatment.

A. Corona treatment

Corona treatment is used for surface modification by oxidation activation. This process changes the surface energy of the fibers. In other words, the polarity of fiber increases by the formation of aldehyde groups on the surface.

Gassan and co-workers treated jute fibers with epoxy [39]. It was concluded by their research that corona treatment increases the overall polarity of the fibers.

Studies on hemp fiber and PP composites were carried by Ragoubi et al;[40]. They concluded higher tensile strength and Young modulus of the composite. SEM analysis revealed improvement in the fiber-matrix adhesion.

Pizzi et al; worked on the treatment time of corona treatment for hemp and flax fibers. It was found that only the treatment for 5 minutes will result in the maximum tensile breaking strength of the composite. After that, the strength value decreases rapidly [41].

As the fiber is three dimensional in nature, the effect of corona treatment is small. The tenacity of fiber has an indirect relation to the corona power as it decreases with increasing power.

B. Plasma treatment.

In connection with corona treatment, various forms of surface modification could be achieved by using plasma treatment. This treatment introduces cross-linking of the surface while energy could be increase or decrease and free radicals could be produced. Helium cold plasma treatment was done on flax fibers reinforced polyester composites. It was concluded that water permeation was decreased with the increased mechanical stiffness of composite. This was accompanied by good interfacial adhesion of fiber in matrix due to the formation of free radical groups on the surface of fibers[42].

The effect of roughness on the jute fiber due to etching caused by plasma power was observed by Sinha et al; the hydrophobicity of the fiber increases. The fibers were mixed with the polyester matrix for composites to enhance mechanical performance improvement. It was concluded that 10 min treatment of fiber results in 14% improvement in strength[43]. Jute fibers were also treated with radio frequency (RF) and low-frequency (LF) plasma powers by using O₂ for composite with the polyester matrix. It was concluded that interlaminar shear strength increases as a result of good adhesion between fibers and matrix. Improved tensile and flexural strength were also achieved with both plasma systems. RF treatment was found better than LF treatment using O₂ in Plasma system [44].

C. Gamma radiation

Gamma rays are an electromagnetic radiation produced by radioactive decay. The penetration of gamma rays through the material alters the fibers internal structure, therefore, the strength of fiber itself decreases. Haydaruzzaman and coworkers studied the effects of gamma radiation on jute fiber and PP composites. It was found that 500 krad treatment of both the fiber and matrix give best results of tensile strength, bending strength and impact strength[45].

D. Silane treatment

Silane treatment is used in many industrial applications as a coupling agent to improve adhesion between a fiber and polymeric matrix. The coupling reaction is related to the effect of two reactive surface groups. Alkoxy groups allow the coupling agent to bind with the –OH groups initially, then the alkyl groups increase the compatibility with organic compounds and the hydrophobic character of the surface. This will lead to an increase in the strength of the interface [46, 47]. Figure 3 shows the mechanism of silane treatment of fiber.

Kenaf and abaca fibers composites with polyester resin were made by using silane coupling agent to modify the surface of fibers. The results show an increase in storage modulus of the composites. *Tan delta* values were reduced as compared to original fiber which shows an increase in interfacial adhesion of fibers within the matrix[48].

Two different types of flax fibers (natural and pulp) were compared for three different types of coupling agents; Vinyl trimethoxy silanes, maleic anhydride, and maleic anhydride PP copolymer. The last one gave the best results in terms of mechanical properties for both types of fibers, while no changes were seen in the properties by the application of first two agents on the flax fibers[49].

Some studies also suggest for silane treatment in combination with alkali treatment [50]. Jute fibers were treated with oligomeric siloxane to prepare a polyester composite. It was concluded that treatment of only 1% concentration of such agent can increase the interfacial adhesion of alkali treated fiber thereby increasing tensile strength and flexural strength of the composites.

E. Alkaline treatment

This is the simplest of all methods available to improve the surface interaction of fiber with the matrix. Surface roughness is improved by chemical modification and cleanliness of fiber surface. Many material scientists use this procedure for improvement of NFRP composites.

Sisal fiber reinforced epoxy composites were made after alkali treatment of 0.5 N Solution. The results showed more rigid composites with less porosity. The compressive strength was enhanced [51].

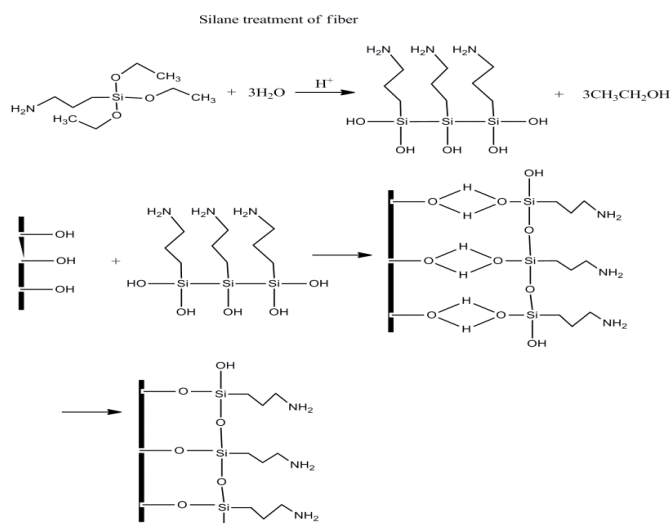


Figure 3 Silane treatment mechanism of natural fiber

Ramie fiber was treated with sodium hydroxide to assess the change of properties. It was discovered that tensile strength was improved by 18%. The fracture strain was also improved approximately three times to that of untreated fiber. This change was due to change of morphology and structure of fiber [52]. Jute fiber was subjected to sodium hydroxide treatment. The concentration of 5% solution increases tenacity, flexural strength, flexural modulus and laminar shear strength [53]. Other literature includes ramie fiber [54], sisal fiber [55], jute fibers [56] and coir fibers [57] composites with different polymer matrices. All of the treated fibers show an increase of mechanical properties of composites compared to untreated fiber composites. Figure 4 shows the mechanism of treatment of fiber with NaOH solution.

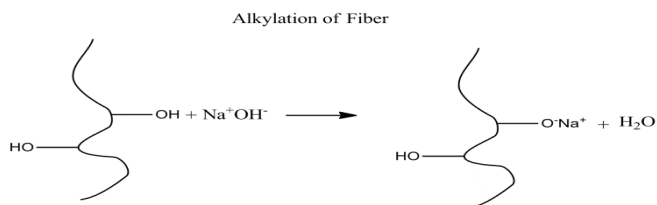


Figure 4 Alkali treatment mechanism of natural fiber

F. Acetylation

This process is an esterification method which reduces the hygroscopicity of natural fibers and increases the dimensional stability of NFRP. Acetylation changes the surface topography of natural fibers resulting in better free energy surfaces and stress transfer efficiency. This process also removes uncrystalline contents of fibers [58].

Flax fibers were modified by acetylation and reinforced with PP matrix. It was observed that the fibers moisture absorbency properties decrease due to the reduction of -OH groups of the fiber surface. This has also an effect on the degree of polymerization which decreases slowly until 18% acetylation. After 18% contents were acetylated, the polymerization degree

decreases rapidly because of the degradation of cellulose [59-62]. Figure 5 shows the mechanism of fiber treatment with acetylation.

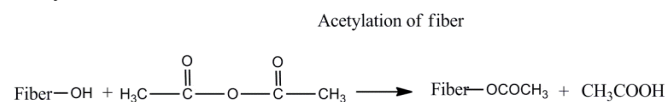


Figure 5. Acetylation of natural fiber

G. Maleated coupling

Maleated coupling agents are used to enhancing mechanical performance of NFRP. This type of grafting uses the free carbon-carbon unsaturation of the maleic anhydride group to form bonds to the polymer chain and leaving the anhydride group free to react. Due to the presence of the polar anhydride group on the olefin, this makes these polymers good couplers for natural fibers.

Several studies suggest the use of maleic anhydride PP as a coupling agent for NFRP. MAPP of 0.5% concentration and treatment time of 5 min was found sufficient to obtain maximum improvement of mechanical properties. Mechanical properties like Young's and flexural modulus, flexural strength, impact strength and hardness of composites were also improved after the treatment [63]. Figure 6 shows the mechanism of coupling agent treatment of natural fiber.

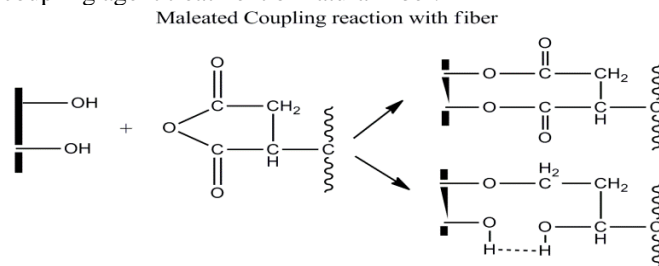


Figure 6 Mechanism of Maleic anhydride treatment of natural fiber

Fatigue and impact loadings of composites also get improvement for MAH-PP treated jute fiber reinforced composite. Fiber-matrix adhesion improves which results in higher dynamic modulus and also reduction in stiffness degradation [76]. Many types of composites are reported for a variety of different properties improvement for different fibers. Moisture absorption properties of flax fiber and PP composites [77], thermal and crystallization change of sisal fiber and PP composite [78], surface and water uptake behavior of flax, hemp reinforced PP composite [79], trans-crystallinity of jute fiber and PP composites [80] and dynamic mechanical properties of flax and hemp fiber with PP composites [81].

H. Biological treatment

The use of enzymes or bacteria to degrade non-cellulosic substances is the most suitable eco-friendly option available. Four kinds of enzymes are used for this purpose; pectin lyase, xylanase, laccase, and cellulose. After this treatment, the structure of fiber loosens and micro-fibril structure opens up with increased fineness for improved fiber/matrix interaction [82].

Hemp and flax fibers were treated with enzymes for the production of the composite with thermoplastics. Improvements in the mechanical properties of composites and morphological properties were enhanced[83]. Figure 7 shows how enzymes behave in the surface alteration of natural fiber.

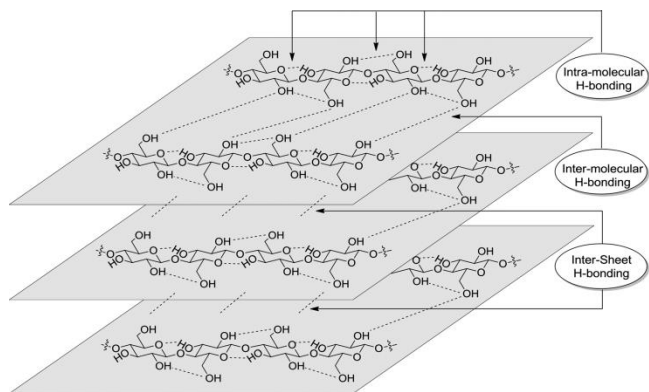


Figure 7 Effects of enzymes on natural fiber

IV. POLYMER MATRICES

A. Thermoplastics

The use of thermoplastic polymers for composite lies in their own that they are easily recyclable. There are various studies on using these polymers with fibers as filler. Studies suggest of using 40 to 70% filler contents. The most commonly used thermoplastics are PP, PE, polystyrene (PS) and polyvinyl chloride (PVC).

Table 4 shows some of the composites of thermoplastics with jute, kenaf, sisal and hemp pretreated with maleicanhydride coupling agent before use, along with mechanical properties reported.

B. Challenges in the application of NFRP.

The absorption of moisture from atmosphere limits the performance of such composites when exposed to outdoor environments. Color fading of surface and erosion, fungi accumulation, the growth of mold and loss in strength are all

Table 4 Mechanical properties of composites with different natural fibers reported in the literature

Polymer	Fiber	% of fiber	Tensile Strength MPa	Tensile Modulus GPa	Flexural Strength MPa	Flexural Modulus GPa	Impact Strength Izod I/Charpy C	Reference
Polypropylene	Jute	60	74	11	112	12	195i	[64]
	Hemp	40	52	4	86	4	210i	[65]
	Kenaf	30	46	5	58	4	39i	[66]
	Sisal	30	28	2.1	62	3.2	14i	[67]
HDPE	Jute	30	34.42	1.03	31.97	1.34		[68]
	Hemp	35	31.4	2.72			97.5i	[69]
	Kenaf	30	12	0.72	28	1.6		[70, 71]
LDPE	Sisal	30	44.3	1.24	59.2	2.856	68.4i	[72]
	Jute	30	27.7	1.6	10.7	1.82	8.9i	[73, 74]
	Hemp	40	16	0.9			14c	[75]

results due to the environment[84-87]. These problems are highest on the surface compared to the internal core of the composite as the surface is more saturated with moisture. Hydrophobization of fibers protects the composites potential damage associated with moisture absorption of fibers. Jute fiber treated with gamma-glycidoxypropyltrimethoxy silane and sisal fiber composites with epoxy results in reduced water uptake by composites [88, 89]. Coir and oil palm fibers modified with maleic anhydride polyester shows better results in soil burial test for one year. Mass loss by fungal decay was decreased due to the fact that less water was absorbed by composite surfaces [90, 91].

C. Hybridization

The incorporation of the third particle for specific property improvement of NFRP is the effective way and promising field. The key issue is to decrease the moisture uptake along with any other mechanical enhancement as reported in the literature. One study was done on kenaf and pineapple leaf fiber reinforced high-density polyethylene composites suggest an increase of tensile and flexural strength of pineapple leaf fiber while kenaf fiber increases impact strength and also reduces the water uptake of the composite [92]. Sisal fiber and polyester resin composite along with the presence of banana fiber result in increased tensile properties of the composite comparatively in the absence of banana fibers [93]. Many researchers have also used nanoparticles of different materials other than natural fibers to increase one or several properties of these NFRP but still, there are enough materials available that can be tried.

V. CONCLUSION

The above studies derive the following conclusions:

- NFRP can be made possible by suitable pretreatment of fibers. The most important thing is to remove the unwanted material naturally present in the fiber so that fiber/matrix interaction can be improved.
- The improved properties of the composite are the result of greater fiber and polymer attraction which is made possible by incorporating some coupling agents in the composites.

- The type of fiber is an important area to be considered for NFRP. Even with the same proportion of two different fibers for the same polymer, there seems to be a very high difference in properties even if the fibers are pretreated with the same coupling agent. The results are very different from the two composites. The difference in the fiber characteristics, cellulose content or structure may be considered as affecting the composite performance along with binding of the fibers to the resin.
- Radiation treatment (gamma and corona) alters the moisture uptake of fibers which eventually results in more linking points between fiber and matrix. As the constituents of each fiber vary, care must be taken in selecting the process parameters to avoid cellulose degradation. Optimization of process conditions is suggested before using this for NFRP manufacturing.
- Biological treatment is the most eco-friendly option and is recommended over the radiation treatment if the time for processing is not a key issue.
- Hybridization is an effective way of improving the composite properties more as compared to single fiber type. Especially, nanoparticles distribution along the matrix. Low specific gravity and higher surface area of nanoparticles, results in a good dispersion.
- Though NFRP has played an important role in replacing many synthetic made materials, further research is still required in improving the moisture resistance and fire retardancy to increase its application area.

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