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Effect of Nano-filler on the Manufacturing and Properties of Natural Fiber-based Composites: A Review

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ABSTRACT

Natural fiber reinforced polymer composite offers ecological safety towards a sustainable environment. Meanwhile, the deficiency of the poor interfacial bonding between fiber and matrix draws the attention of researchers to be sorted out. The use of inorganic nanofiller is considered as a possible solution to overcome the hurdle nowadays besides strengthening the composite properties. This article thoroughly reviews the use of inorganic nanofillers in natural fiber composites, covering different manufacturing processes and properties. Factors of various manufacturing techniques occupied for composite fabrication are investigated. Moreover, the influences of different nanofillers on mechanical, thermal, chemical, and physical properties of composites are discussed. In addition, Scanning Electron Microscopy (SEM) images of the bio composites are critically reviewed that usually exhibit the interfacial bonding and the fractures of the specimen. Furthermore, application of such natural fiber composites and the future investigation pathway in using inorganic nanofiller in composite are narrated.

Keywords: Natural Fiber-based Composites, Nanofiller, Manufacturing, Properties

1 Introduction

Polymers reinforced with natural fiber are nowadays a challenging and promising field in composite material research due to the rising need for sustainable and biodegradable materials. The policy of using petroleum-based synthetic polymers is under scrutiny in many countries because they are difficult to recycle and environmentally dangerous due to their non-degradability characteristics, which ultimately has a detrimental influence on nature and human health [1]. Natural fiber, on the other hand, is an excellent alternative to synthetic fiber because it is non-toxic, environment-friendly, less expensive, renewable, and recyclable, has superior damping qualities, and is widely available [2]-[4]. Performance of such composites is significantly influenced by the fiber properties (physical, thermal, mechanical, water absorption, etc.), fiber-matrix composition (volume percentage, stacking sequence, fiber orientation, etc.), and surface treatment of fiber [5], [6]. Natural fibers also have high tensile strength which offers their polymer composites good mechanical and thermal properties in applications including automobiles, load-bearing applications, packaging, and aerospace [7],[8]. A study found that using fiber-based composites reduced overall aircraft weight by 35% and had a positive effect on fuel efficiency and performance [9]. Due to these qualities, most of the components used by aircraft producers including Boeing, Dreamliner, and Airbus now employ composites [10]. Furthermore, it's been claimed that employing fiber composite parts can reduce vehicle weight and prices by 20 and 30 percent, respectively [11].

Three fundamental categories, including animal fiber, plant fiber, and mineral fibers, can be used to classify natural fiber [12]. Plant fibers like cotton, flax, hemp, and jute are formed of cellulose, while animal fibers such as hair, silk, and wool are made of proteins. In plant-based composite material, cellulose containing jute, sisal, banana, bamboo, rice, corn, hemp, kenaf, coir, flax, banana, aloe vera and pineapple are frequently used. Though having several benefits, such composites have several drawbacks, including poor interfacial bonding, dimensional instability, excessive moisture absorption due to the hydrophilicity of the fibers, brittle failure of the polymers, a tendency to aggregate during processing, and average strength [13]-[15]. It is troublesome for industrial and structural applications when the properties of a polymer composite deteriorate due to inadequate interfacial contact between natural fibers (hydrophilic) and a polymer (hydro-repellent) [16].

A variety of methods, including surface modification through chemical or enzymatic treatments, the addition of various interfacial additives or fillers, hybridization, coupling agent addition etc. are highly effective overcoming these difficulties [17]-[19]. The hybrid composite qualities are solely attributed to the fiber content, fiber length, and orientation, as well as the degree of fiber intermingling, fiber arrangement, and fiber-matrix bonding [20]. To adhere the bonding between fiber and matrix, surface treatment of the fiber is often made. Common surface treatments are alkali, silane, acetylation, benzoylation, peroxide, permanganate, and sodium chlorite [16], [21]-[25]. Alkaline treatment is a very well-known and straightforward technique for improving the adhesive properties of the fiber matrix. Commonly, sodium hydroxide (NaOH) is used in this process to change the cellulose structure of natural fibers, accelerating the breakdown and disaggregation of the fibers [26]. During the alkaline process, lignin, pectin, wax, and oil are removed from the fibers, leaving a smooth, clean surface and a higher elasticity [27]. The following chemical reaction happened [24],[28],[29].

$$Fiber - O^{-}H^{+} + Na^{+} - OH^{-} \rightarrow Fiber - O^{-}Na^{+} + H_2O \quad (1)$$

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Nanocomposites are designed by adding nanoscale fillers to a polymer matrix to fulfill the expanding needs for specific qualities in a variety of industrial and practical applications [30]. Nanocomposite materials have at least one phase with a dimension of 100 µm or less [20]. Mechanical, optical, electrical, magnetic, and thermal characteristics of nanomaterials are different from those of pure polymers [31]-[33]. Therefore, these particular properties can be improved by incorporating nanofillers, enabling a wider range of applications [28],[34],[35]. Numerous research is currently being conducted on different filler materials. Chowdary et al. [36] examined the influence of nano-silica on Sisal/Kevlar composites and found out that at 4% nano-silica addition, mechanical strength improved by a significant percentage. In the addition of nano-silica, thermal, flammability, and morphological characteristics of the composites are improved [37]. The incorporation of nano-clay considerably improved the fibers-matrix interface adhesion and compatibility of kenaf-coir hybrid composites [38]. Impact strength increases and water absorption percentages decrease with the increasing number of nanoparticles [39].

This study provides a comprehensive review of mechanical, thermal, and morphological studies on nanocomposites made from natural or bio fibers, various types of fabrication processes, and the application of the nanocomposites in different sectors.

2 Nanocomposites: Types and Properties

Nano-sized silica, zinc, alumina, titanium dioxide, calcium carbonate, lead oxide, silicon carbide, carbon black, nano clay, and various kinds of nanofibrils and cellulose nanocrystals are commonly incorporated in composite materials. They can be differentiated as organic and inorganic nanofillers. Different types of nanofillers that improve material characteristics without sacrificing density, toughness, or processability [12], [40] are depicted in Fig 1. Modern microscopic techniques such as SEM, TEM, STM, NMR, XPS, WAXS, AFM, DSC, and FTIS are commonly used to analyze nanoparticles [25], [41].

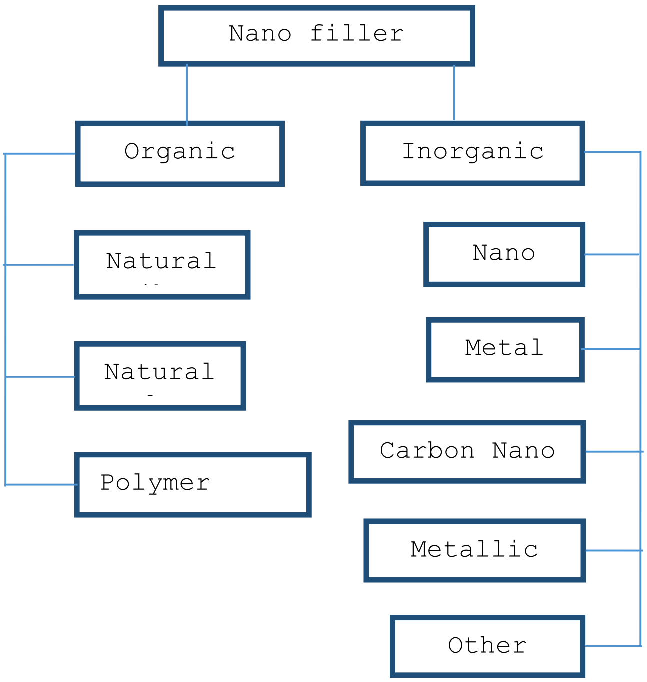

Fig 1 Different types of organic and inorganic filler material [42]

Nanoscale fillers have an extremely high surface-to-volume ratio due to their properties such as catalytic reactivity, electrical and chemical resistivity, etc., [43],[44]. Adding nano fillers provides a large interaction zone between them which involves several interaction mechanisms, particularly based on the type and nature of the filler and matrix used. The schematic interpretation of filler-matrix interaction is interpreted in Fig 2. Significant improvements in crystallinity, the formation process, the polymer chain's order, chemical properties and corrosion resistance are also addressed [45],[46]. At the nanoscale, quantum confinement, energy quantization, molecular mobility, and electromagnetic forces become more prominent. As a result of these processes, there will be an increase in intermolecular bonding, hydrogen bonding, van der Waals, hydrophobic effect, catalysis, magnetism, surface energy, and other effects. Based on the effects of hydrophobicity, catalysis, hydrogen bonding, surface energy, etc., nanotechnology and nanostructured materials have been developed [42]. Due to the very small number of nanoparticles introduced in comparison to the bulk phase, the material weight also decreases [14]-[16]. However, the incorporation of higher filler concentration may cause in more microscopic voids, eventually lowering the properties due to weaker bonding between the reinforcement and matrix [47],[48].

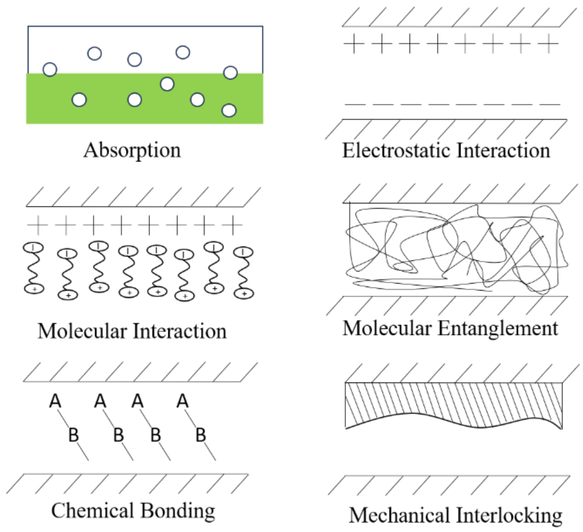

Fig 2 Schematic of the interaction mechanism at the fiber-matrix interface [49]

Aside from these, nanoscience and nanotechnology researchers have recently become interested in cellulose-based nanomaterials because of the abundance of renewable natural sources [50]. The terms nanocellulose and "nano-fibrillated cellulose" are usually used to describe cellulose nanofibers (CNF) and cellulose nanocrystals/whiskers (CNC). In the host polymer matrix, CNC exhibits a strong propensity for self-association, which is helpful for the development of load-bearing percolating structures [51]. Various techniques, including chemo-mechanical, grinding, cryo crushing, micro fluidization, and ultrasonication have been used to create CNFs from natural fibers [52]-[54] while CNCs are commonly produced using acid hydrolysis of cellulosic materials dispersed in water [55]. Notable properties of Poly Vinyl Alcohol (PVA) based nanocomposites reinforced with sugarcane bagasse nanocellulose investigated by Mandal et al. [56] and it was found that Crosslinked PVA and linear PVA nanocomposite exhibited the highest tensile strength at 5 wt.% and 7.5 wt.%

of nanocellulose respectively. Rosamah et al. [57] examined the impact of bamboo nanocellulose in kenaf fiber-polyester composite. It was found that the addition of 3% of nanofillers contributed to a strong bonding and increased wettability with the matrix, resulting in superior mechanical properties and thermal properties of the composite.

3 Manufacturing Techniques

In general, the manufacturing process of nanocomposites is like that of conventional polymer composites. Before compounding the matrix with the fiber, nanoparticles are usually mixed with the matrix using various stirrers as per the required proportion. Common compounding techniques are single/twin screw extruders, two- and three-roll mill ball machines, Brabender, Ragogna, and HAAKE mixers, k-mixers, mechanical and magnetic stirrers, and other common pieces of machinery are some of the prominent ones used to compound filler and polymer matrix [38],[58]-[62]. Nowadays, mixing matrices with nanomaterials frequently involves the use of mechanical stirrers and ultrasonic probes [38],[63]-[67]. To introduce specific nanofillers into the matrix and to prevent contamination, uniform dispersion of nanofillers is a crucial step in the manufacturing of nanocomposites [42].

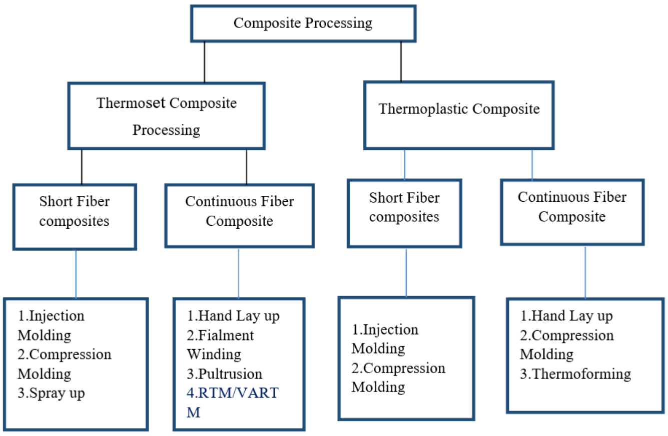

Fig 3 Various fabrication processes of composite

Fig 3 depicts the types of manufacturing techniques where nanoparticles can take place as fillers.

3.1 Hand lay-up

The Hand Lay-up technique can be used to manufacture hybrid composites using long natural fibers, although this method does not permit considerable fiber loading [68],[69]. Nanoparticles are inserted in the sample by mixing the particles with a matrix with an appropriate ratio. Samples are used to be cured at room temperature [70]. Modern technologies for curing composite parts include autoclaves, electron beams (E-beam), microwaves, X-rays, and ultraviolet (UV) light [71]. This approach is constrained by factors like the greater possibility of voids due to the uneven distribution of resin. Mixing of resin and the composition is crucial in this labor-intensive process [69]. The composite quality is significantly influenced by fiber loading. Additionally, this is influenced by the fibers' physical traits, such as the lumens in their intra-fiber gaps.

3.2 Compression Molding

A common method of producing high-volume natural fiber-reinforced polymer nanocomposite is compression molding [72]. Fabrication of natural fiber-reinforced nanocomposites typically involves preheating the fiber components, followed by compression at high pressure until solidification takes place [16],[73]. The hot and cold compression methods are two different ways to produce the composite by this technique. Using two flat plates, the mixture is compacted in hot compression before being heated to cure it. Consequently, in this process, both pressure and temperature are required at the same time, in contrast to cold compression curing, which occurs at room temperature and only uses pressure [69].

Less waste, excellent productivity, and high repeatability are all present together with low cycle time and cost [71],[74]-[76]. Because the components are placed carefully, there is relatively little fiber damage throughout the composite's production process. Long and short fibers can both be employed as reinforcement; however short natural fibers can be pre-combined with resin and nanofiller to lessen the shrinkage of the final specimen [72]. This method is primarily suitable for fabricating small parts [69]. Molding conditions, curing temperature, heating time, interfacial interaction between fiber and matrix, and cooling time are all different factors that should be considered. A careful consideration of temperature is required since, in general, there is a small gap between the processing temperature of a given matrix and the temperature at which fiber deterioration will take place [77]. The tensile strength of fibers is demonstrated to be reduced at temperatures between 150° C and 200° C, with a 10% decline in strength occurring in just 10 minutes [78]. However, depending on the sheet's thickness and the material used, these variables may change [79].

3.3 Resin Transfer Molding Process

Resin transfer molding (RTM) has recently gained a lot of popularity for fabricating high-performance thermosets. A mechanically clamped, stiff, closed mold is used in which long or weaved fibers are inserted. Then, at low pressure, the resin, nanoparticles, and other additives are pressed into the mold holding the preform, and then heat is applied to solidify them [80]. The items produced with this technique feature great surface finishing, good dimensional tolerances, minimal void content, cheap tooling costs, nearly no air entrapment, and no thermomechanical degradation [74],[81]. Major challenges to this method are the need for resin with low viscosity, variation in the composite part's thickness as a result of uneven compaction pressure, less productivity, and poor shrinkage quality [82],[83],[84]. Low viscous resins are commonly used in this process. Mold configuration, resin properties, resin preheated temperature, injection pressure, and fiber content all affect this technique [85],[86]. Due to lower degrees of fiber alignment, natural fiber composites are less compactable than glass fiber composites during this phase where the structure of the fibers, particularly the impact of lumen closing, influences the compaction [87]. However, a high injection pressure could result in the fiber preform being washed out of the mold and deformed. Premature resin gelation and short shot can be caused by an extremely high mold temperature. Each process variable is connected to the others and influences how final goods behave mechanically [88].

The RTM process known as VARI/VARTM is one of the new molding technologies that is expanding very fast. The main distinction between the two is that resin is injected under pressure in RTM rather than employing a vacuum and high heat or pressure [71]. The VARTM process creates a vacuum-tight seal. Typically, a "manifold" made up of several carefully placed ports and feed lines allows resin mixed with additives to enter the structure. A vacuum pump is typically used to remove the air [89],[90]. Air cavities can thus be minimized to the point that the resulting composite part has little open space, producing composite products with superior mechanical properties [91],[92]. A network of internal passages helps to wet out the fibers as it is pulled into the reinforcements by vacuum. For the impregnation of the fiber tows during the VARI process, the capillary effect of natural fibers was crucial [52]. With the use of VARTM's low-cost tooling, it is affordable to make large, complex parts with a larger fiber volume fraction in a single step [93]. They do, however, have several disadvantages, such as thickness variance, poor surface smoothness, installation challenges for supplemental equipment, such as sealant tape, porous peel ply, vacuum bag, distribution medium, and breather, non-reusability, high waste production, etc. [94],[95].

3.4 Spray Up Method

In this process the fiber can be uniformly encapsulated into the polymer matrix directly through spray drying [76]. The mold is initially prepared with mold release and gel coat. Then, using a handgun, resin, nanoparticles, and finely chopped fiber are sprayed into the mold. The fiber is then aired out and moistened using a roller and brushes. It is then possible to add a second layer made of wood, foam, or another core material. After that, the portion is dried, chilled, and taken out of the reusable mold [71]. This approach only provides one-sided surface finishing and uses low viscosity resin, which is not suited for high dimensional accuracy items [72],[75]. Before the specimen is completely cured, the sprayed fiber and resin combination is rolled to prevent bubbles and voids [96].

3.5 Pultrusion

Pultrusion is a continuous method for producing composites with unique cross-sections and long lengths [97]. It is the best approach for large-scale, swift, long, and consistent cross-sections of parts and continuous composite in any dimension [71],[75]. In this technique, warmed dies are used to force integrated continuous fibers through a heated resin-nano filler solution. Material is subsequently molded into the desired shape after passing through several forming guides. The finished shape is then cut to the correct length after cooling employing a cut-off saw [98]. Pulling is done to prevent fiber damage. As matrices, epoxies, polyester, phenol, and vinyl ester are frequently utilized. The advantages of this approach include stronger strength, better surface finishing, thin wall construction, a wide range of cross-sectional forms, less fiber damage and the potential for a high level of automation [75],[79]. The mechanical qualities are impacted by variables like die temperature, pulling rate, and fiber content. The qualities are found to increase with higher die temperatures, higher fiber contents, and lower pulling rates. Post-curing is also a useful tool for enhancing the characteristics [99].

3.6 Injection Molding

One of the quickest mass production methods for intricate parts with a range of sizes and forms and minimal labor costs is injection molding [100]. Injection molding is superior to other processes in several ways, including mass production, a shorter production cycle, and tighter tolerances for complicated products [101]. Through a feed hopper with a funnel-like shape and a rotating twin-screw extruder, the fibers-typically pellets containing chopped fibers, nanoparticle mixed with resin are fed individually into a heated compression barrel. Through thermal compression, the pellets are forced into closed mold cavities made of matching metals, which facilitates thoroughly transferring the stress from matrix to fiber. The polymer solidifies inside the mold, which is tightly pushed against injection pressure. After proper solidification, the mold is removed from the closed mold with the proper shape [72]. The resulting products have a high level of surface finish and outstanding dimensional precision, and this process works with both thermoset and thermoplastic composites. It is a very significant and productive technique for making everything from very small things like bottle tops to extremely huge automotive body sections [79]. The critical length of the fiber, fiber content, residual stress, mold temperature, injection pressure and cooling time are some significant parameters that need to be controlled [102],[103]. If the fiber exceeds the critical length and may risk breaking before the matrix fails. The modulus distribution of the composites is impacted by residual stress and fiber orientation [104].

3.7 Filament Winding

Although filament winding has received very little research so far, it is a viable alternative for producing symmetrical and convex-shaped components [105],[106]. It is a continuous fabrication technique that is inexpensive to fabricate highly automated, and reproducible.The technology for continuous thermosetting resin adhesive-impregnated synthetic fiber winding is quite advanced [107],[108]. The mandrel rotational rate and the way the fiber is fed into the mandrel both affect how the fiber is oriented in this procedure. In the x-direction, fibers that have been resin-impregnated are coiled and formed around a revolving mandrel. To regulate the resin in this process, nips and dies are employed. Inside the oven, the damaged area is repaired [75].This method is quick and economical. Discontinuation of natural fiber is the limitation of this process. To prepare continuous pre-preg belts, natural fibers can be twisted or weaved. Most frequently preferred are roving or stand fibers coupled with low-viscosity resins. The filament winding method, which is quick and affordable, is used to create pipelines, oxygen tanks, etc.

A summary of the various types of Fabrication processes with their benefits and limitations are listed in Table 1.

Table 1 Various manufacturing techniques along with their relative advantages and limitations

Process	Fiber type	Curing Temp. and pressure	Advantages	Limitations
Hand Lay-up	Long, short, woven	Room/High temp	Suitable for large components parts.	Heterogenous distribution of Resin.
Compression Molding	Long and short	High temperature and pressure for hot compression and room temp. for cold compression	Less cycle time, cost and waste, High productivity, and Reproducibility.	Suitable for small parts' production.
RTM	Long/woven	Room temp. and high pressure	Good surface finish, fewer voids, less waste, and less tooling cost.	Less productivity and low shrinkage quality.
VARI/VARTM	Long/woven	High heat and pressure	Excellent mechanical qualities, Fewer voids.	Bad surface finish, Thickness variation, High Waste.

Process	Fiber type	Curing Temp. and pressure	Advantages	Limitations
Spray Up	Short	Room temperature	Low cost, large product components.	One-sided surface finishing, difficulties in fiber volume and thickness control, unsuitable for high dimensional accuracy parts.
Pultrusion	Long and continuous	Controlled temperature	Higher strength, better surface finishing, thin wall structures, a large variety of cross-sectional shapes.	High tooling cost, restriction for a particular type of cross-section Components.
Injection Molding	Chopped	High injection pressure and high heat	Low labor cost, high quality of surface finish, and excellent dimensional accuracy.	High Initial cost, limitation in mold design.
Filament Winding	Long continuous, roving or stand fiber	Room temp. and pressure	Convenient for symmetrical and convex-shaped parts, comparatively economical.	Difficult to manufacture.

4 Mechanical Properties

It is reported by numerous researchers regarding the enhancement of mechanical properties by incorporating nano clay, nano silica, carbon nanotubes and other nano metal oxides as well as nano cellulose [109], [110]-[114]. In Jute fiber with nano clay, the tensile properties show the best results for 30 mm fiber length and nano clay of 5 wt.% [115]. For Nano clay infused banana fiber composite the compressive properties were enhanced by about 28%, Young's modulus increased by 25% and maximum strain energy shown in tensile mode increased from 19.27 J/m^2 to 71.83 J/m^2 when compared with pristine banana fiber composite [60]. The hybrid composite composed of 2 wt.% of MMT (montmorillonite-clay) with 5 wt.% of curauá fiber exhibited the best mechanical performance. Adding TiO_2 in epoxy polymer causes higher reluctance to impact effect during 5% volume fraction of TiO_2 and the property decreases with the increased portion of TiO_2. By increasing the value, this behavior was found to be the same as pristine epoxy when the volume percentage reaches 10% TiO_2 [116] . Effect of 2 wt.% to 3 wt.% nano clay on the mechanical properties of bamboo fiber-reinforced composite was found significant [117]. It is reported that the flexural performance of Napier grass-based composite is improved by incorporating of 3 wt.% of nano clay [118]. The tensile modulus retain rate was improved by 33.8% after the grafting of nano-clay into flax fiber-based epoxy [119]. Hybrid composite characterization with 10% epoxidized soybean oil and 1.5 wt.% nano clay causes retention of the real stiffness while improving toughness [120]. Nano clay inclusions of 5 wt.% in curauá fiber-based polyester composites reinforced enhance the tensile strength by 39.22% and flexural behavior by 25.43 % [121]. A hybrid composite of pineapple leaf fiber and polypropylene matrix with nano clay produces better mechanical response [122].

Alongside TiO_2, SiO_2, Al_2O_3, and ZnO metallic oxides, inorganic nano-sized powders are used as nano-fillers in different natural fiber nanocomposites. Stiffness behaviors are mostly enhanced by adding these particles with various polymer matrices. Reduction in crack growth, better interfacial bonding and lamination in basalt fiber-based composite was noticed when it was doped with SiO_2 powder. Adding TiO_2 in epoxy polymer causes higher reluctance to impact effect during 5% volume fraction of TiO_2 and the property decreases with the increased portion of TiO_2 [116]. By increasing the value, this behavior was found to be the same as pristine epoxy when the volume percentage reaches 10% TiO_2.When nano-SiO_2 of 5 wt.% was added to bagasse fiber-reinforced composites, MOE increased by 221.84 percent in comparison to pure HDPE. The flexural strength and modulus of rupture (MOR) of the composites improved as the nano-SiO_2 concentration of the composites increased [123]. When bagasse fibers and nano-SiO_2 (5 wt.%) are combined, the tensile modulus of pure HDPE is increased threefold [113].

Behzad Kord et al [124] examined the influence of including nano-SiO_2 on mechanical and physical properties of hemp fiber-based polypropylene composite. They concluded that the tensile strength and modulus improved up to 3% loading percentage of nano-infusion and then decreased. On the other hand, Singh et al. [125] found that in case of hemp-sisal-based epoxy hybrid composite with 0-4 wt.% of nano-silica inclusion, the addition of 2 wt.% nano-silica revealed higher tensile and impact strengths and 3 wt.% of nano-silica showed highest flexural properties. The addition of bamboo nanoparticles of concentration (0-5) wt.% into kenaf/unsaturated polyester natural composite results in the enhancement of mechanical characteristics for up to 3 wt.% of bamboo nano-infusion in the experiment conducted by Rosamah et al. [57]. Zhou et al. [126] added a small amount (0-1wt%) of CRN (carbonized ramosissima nanoparticles) in the bamboo fiber-reinforced epoxy composite. In this study, the tensile properties increased by 18.35% when CRN was 0.5 wt.% and shear strength was maximum for 0.25 wt.% which was increased by 38.96%. Ghalenho et al. [127] studied pine wood/polyethylene with a different weight percentage of TiO_2 nanoparticles (0%, 1%, 3%, and 5%) where the highest tensile and bending strength was achieved for 3 wt.% of TiO_2 through the addition of TiO_2 nanoparticles lowered the impact strength due to the higher brittleness. Sumesh et al. [128] examined the effect of nano-alumina up to 3% mass fraction on hybrid banana coir, hybrid sisal coir and hybrid sisal banana composites. The study showed improved results for mechanical properties but due to agglomeration, these characteristics tend to decrease at 5 wt% nano-addition.

The influences of nano-fillers (metal oxides, nano clay cellulosic fibers, carbon nanofibers) are shown in Table 2. In most cases, 1-5wt.% of nano clay and metal oxides were used and showed some prominent results. In the case of nano-graphene addition, positive results were achieved using 0-2% weight percentage. However, exceptions were found for nano-cellulose as it is sometimes used as reinforcing material for composites.

Table 2 Influence of various nano materials on the mechanical properties of natural fiber

Fiber	**Fiber content**	**Matrix**	**Manufacturing method**	**Nanofiller (%wt.)**	**Tensile strength (MPa)**	**Tensile modulus (G Pa)**	**Flexural Strength (MPa)**	**Flexural modulus (G Pa)**	**Impact Strength (kJ/mm^2)**	**Ref.**
Banana	40 vol%	Epoxy	Hand Layup	Na^+ Cloisite (3%)	173	10	88	8.102	—	[60]
Agave	—	epoxy	Vacuum Assisted Resin Infusion (VARI)	ZnO (2%)	23	2.8	—	—		[129]
Coccinia indica	—	Epoxy	Hand Layup and Compression molding	Cloisite 30B (3%)	38.29	—	92.77	—	67.25E-6	[130]
Curaua	5 wt.%	SBS	Melt blending	Cloisite 10A (2%)	7.8 ± 0.2	4.8 ± 0.2E-3 —	—	—	2	[121]
Vakka	44 wt.%	Polyester	Hand layup	Nanoclay	94.75	2.486	145.67	145.67	—	[110]
Ginger	—	Polyvinylidene fluoride	Sol-gel technique	Nano-silica (0-1%)	8-13	—	—	—		[131]
Jute fabric mat	41 g	Polyester	Compression molding	MMT K10/Egg shell powder (±1.5%)	29.5 ± 0.24	—	39.52 ± 0.52	—	0.312 ± 0.018	[132]
Woven basalt fibers	50 wt.%	Aluminum-Epoxy	Hand layup	MMT K10 (5%)	—	—	641.30 ± 11.56	122.22 ± 2.55	—	[133]
Sugar palm	—	Polyester	Hot pressing	OMMT (4%)	24.56	3.683	68.12	3.786	69.19E-6	[134]
Jute	—	Natural rubber	Roll-milling mixing	Cellulose nanocrystals (CNC) (5%)	21.8±0.7	—	—	—	—	[135]
Jute	—	Polyester	—	Bentonite	40.39	2.62	337.93	12.51	0.157	[136]
Sisal	40 wt.%	rPP	Compression molding	Cloisite 30B (5%)	55.95	1.7003	—	—	8.76E-6	[59]
Sawdust	80%	epoxy	Compression molding	ZnO (5%)	—	—	89.60	—	3.82	[137]
Sisal	50 wt.%	Epoxy	Vacuum-assisted resin infusion molding	Cloisite 30B (3%)	55	23	—	—	—	[60]
Hemp	21 wt.%	Polyester	Compression molding	Cloisite 30B (1.5%)	24	6	—	—	—	[120]
baggage	15-30%	Polypropylene	Injection Molding	Nano-graphene (0-1%)	32-46.3	2.2-3.4	55-59	1.8-2.6	39-44 J/m	[138]
Rice straw	40%	Polypropylene	Injection Molding	Nano-silica (0-3%)	—	2.3-2.5	—	1.9-2.8	20.5-21.5 J/m	[139]
Coir	30 wt.%	Polypropylene	Hot pressing	MMT (2%)	9	3	—	—	—	[140]
Wood	30 wt.%	Polypropylene	Hot pressing	MMT (0-2%)	12	4	—	—	—	[140]
Coir fiber	0-10%	Synthetic epoxy	Hand layup	TiC nanoparticle (0,5,10%)	30-58	1.2-2.2	115.05-124	3.9-4.8	4.5-8.8 kJ/m^2	[67]
Pineapple leaf	30 wt.%	Polypropylene	Compression molding	Cloisite 20A (3%)	45.14	6.45	65.01	4.46	—	[122]
Kenaf	30%	Unsaturated Polyester	Hand-layup	ZnO (5%)	58	5.60	68	13	31	[114]
Jute	20 wt.%	Epoxy	Hand layup and Compression molding	Cloisite 20A (5%)	103.05	1.298	162.8	2.8	0.358	[110]
Curauá	10–30 wt.%	Polyester	Cold Pressing	Organophilic Clay (2.5-10%)	36.13	—	32.55	—	—	[121]

Fiber	Fiber content	Matrix	Manufacturing method	Nanofiller (%wt.)	Tensile strength (MPa)	Tensile modulus (G Pa)	Flexural Strength (MPa)	Flexural modulus (G Pa)	Impact Strength (kJ/ mm^2)	Ref.
Coconut (coir pith)	—	Polyester	Hand layup	Nano-alumina (1%)	46	—	83.1	—	—	[141]
Hemp		epoxy	Hand layup	Graphene (0.3%)	68	—	47.22	—	—	[142]
Luffa fiber	20%	epoxy	Hand layup	Graphene (2%)	12.228	2.10	67.05		3.548	[29]
Flax/PLA	28%	epoxy	Vacuum Bagging	Al_2O_3, MgO (3%)	46, 50	4.2, 5.2	95, 64	5.8, 3.8	70, 76 kJ/m ^{2}	[34]
Jute	15 wt.%	Polyester	Hand layup	MMT (5%)	40.38	—	234.93	—	—	[34]
Hemp	—	Epoxy	In-situ polymerization	Hemp nano cellulose (2%)	77.09	2.43	95.78	4.14	21.82 kJ/m ^{2}	[65]
Wild cane grass	40 vol%	Polyester	Compression molding	MMT (4%)	99.57	2.26	221.61	4.192	—	[143]
Sisal	25 wt.%	General polymer	Compression molding	Garamite (3%)	109	—	6980	—	—	[144]
Jute/Coir	105 g fiber/ 630 g of resin	Polyester	Compression molding	Garamite (3%)	43	—	—	—	—	[145]
Wood craft pulp nano-cellulose	5-20%	Polyvinyl alcohol	Mechanical stirring and casting	Nano-silica (5-20%)	2.85-4.69	—	—	—		[146]
Sugarcane baggage	—	Poly vinyl alcohol	Mechanical stirring and casting	Sugarcane baggage nano-cellulose (2.5, 5, 7.5, 10%)	41.3-57.7 (linear) 52-83 (crosslinked)	—	—	—		[56]
TEMPO-mediated oxidized cellulose nanofibers	—	Polyvinyl Alcohol-Chitosan blend	Mechanical stirring and casting	Cellulose nanofiber (0-1.5%)	15-29	—	—	—	—	[50]
Kenaf, coconut	40%	Polyester	Hand layup	Oil palm sell nanoparticles (1-5%)	30.10±1.03-37.56±1.12	0.76±0.04-1.15±0.05	60.42±2.19-75.27±2.43	4.41±0.24-6.17±0.38	10.84±.56-13.42±0.49	[147]
Basalt fiber	—	Epoxy	Hand layup	Coir micro particles and TiC nanoparticle (5-10%)	46.32±1.09-112.87±5.09	1.5-7.8	60-220	2-11	4.8-27.67±1.14	[67]
Hemp fiber	—	Epoxy	Hand layup	Eggshell nanoparticles (0, 7, 14, 21%)	69.99-74.32	—	170-220	—	6-10	[39]
Flax	—	Epoxy	Vacuum-assisted resin infusion	OMMT (1.3%)	87.5	7.55	140	6.2	—	[119]

5 Thermal Properties

Nano clay-infused natural fiber nanocomposites express a higher value of thermal expansion coefficient, enhanced thermal resistance, flammability, and composites [110],[112]. Higher heat deflection temperature of the composites is achieved by these nanofillers which elevated dimensional stability and flame retardation. Moreover, the thermal barrier characteristic is the outcome of the high aspect format of nanoparticles. The larger aspect ratio of nano clay provides a convoluted gateway making it tough for the vapor and gas particles to permeate the composite material [112]. Nanofillers cause layers of lower heat flux over the surface of a material which is used as a fire retardant in composites. These are used as the replacement of halogen to act as a fire inhibitor [148]. The thermal stability of the nanocomposite increased with the addition of Nano cellulose and Nano oxide silicon, particularly at the 5wt% loading, possibly because of the high thermal stability behavior of Nano silicon dioxide [146]. Three-dimensional inorganic ZnO nanoparticles show better thermal stability in different polymer composites [135]. The existence of ZnO significantly influences the mechanical and thermal properties of kenaf/polyester composites [149].

The effect of nano organic and inorganic particles on the thermal properties of different natural fiber composites was introduced in Table 3. In most of the cases, higher thermal stability was attained for adding (0-5) wt.% of nano-fillers. Furthermore, the thermal degradation temperature was mostly above 300°C for these nanoparticle-aided composites and crystallization temperature and enthalpy were also found higher in several cases.

Table 3 Effect of Nanomaterials on Thermal property of various composites

Nano-filler	Type	Fiber/matrix	Thermal property	Ref.
Nanoclay	organic	Sisal-polypropylene	Higher thermal stability	[59], [150]
Nanoclay-MMT (montmorillonites)	organic	Jute- polypropylene	Better results for 1% and 5% nanoclay in thermogravimetric analysis	[151]
MWCNT (multi walled carbon nanotube)	organic	epoxy	The glass transition temperature, thermal stability and decomposition temperature increased	[148]
ZnO nanoparticles	inorganic	Kenaf/polyester	Significant improvement in thermal stability	[114]
ZnO powder	inorganic	polymer	Enhanced thermal stability	[123]
Nano-alumina	inorganic	Coir/polyester	TGA) and DTG shows highest thermal stability.	[141]
Nanoclay-MMT	organic	Rice husk/high density polyethylene	The crystallization temperature, crystallization enthalpy and crystallinity level increased	[152]
Nano-Al_2O_3	inorganic	sisal/coir/epoxy, sisal/banana/epoxy, banana/coir/epoxy	Degradation temperature improved	[153]

6 Physical and Chemical Properties

The water absorption capacity of nano clays depends on the number of exchangeable cations in interlayer [148]. Inorganic SiO_2, ZnO and TiO_2 nanofillers are used as corrosion-resistant substances in organic coatings [135]. The Addition of Montmorillonite nano clay improved the water absorption capability of hybrid composite combinations of various natural fibers (kenaf, coir, and wood) and polypropylene [154]. On the contrary, the addition of montmorillonite nano clay in natural fiber-reinforced hybrid nanocomposites, manufactured by compression molding technique with wood particles, hemp fiber, and polypropylene decreases the water absorption property [99]. Ferric oxides added to fiber boards result in enhanced thickness swelling and water absorption properties. The brittleness of the thermoset can be significantly reduced by adding a metal oxide nanofiller where hardness is improved due to the improved density [135]. It is generally believed that because of the barrier effect of the nano clay, which impedes penetration of O_2 into the sample, the destruction of the nanocomposite occurs at a higher temperature [154]. Besides, natural fibers have better specific properties than synthetic fibers, which in combination with another reinforcing (nanofillers) enhances the performance. Weight gain and FTIR spectrum analysis indicated that 5% nano clay addition gave favorable reduction in the water absorption behaviors of vinyl-ester eco-nanocomposites [155]. Alumina and Magnesia were used as nanofillers and found improved water absorption characteristics in flax/PLA bio composites [156] and jute/epoxy nanocomposite [157].

It is obvious from Table 4 that the hydrophilic characteristics of natural fibers give rise to weight gain whereas the hydrophobicity of nanoparticles reduces the water uptake of the hybrid composites. Moreover, nanoparticles decrease the thickness swelling. The effect continued with increased density and decreased void fraction of the composites by the addition of nanoparticles. The higher percentage of fiber shows higher biodegradability in composites.

7 Scanning Electron Microscope (SEM)

Many researchers have presented studies that provide information on the influence of the addition of nanofillers to interactions between the filler and matrix at the interface via SEM analysis. K. R. Sumesh et al. [164] studied the surface property of banana-pineapple fiber after the flexure test with the addition of 0% and 3% TiO_2 nanofiller and it was observed the reduction of void formation and resulting in improved flexural properties (Figure 9). It was reported that the addition of 4 wt.% nano-silica in the kenaf-sisal hybrid composite shows less crack formation after flexural test [165] though fiber pullout was observed.

E. Rosamah et al. [57] analyzed the tensile fracture surface of kenaf/coconut mat with the addition of 0-5% Oil Palm Shell nanoparticles. It was found that an enhanced portion of nano OPS up to 3% provided better matrix formation with reduced voids and fiber fracture. However, after increasing the amount from 3-5%, higher numbers of voids were detected which is probably due to the poor wetting of the fiber due to higher OPS nanoparticles content in the polymer matrix.

In an experiment conducted by K. G. Ashok et al. [29] with the incorporation of PbO nanoparticles with Luffa Fiber, it was figured that pull out of fibers from the surfaces which is due to the agglomeration of nano PbO particles and particle-particle interaction at higher weight percentage, thus restricting the interaction of the nanoparticles with the matrix fiber interface (Figure 10).

From the micrograph of ZnO nanofiller-based composites, it is concluded that the void content decreased with the incorporation of ZnO [163].

Fig 4(a) shows the fractured surface of the Sugar Palm fiber/Polyester composite without any additive whereas Fig 4(b) depicts the composite with 2% nano clay [134]. By investigating the impact energy for both samples, it was reported that the sample without nanofiller exhibits poor interfacial bonding.

Table 4 Effect of nanoparticle in the physical properties of different composites

Nano-filler	Fiber-matrix	Types of tests	Result	Reference
Montmorillonite (2 phr)	Kenaf-coir-polypropylene	Water absorption, Biodegradability	The water absorption increased steadily until 100 days then became constant and hybrid composites absorb more water than the single composites.	[158]
Nano-SiO_2 (0, 2, and 5 wt%)	Baggage-high density polyethylene (HDPE)	Water absorption, Thickness swelling	The percentage of water absorption depended upon the wt% of SiO_2 nanoparticle. The maximum absorption of water was found at 5 wt% of nano infusion. The highest percentage of thickness swelling was at 2 wt% of nano-SiO_2 after 2h.	[113]
Nano-ZnO (5 wt%)	Kenaf-unsaturated polyester	Water absorption, biodegradability	Water uptake was tested with 4 different layers of kenaf fiber where 2 and 3 layers of kenaf showed more water absorption compared to 1 and 2 layers of kenaf in 5 wt% ZnO filled nanocomposite. Because of the highest kenaf contained, 4 layers kenaf/ZnO/polyester composite gave higher degree of degradation compared to other	[114]
Silica nanoparticle (0, 1, 2, 3, and 4 wt%)	Hemp-sisal-epoxy	Theoretical density, experimental density, and void fraction	Density increased with the addition of nanoparticle and void fraction decreased with the increasing nano silica content	[125]
Bamboo nanoparticle (0-5 wt%)	Woven/nonwoven kenaf-unsaturated polyester	Theoretical density, measured density, void fraction, and water absorption	Unfilled composites had the highest void contents whereas 3 wt% of nano particle for both woven and nonwoven kenaf fiber composite provided lowest voids. The percentage of water absorption decreased with concentration of nanoparticles.	[147]
Calcium carbonate nanoparticle	Kenaf-polyester	Water resistance test	$CaCO_3$ caused increment of water absorption in the composite	[159]
Nanographene (0,1,2, and 4 phr)	Wood flour-HDPE-foaming agent	Water absorption, thickness swelling	Water absorption and thickness swelling reduced with the addition of graphene nanoparticle	[160]
Carbon nanotube (0, 1 and 2 phr)	Wood flour-PVC	Cell density, water absorption, thickness swelling	Highest cell density was found by the incorporation of 2 phr CNTs into the composite.	[161]
Nanoclay (0, 2, and 5 phr)	Wheat straw flour-foamed HDPE	Water absorption, thickness swelling	the water absorption and thickness swelling found lower	[162]
Nano-ZnO (0, 1, 2, and 4 phc)	Wood flour-polypropylene	Water absorption, thickness swelling	The moisture absorption and dimensional stability of the composites progressively decreased with an increasing nano-ZnO loading. Highest thickness swelling was found with 4 phc ZnO-nano filler	[163]

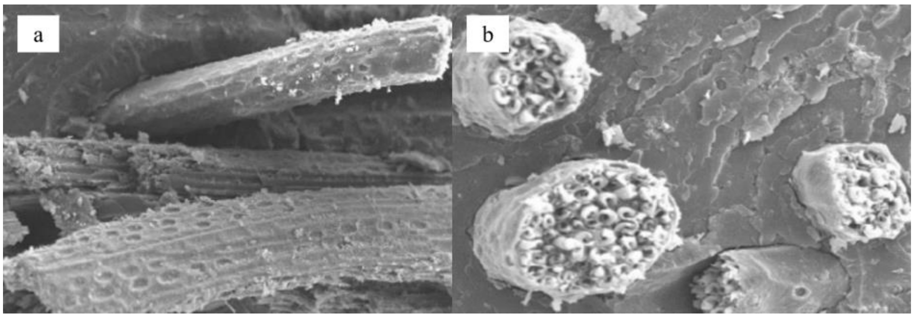

Fig 4 SEM images of the (a) 0% Nano Clay and (b) 2% Nano Clay composites [134]

Majid, et al. [2] investigated the mechanical and morphological properties of napier/epoxy composites. The SEM interrogations of the fabricated specimen reveals that the higher wt% incorporation of the filler material reduces the fiber pull out from the matrix. Delamination or low interfacial properties can be considered the key drawback of using such green composites. Delamination can be occurred due to the presence of residual stresses during the manufacture. For designing such structures, it's inevitable to gather the concept of interlaminar fracture properties. Influence of fiber architectures on the mode-I and mode-II interlaminar fracture toughness of flax fiber epoxy composites were investigated [166]. The mode-I interlaminar fracture toughness (GIc) of flax, glass and hybrid flax-glass fibre woven composites were studied using a DCB test and observed by SEM images [167]. The effect of water absorption on the interlaminar fracture toughness behavior of woven flax and flax/basalt reinforced vinyl ester composite laminates were examined and it was found that hybridization of basalt fiber improved the interlaminar fracture toughness [168]. Results of adding TiO_2 on flax fiber reinforced epoxy composites depicts the significant improvement in the Mode I and Mode II interlaminar fracture toughness [169]. It was found that nano

TiO_2 at 0.4 wt% addition improved the GIc value by 52% and 0.5 wt% addition resulted in 73% improvement in the GIIc value. Mode I and Mode II interlaminar fracture toughness for 0.4 wt% and 0.5 wt% addition of TiO_2 correspondingly is depicted in Fig 5. SEM images of the composites (fiber failure at various tests) depicts that the addition of various nano particle at certain percentages enhance the mechanical properties though at higher percentages the properties deteriorate due to the agglomeration of the nano fillers. Moreover, it can be said that the void between the matrix and fiber significantly reduced by incorporating nano filler. Therefore, it is highly recommended to add nano fillers with calculated proportion in the fabrication of composites for getting the optimum characteristics.

8 Applications

Bio-based composites provide a versatile area for potential use in various sectors such as in automobiles, home appliances, medicine, weapons, civic infrastructure, the navy, sports, packaging, electronics, etc. [149], [170]. Ford was the first automaker to use plant fibers in composites in 1930 for body panels [171]. Following that, Mercedes-Benz constructed the door structure of jute/epoxy in the 1990. It was reported that the annual use of natural fiber composites by the German car industry is 19 kt, of which 64% are flax, 11% are jute/kenaf, 10% are hemp, and 7% are sisal [172].

For increasing the mechanical and physical properties of polymer composites, nanoparticles are currently considered to be a viable filler material. Bio composites with nanoparticles are employed to obtain fine properties of organic coatings. As a result, these nanocomposites have been thoroughly researched to produce organic anticorrosion coatings that are also advantageous to the environment. The functionalization of natural fiber-based composites with different nanoparticles has a good prospect such as waterproofing, flexible processability, fire resistance, high magnetic properties antibacterial qualities, UV protection, insulation, self-cleaning capabilities, etc. The fact that they are made of bio-based ingredients and naturally decompose makes them significant environmentally safe and harmless materials. Recently, in the biomedical field, nanocellulose is increasingly gaining popularity for its use in tissue engineering, bone regeneration, drug delivery systems, skin replacement after burns, and wound dressings [173]. A CS-based composite containing selenium nanoparticles was developed by Kalishwaralal et al [174] for tissue engineering applications. By employing nanocellulose, food storage, transportation, and shelf life are all improved. White cheese was kept in storage for 30 days at 7° C. After 15 days of storage, the composite with 2%, 4%, and 8% filler added lowers coliforms when compared to the control film [175].

A summary of the applications of different natural fiber based nano composites are listed in Table 5.

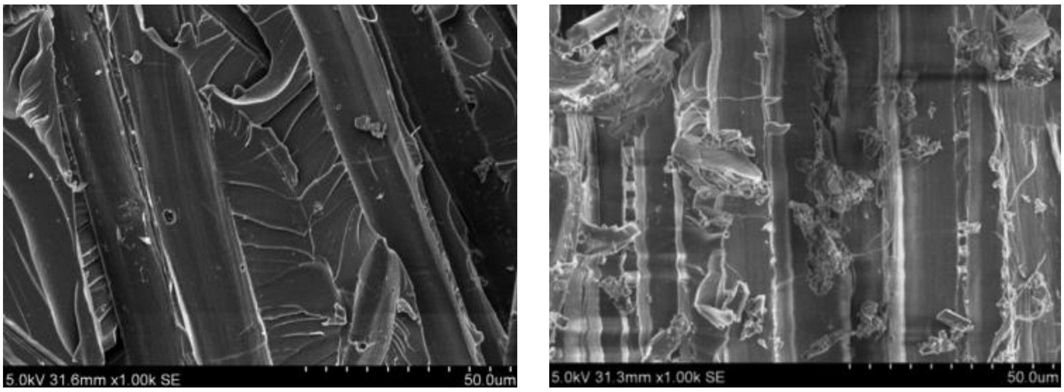

Fig 5 SEM images of (left) Mode I test specimens fractured surface for 0.4 wt.% (right) Mode II test specimens fractured surface for 0.5 wt.% added of TiO2 in flax fiber reinforced epoxy composites [169]

Table 5 Application of natural fiber based nano composites in different sectors

Sectors	Fiber used	Nano particle used	Application Sectors
Automobile and Aerospace	abaca, pineapple leaf, coir, oil palm, bagasse, bamboo, wheat straw, curaua, and rice husk	Graphene, nano clay,	seats, body panels, carpets, glove box, seat backrest panel, trunk panel, sun visor, roof cover, car dashboard
Food Packaging	Hemp, Ramie, Jute, kenaf, Chitosan	TiO_2, ZnO NPs, nano clay, Graphene	Packaging of Fruits,
Construction	Hemp, Oil palm, Jute, wood, coir, sisal	Graphene, nano clay	Roofs, windows, panels, Railing
Electrical and Electronics	Hemp, Rice Husk	Nanocellulose, Cu NP, MgO NP	Mobile cases, laptops cases,
Tissue Engineering	Chitosan, Silk,Feather	SeNPs, crystal nanocellulose	cardiac patches
Cosmetic industry	cotton	Nano clay	Cosmetic skin mask
Medical sectors	Wood, Bagasse, CNT	Graphene, nano clay, Cu NP,Se NP, SiO2 NP	dental brackets, bone and tissues repair and reconstruction, and implant,
Sports Equipment	Flax	Graphene,	Paddle, golf, hockey stick, skeleton of tennis rackets, helmets, bicycle, Frames, Snowboards
Others	Coir, Ramie, Wood, Flax, Kenaf,	CuO,	helmets and post-boxes, mirror casing, paperweights, industrial sewing threads, fishing nets, canvas, Bricks, pipes, UV shielding equipment

9 Conclusion

Natural fiber could be the best alternative to synthetic fiber for manufacturing industrial goods considering the global environmental challenges [176]. Composites with fiber or particle reinforcements have achieved some incredible achievements in recent years. The research area on nanocomposites has been identified as being dynamic and competitive. Focusing on the fabrication processes of nanocomposites, this review article has examined the mechanical, thermal, and physical properties as well as applications.

Although natural fiber-reinforced composites are made for distinct applications with different materials, their characteristics depend on the matrix characteristic, and nature and compositions of the nanomaterials and fibers used. Therefore, a crucial stage in the creation of high-quality nanocomposite materials is the selection of an appropriate nanofiller, fiber, and matrix. Moreover, as the manufacturing process also has a great impact on productivity, composite quality, and the process's overall success, it is one of the most essential factors to consider and should be further researched. Additional research is needed to identify the various techniques of nano-reinforcement leading to large changes in material properties and their potential future. Most importantly, the harm of plastic use and other non-environment friendly materials and the benefits of natural fiber-based nanocomposites should be made more widely known. Furthermore, emphasis can be provided on the synthesis of nanocellulose and use as a nanofiller in the bio composites as reinforcement. Eventually, it can be said that there is plenty of scope of study to reveal the benefit of using nano filler in composite processing by modifying the technique, surface treatment and the quality of the raw materials.

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