REVIEW

A Comprehensive Review on Epoxy Biocomposites Based on Natural Fibers and Bio‑fllers: Challenges, Recent Developments and Applications

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Received: 17 August 2021 / Accepted: 31 January 2022 / Published online: 19 April 2022 © Donghua University, Shanghai, China 2022

Abstract

Natural fber-reinforced polymers remained a hot topic of interest in material sciences over the last two decades. Such fbers are appealing in composite materials due to their renewability, low density, better specifc strength, biodegradability, accessibility, and low cost. Polymers reinforced with natural fbers provide improved mechanical performance at a lower cost and could be the best alternative to synthetic fbers. Bio-fllers have gained much attention nowadays due to their cost-efectivity and the ability to modify base materials in the composite structure. Natural materials reinforced epoxy composites have a high capacity due to their eco-friendliness, economic feasibility, and technical viability. However, some parameters directly infuence desired product performance. This review discusses various properties of natural fbers and their impact on the overall performance of natural fber-based epoxy composites. It summarizes the recent research on natural fbers/bio-fllers reinforced epoxy biocomposites.

Keywords Biocomposites · Natural fbers · Bio-fllers · Reinforced epoxy composites

Introduction

Due to various environmental benefits, researchers are working to produce renewable composites that are fully or partially bio-based [\[1](#page-16-0)]. A bio-composite is a composed of a polymer matrix with natural fbers and reinforced bioparticles. Synthetic fber-reinforced polymer composites have been widely used recently for various technical applications due to their high strength-to-weight ratio. Nevertheless, they have drawbacks like the high cost of initial material, increased energy demand, non-biodegradability, and environmental impact. Natural fbers are considered to be the best substitute for synthetic fbers. Natural fber-reinforced composites are less expensive and more environmental friendly than synthetic fiber composites [[2](#page-16-1), [3\]](#page-16-2). Natural fber-based composites consist of natural fber in the polymer matrix. By 2024, the market for natural fber-reinforced composites is predicted to reach \$10.89 billion. The natural fber-reinforced polymers have found successful applications in automobiles, aircraft, constructions, electronics, and other industries to replace the internal and external work because of their reduced price, high stifness, strength, degradability and reduced weight [[4–](#page-16-3)[7](#page-16-4)]. Load-bearing components for structural and infrastructural purposes have been made from natural fber composites. A few examples of such fbers include multipurpose panels, beams, roofng, storage tanks, and pedestrian overpasses [[8\]](#page-16-5).

Two main types of polymers include thermoplastics and thermosets. Polyester, phenolic, and epoxy are the most commonly used thermosetting resins [\[9](#page-16-6), [10\]](#page-16-7). Epoxy-based composite materials possess good mechanical properties, super adhesiveness, high specifc strength, good heat, and solvent resistance at a lower cost and are widely used in load-bearing applications. Currently, there is a great need to understand the properties of natural materials to fulfll

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engineering requirements. The use of natural fbers-based epoxy composites for structural materials has received great attention in the last two decades [[11](#page-16-8), [12\]](#page-16-9). Researchers are therefore continually working for more practicable and durable development and industrial application of biocomposites. This review encompasses various parameters affecting the performance of natural fbers and specifcally discusses the recent research done on bio-fllers and natural fbers-based epoxy and hybrid composites.

Challenges Using Natural Fibers for Composites

Efect of Chemical Composition

Natural fbers consist of proteins, lignin, waxes, cellulose, pectin, and hemicellulose. Table [1](#page-1-0) summarizes the chemical composition of diferent natural fbers [[13–](#page-16-10)[15\]](#page-16-11). The chemical composition of plant fber, and resultant the composites has a signifcant impact on their properties. Cellulose and hemicellulose have a hydrophilic tendency, indicating that plant fber has high moisture content and, as a result, the composite has low moisture resistance [\[16\]](#page-16-12). The primary parameters to achieve excellent performance are a high degree of polymerization, a higher cellulose content, a smaller microfbrillar angle, and a higher cellulose crystallinity. Fibers' Young's modulus and tensile strength typically increase as cellulose content rises. The stifness of fbers is determined by the microfbrillar angle. If the microfbrils angle is low, fbers are more ductile, whereas fbers with high microfbrils angle will be infexible and stif with high tensile strength. Hence, all these factors are taken into account while selecting plant fbers as reinforcement in structural applications. Bast fbers have a low microfbril angle, a high crystallinity, and a high cellulose content as they support the plant's stalk [[17](#page-16-13)].

Interaction between Fiber and Matrix, Adhesion Problems

The majority of natural fbers contain a cellulose-rich core as well as waxes, fats, lignin, pectin, and hemicellulose. Lignin, oils, and wax cover the outer surface that serves as a cementing layer and prevents the formation of powerful interphase. It renders the surface of fber highly hydrophilic and incompatible with hydrophobic polymer matrices. Therefore, the main objective is to improve the interphase by removing this cementing layer. Removal of this layer improves mechanical interlocking by increasing interfacial shear strength along with fiber wettability [[18](#page-16-14)]. The fiber/matrix interface and fber surface wetting property are critical in determining the mechanical properties of composites. The fexural strength, tensile strength, and toughness of composites are all afected by fber wettability [[19](#page-16-15)[–22](#page-17-0)]. There are various physical and chemical techniques to increase the interfacial strength of a composite.

Mercerization of natural fibers is commonly used to increase the compatibility of natural fber and polymer resin. The alkaline treatment exposes the amount of cellulose on the fber surface, enhances surface roughness, and increases the number of possible reaction sites, hence providing a better mechanical interlocking [[23\]](#page-17-1). The higher the surface energy of plant fbers, the greater the fber wettability, and higher the surface roughness, the better mechanical interlocking and the greater the fiber epoxy matrix adhesion. Furthermore, the combination of alkali, organosilane, and epoxy dispersions also improves the jute fber-epoxy adhesion strength. The interfacial shear strength of jute/epoxy composites increases signifcantly by up to 40% [[18\]](#page-16-14). Acetylation increases the fber's surface roughness, resulting in enhanced mechanical interlock with the matrix [[24](#page-17-2)]. The maleated coupling treatment modifes the polymer matrix and fber surface, augmenting interfacial bonding between matrix and fber [\[25,](#page-17-3) [26\]](#page-17-4). Silanes increase the hydrophobicity of natural

Table 1 Chemical composition of various natural fbers [[13](#page-16-10)–[15](#page-16-11)]

fibers, thus improving the strength of natural fiber reinforced composites $[27]$ $[27]$. UV treatment induces the polarity of fibers and improves the bending strength of epoxy matrix composites up to 30% [\[28](#page-17-6)]. Plasma treatment enhances the surface roughness of fber, consequently increasing interfacial adhesion [[21\]](#page-17-7). Plasma treatment also improves bending strength and interlaminar shear strength in natural fber composite up to 30% and 35%, respectively [[29\]](#page-17-8). Chemical treatments remove the waxy substances such as lignin and hemicelluloses from the fber surface, leaving the fber surface rough (Fig. [1](#page-2-0)). This roughness boosts mechanical interlocking and promotes fber-matrix adhesion, resulting in better mechanical performance [\[30](#page-17-9)].

Fiber Strength Variation

One of the major problems with natural fbers is that their qualities vary even within the same type of fber. The properties of natural fbers change signifcantly depending on chemical composition and structure, harvesting period, growing conditions, extraction techniques, treatments, storage methods, and even testing methods/standards. As previously stated that due to its inherent nature, chemical composition strongly afects fber properties. In addition to cellulose content and microfbrillar angle, other structural parameters such as cell dimensions, and defects also play an important role in the determination of fber properties [[31](#page-17-10)]. The harvest season has an impact on fiber properties as well. Harvesting in the autumn and winter is best for mechanical properties [[32](#page-17-11)]. The studies have revealed that the tensile strength of hemp fber varies during the fowering stage and increases up to 114 days of growth [[26\]](#page-17-4). The retting technique also infuences the chemical composition,

structure, and performance of the natural fber. Biological retting produces high-class fbers, but it increases environmental pollution as this method generates a large amount of wastewater, making it unsuitable for most commercial applications $[15]$ $[15]$ $[15]$. The tensile strength of elementary flax fiber varies depending on the isolation process and ranges from 1500 to 1800 MPa. The strength of fax fber extracted by hand was found 20% higher than that of flax fiber extracted by machine [\[33\]](#page-17-12). Other factors infuencing the natural fber properties include fber diameter and cross-section area. The diference in determining the cross-sectional area of plant fbers causes a signifcant variation in the strength and modulus of the same plant fber. The tensile modulus of a single natural fber decreases as the diameter of the fber increases [[14](#page-16-16)].

Moisture Absorption

Due to abundant hydroxyl groups, the moisture resistance of natural fbers is poor. They absorb moisture from the environment until equilibrium is achieved. This moisture absorption in hydrophilic natural fbers has a signifcant impact on their physical and chemical properties, resulting in dimensional variations, changes in mechanical and chemical performance, and composite failure. The amount of moisture absorbed by the fbers infuences their volume in the composite to reinforce natural fber in a polymer matrix. Secondly, polymerization at temperatures above 100 °C vaporizes the water contained within the natural fber. These efects cause internal stresses in the composite which can lead to a signifcant decline in the preliminary properties of the composite. Assarar et al. [[34\]](#page-17-13) fabricated epoxy composites out of glass and fax fber and compared the efects of

Fig. 1 Scanning electron microscopic images of **a** untreated jute fiber, **b** alkali treated jute fber, **c** silane treated jute fber, and **d** alkali and silane treated jute fiber [\[30\]](#page-17-9)

water aging on both composites. They claimed that moisture absorption of fax fber composite was excessively high when compared to glass fber reinforced composite. The saturated weight gain of flax fiber epoxy composite was 12 times higher than that of glass fber epoxy composite. Espert et al. [[35\]](#page-17-14) investigated the impact of water absorption on the mechanical properties of cellulose fber-reinforced composite. They discovered that increasing fber content increases moisture absorption of the composite due to the hydrophilic nature of the natural fber. Le duigou et al. [[36\]](#page-17-15) analyzed the efect of seawater on biodegradable fax reinforced polylactic acid (PLLA). The unreinforced PLLA had a saturated weight gain of 0.32%, while that of flm-stacked specimens was 5.6%. It means that the weight gain of flax fiber at 20 $^{\circ}$ C was 17 times higher than that of PLLA.

All of the preceding discussion indicates that the natural fbers absorb a lot of moisture. Table [3](#page-9-0) presents the moisture regained from various natural fbers. This low moisture resistance noticeably infuences the mechanical performance of the composite. Many researchers have investigated the efect of moisture absorption of these natural fbers on the mechanical properties of the resultant composite. Le duigou et al. [[36](#page-17-15)] reported that moisture absorption degrades the bonding between fber and matrix and reduces tensile stif-ness and strength. Yan et al. [\[5](#page-16-17)] analyzed the effect of various aging solutions on the mechanical properties of fax epoxy composite. The results show a signifcant reduction in the tensile and fexural properties of the composite. They noted a decrease of 22.6–31.1% in tensile strength, 24.0–36.4% in tensile modulus, and a reduction of 9.3–23.5% in fexural strength and 13.9–25.2% in fexural modulus. Dhakal et al. [\[37\]](#page-17-16) found that flexural properties for both jute-reinforced composite and fax-reinforced composite decrease as the percentage moisture uptake increases. Compared with dry fax samples, the strength and modulus of the wet fax samples decreased by nearly 40 and 69%, respectively, while wet jute sample strength and modulus decreased by 60 and 80%, respectively. Moreover, water can have a plasticizaing effect and can plasticize the internal chemical structure [\[38](#page-17-17)].

Treatment of natural fbers with diferent chemicals reduces their moisture regain to a large extent [[39\]](#page-17-18). Nayak et al. [[40\]](#page-17-19) treated areca sheath fber with NaOH,

sodium chlorite, permanganate, and benzoyl chloride. They observed that all chemical treatments reduced water absorption. However, when compared to other treatments, benzoyl chloride and sodium chlorite-treated fbers exhibited high moisture resistance. Dilf et al. [\[30\]](#page-17-9) investigated whether the combination of alkali and silane treatment reduces the moisture absorption rate of jute fber epoxy composite. Teklu et al. [\[41\]](#page-17-20) noted that sisal fber with an alkali-treated surface and polyaniline coating, shows signifcant resistance to water uptake. The water absorption reduced from 20.9 to 9.8% after surface treatment, compared to untreated sisal fbers. Polyaniline-coated fbers had a much lower absorption (3.5%) as compared to alkalitreated uncoated fbers.

Polymer Matrix and Curing Agents

Epoxy Resin

Epoxy resins are special amid various thermosetting resins due to their unique properties: such as low creep, frst-rate corrosion weather resistance, high-temperature performance, suitable electrical properties, lower shrinkage, and high availability of the resin in diferent forms (ranging from low viscous liquid to highly solid). Similarly, it requires minimum pressure for the product manufacturing. Their physical and mechanical properties can be adjusted to achieve extreme flexibility or high strength [\[42](#page-17-21)]. The resin can use several materials like glass, metal, wood, and natural fbers [[43\]](#page-17-22). Due to these unique characteristics and valuable properties, epoxy resin is widely used in engineering composites, surface coatings, structural adhesives, and electrical laminates [[44,](#page-17-23) [45\]](#page-17-24).

Epoxy resin is a thermosetting polymer that contains at least one epoxide or oxirane group. The basis for all types of epoxies is the diglycidyl ether of bisphenol A (DGEBA). Nearly 90% of the world's epoxy resin development today is based on the reaction between bisphenol-A and epichlorohydrin in the of a simple catalyst, manufacturing DGEBA, as shown in Fig. [2](#page-3-0).

Fig. 2 Synthesis of DGEBA

Curing Agents for Epoxy resin

Curing agents are also known as cross-linkers or hardeners. They are used to transform epoxy into thermoset networks. They can catalyze the curing reaction or initiate it through polyaddition and/or copolymerization with epoxy monomers. The structure of cross-linking agents considerably afects the properties of the cured products. Curing retailers are extremely important in the epoxy resin curing system because they are related to curing kinetics, reaction rate, curing degree, viscosity, and curing period, and thus play a signifcant role in determining the overall attributes of cured products. Active hydrogen compounds and their derivatives are the most common type of cross-linking agents. These hardeners entail amine, amide, hydroxyl, acid or acid group anhydride compounds. They generally react using polyaddition with epoxy resin to provide an amine, ether, or ester. Another type comprises of anionic and cationic initiators and is applied to catalyze epoxy resin homopolymerization. Molecules capable of creating anions such as tertiary amine, secondary amines, and metal alkoxides are efficient anionic initiators for epoxy resins. The third type of hardeners, also known as reactive cross-linkers generally, possess larger equivalent weights and crosslink with the 2nd hydroxyls of the epoxy resins or by self-condensation [\[46\]](#page-17-25). Table [2](#page-4-0) presents some commonly used hardeners along with their chemical structures.

Natural Fiber Reinforced Epoxy Composites

Diferent physical and chemical approaches (Fig. [3\)](#page-4-1) have been used for better fiber reinforcing, as mentioned in Sect. [2](#page-1-1). Various researchers have analyzed and investigated these types of treatments [[15](#page-16-11), [47](#page-17-26), [48](#page-17-27)].

Herein, we focus on improved treatment procedures, the use of bio-fllers, and hybrid approaches, all of which are increasingly being used to improve the quality of

Modification techniques

PHYSICAL METHODS	CHEMICAL MODIFICATION
• Corona treatment Plasma treatment Gamma Radiation	• Alakali treatment • Silane treatment • Enzyme treatment • Peroxide treatment • Stearic acid treatment • Etherification • Acetylation • Sodium chlorite treatment • Maleated coupling Ozone treatments • Sodium chlorite treatment • Benzoylation Grafting \bullet

Fig. 3 Physical and chemical treatments for modifcation of natural fibers

composites. The efect of hot alkali treatment on the tensile and bending properties of fax epoxy composite was investigated, in a typical study, as shown in Fig. [4.](#page-6-0) The study revealed that higher temperature fber treatment accelerated the conversion of cellulose 1 to cellulose II, as obvious in XRD result in Fig. [4](#page-6-0)B. The treatment removed hemicellulose, some lignin, and other surface impurities such as wax and oil, creating cracks on fiber surface due to damage caused by alkali treatment (Fig. [4C](#page-6-0)). However, the removal of these impurities results in a cleaner fber surface and a rougher topography, shown in Fig. [4C](#page-6-0); the roughness increased with increasing treatment temperature, Fig. [4](#page-6-0)C(e). These rough fber surfaces help in mechanical interlocking with the matrix; the cleaner fiber surface improves chemical interaction between fber and matrix, resulting in increased fber-matrix adhesion; and the appearance of cellulose II improved mechanical properties. The maximum

Fig. 4 A Process of alkali treatment. **B** XRD patterns of alkali treated ◂fax fber at diferent temperatures. **C** FE-SEM images of fax fber **a** UF, **b** AFRT, **c** AF80, **d** AF120, **e** AF160. **D** Tensile properties of different temperature alkali treated fber reinforced composite **a** tensile strength, **b** elastic modulus. **E** Bending properties of diferent temperature alkali treated fber reinforced composite: **a** bending strength, **b** bending modulus

improvement in tensile strength (8.59%) and elastic modulus (50.4%) were obtained for composites treated at the highest temperature of 160 °C (the highest treatment temperature in this work), compared with the untreated composite Fig. [4](#page-6-0)D. The composite with fbers treated at 80 °C had 44.5% higher fracture energy. Similarly, bending strength and bending modulus improved 28.8% and 68.0%, respectively for composites with fber treated at 160 °C (the highest treatment temperature used in this study) compared with untreated fber composite [\[49](#page-17-28)].

Another technique to improve the properties is the inclusion of rigid/stifer bioparticles in the epoxy matrix. Nevertheless, various factors such as the composition of the fller, the preparation process, improper dispersion of the fller in the matrix, fller matrix interactions, and fller orientation can all afect the mechanical properties of bio-fllers reinforced composites. In a recent study, the olive powder obtained from three diferent parts: olive tree small branch (OTS), olive tree big branch (OTB), and olive tree leaves, were reinforced in the epoxy matrix. Except for OTL composite, the tensile strength and modulus of the epoxy composites increased after with reinforcement. The fexural strength and modulus improved for composites with all three reinforcements (Fig. [5\)](#page-7-0). The strength of the OTL fllerreinforced composite decreased due to the poor dispersion of OTL fller in the epoxy matrix. The SEM images (Fig. [5\)](#page-7-0) corroborated these fndings. Filler breakage was seen in OTS and OTB and fller pullout in OTL, showing improved interfacial interaction and stress transmission from matrix to fller, resulting in improved tensile and fexural properties for OTS and OTB. However, the impact strength of the composites decreased with the addition of olive fller. This reduction was due to the low strength of natural fber compared to synthetic ones. With increasing immersion time, water absorption and thickness swelling increased for all composites [\[50](#page-17-29)].

The use of hybrid fbers or fllers is another promising technique that is becoming popular. The use of inorganic hard particles can minimize polymer chain mobility, redistribute stress, and improve composite stifness. In recent research, basalt powder (2.5 to 10 wt%) was combined with six-layered basalt fabric in the epoxy matrix. The basalt powder composite with 2.5 wt% of basalt powder had the maximum tensile strength, elastic modulus, storage modulus, and glass transition temperature. It was due to the better dispersion of basalt powder in the epoxy matrix, as obvious in SEM images (Fig. [6\)](#page-8-0). The stifness of the fllers caused a reduction in the elongation at break and impact strength of the reinforced composites [\[51](#page-17-30)]. Further advancements in natural fber composites are covered in the following section.

Fiber Type

Epoxy, as a matrix, can be used to create a wide range of composites. They have broadly grouped into fber reinforced plastic (FRP) composites, particulate composites, and nanocomposites [\[52](#page-17-31)]. The reinforcement of natural materials improves the physical and mechanical properties of the matrix. Natural fbers are divided into three main categories according to their origin; mineral, animal and plant fbers. Figure [7](#page-9-1) depicts the various classifcations of natural fbers [\[47](#page-17-26), [53](#page-17-32)]. Animal fbers consist of proteins, and are not mostly favored for high performance except silk fber. Silk fber can give high strength, but its high cost and availability are the shortcomings. Woven silk fber-reinforced composite exhibite far higher fracture strain abilities than glass fber composites and are the appropriate selection to be used in places where high compliance is required [\[54](#page-17-33)]. Plant fbers are stronger and stifer than animal fbers, making them ideal for structural applications [[55,](#page-17-34) [56](#page-17-35)]. Plant fbers can be categorized as wood, bast, seed, leaf, and fruit fbers, depending on the plant portion from which they are extracted [[57\]](#page-17-36). Cotton, jute, hemp, kenaf, fax, bamboo, coconut, and sisal are the commonly used plant fbers. The bast fbers such as fax, hemp, and jute are preferred for structural applications in composites because of their good mechanical properties [\[4](#page-16-3)]. Mineral fbers were outlawed in many countries due to their carcinogenic properties, hence they are less commonly used in high-performance technical applications [\[58\]](#page-17-37). The qualities of reinforced natural fbers have an impact on the performance of polymer composite. Table [3](#page-9-0) demonstrates some characteristics of natural fbers [[13–](#page-16-10)[15](#page-16-11), [54,](#page-17-33) [59\]](#page-17-38). Natural fbers properties depend on fber type, harvesting time, growing conditions, extraction time, chemical composition and structure, treatment, and storage procedures.

Type of fber is a critical parameter in defning the overall properties of the composites [\[60](#page-17-39), [61](#page-18-0)]. Table [4](#page-10-0) shows recent research work done on diferent natural fber reinforced epoxy composites.

Fiber Volume Fraction

The demand for multifunctional materials in advanced engineering and the medical feld is growing nowadays. The material properties and the structure (chemical and physical) both contribute to the multifunctionality. Materials having high strength, stifness, fatigue, toughness, and low damping properties are classifed as structural-functional materials, while others with electrical resistivity

Fig. 5 SEM images of the tensile fracture surface of epoxy reinforced with **A** OTS, **B** OTB, and **C** OTL composites. **D** Tensile strength and modulus of OTS, OTB, and OTL reinforced epoxy composites.

E Flexural strength and modulus of OTS, OTB, and OTL reinforced epoxy composites [\[50\]](#page-17-29)

and thermal conductivity are classifed as nonstructural functional materials. Generally, the main goal of industrial products is to minimize material weight and cost. Stifness and strength, specifc tensile stifness and strength, and specifc bending stifness and strength are basic mechanical properties. Mechanical, thermal, and water absorption, all these properties are infuenced by the amount of fber in the composite $[82]$. Several natural fibers, especially bast fbers such as jute, fax, and hemp show good specifc tensile stifness than E-glass. As natural fbers have a lower density than E glass, therefore the reinforcement of natural fber in polymer matrices at the same fber content makes the fnal product almost 40% lighter than glass fber reinforced composite [\[83](#page-18-2)[–85](#page-18-3)]. Almari et al. [\[86](#page-18-4)] studied the efect of recycled cellulose fber on the physical and mechanical properties of the epoxy composite. They demonstrated that bending strength, bending modulus, fracture toughness, and impact toughness increased with increasing fber ratio. Furthermore, increasing the fber ratio increases the fber-matrix interaction, and the composites having higher fber content have better interfacial bonding. They also discovered that composites with a high fber content absorbed more water, with diminished interfacial bonds, and less fracture toughness, bending strength, and bending modulus. Fiore et al. [[87](#page-18-5)] studied the efect of fller content and size on mechanical properties of Arundo donax fllers (ADFs) reinforced epoxy composites. They concluded that composites containing Arundo donax fllers display better tensile moduli, comparable to bending moduli, and lower strength characteristics as compared to pure resin. With the increase in particle size and content of ADFs, the storage modulus and loss modulus increased in the rubbery region. Islam et al. [[88](#page-18-6)] studied hemp fber reinforced epoxy composite with diferent fber contents ranging from 40 to 65 wt%. They noted that untreated long fber showed better strength at high fber contents.

Fig. 6 A SEM images of composites **a** BF **b** 2.5BF **c** 5BF **d** 10BF magnifcation ×1000. **B** The dynamic mechanical thermal properties (DMTA) curves of the composites. **C** Mechanical properties of composites [\[51\]](#page-17-30)

Fiber Surface Modifcation

A variety of chemical and physical methods are available to amend the structure and properties of the fber and increase the affinity between fiber and matrix. Fiore et al. [\[89\]](#page-18-7) investigated the reinforcement of untreated and alkalitreated kenaf fber in an epoxy matrix. When compared to neat resin both untreated and alkali-treated composites enhance mechanical properties. According to Mylsamy et al. [[90\]](#page-18-8), both length and alkali treatment of agave fiber epoxy composites show better adhesion for short fbers. Alkali treatment further increases fber wetting and fber-matrix interaction resulting in higher tensile compression, fexural, and impact strength. Yousif et al. [[91](#page-18-9)] analyzed that the

Fig. 7 Classifcation of natural fibers

Table 3 Physical and mechanical properties of different natural fibers [\[13](#page-16-10)[–15,](#page-16-11)

[54,](#page-17-33) [59](#page-17-38)]

bending strength of untreated kenaf fber-reinforced composite increases by 20% while alkali treatment of kenaf fber further improves the bending strength by 36% due to interfacial bonding between fber and epoxy matrix. Moreover, the porosity of the composite decreases with alkali treatment. Mahjoub et al. [\[92\]](#page-18-10) studied the effects of changing NaOH ratios and treatment time on kenaf fber epoxy composite. They found that 5% alkali treatment produces no tension on fber texture and structure when compared to 10% and 15%, and is optimum for kenaf fber treatment. Brodowsky et al. [[18\]](#page-16-14) while analyzing jute fber-reinforced composite discovered that combining alkali and silane coupling agents improves the composite's adhesion strength and mechanical properties. Lu et al. [\[93](#page-18-11)] prepared untreated, alkali-treated, and silane-treated bamboo fbers reinforced epoxy composites. They concluded that alkali and silane treatment boosts composite tensile strength and elongation at break. They also discovered that NaOH treatment of the bamboo fber increased the tensile strength and elongation at the break by 34% and 31%, respectively, compared to untreated cellulose reinforced composite. Likewise, the silane coupling agent also increases the tensile strength and elongation at the break by 71% and 53%, respectively. The efect of kenaf fber on epoxy composite was studied by Azwa et al. [\[94\]](#page-18-12), that the addition of untreated kenaf fbers increases thermal stability. But alkali treatment afects thermal stability, decreases degradation temperature of the composite, and produces less char than untreated fber composite. They also observed that increasing composite exposure time only affects the decomposition rate up to 150 °C and has a minimum role in the decomposition of the composite. Anbukarasi et al. [\[95](#page-18-13)] discovered that NaOH-treated lufa fber increases the thermal and mechanical properties of the epoxy composite. The composites containing particle fbers exhibited better water absorption. Hence, the particle fber-reinforced composites can be employed in damp places like washroom doors, car

Table 4 (continued)

Table 4 (continued)

interiors, etc. Alkali-treated agave fber composites have the lowest Tan δ at higher temperatures, according to Mylsamy et al. [\[90\]](#page-18-8), making it a good polymer composite for high temperatures. Zhang [\[96](#page-18-31)] examined the microstructure and thermomechanical characteristics of natural bamboo fbers at various NaOH concentrations. They reported that as the NaOH concentration increases, so does the thermal stability. The thermal performance of unmodifed and NaOHmodifed coconut fber reinforced epoxy composites was compared by Kumar et al. [[97\]](#page-18-32). They perceived that composites with treated coconut fbers as a reinforcement had better thermal stability. Liu et al. [[98\]](#page-18-33) studied the impact of chemically treated abaca fber on the transverse thermal conductivity of the composite. The results show better thermal conductivity for treated fber-reinforced composite related to untreated fber composite. Shih [\[99](#page-19-0)] examined the infuence of silane treatment on the mechanical and thermal behavior of bamboo reinforced epoxy composites and concluded that silane treatment of bamboo fber increases the thermal stability of the composite.

Bio‑fller/Particles Reinforced Epoxy Composites

Bio-fillers derived from natural resources are increasingly being employed to improve the composite material performance while lowering the costs. Kranthi et al. [[100\]](#page-19-1) prepared pine wood dust reinforced epoxy composites. They concluded that pine powder holds good fller characteristics to improve the sliding wear performance of the matrix. Moreover, they discovered that specifc wear rate depends on fller content, sliding velocity, and normal load. Biowaste -

reinforced epoxy resin composites, such as fish scale reinforced epoxy resin composites, have low permeability and increased micro-hardness. These composites show the potential to be used in applications such as automobile seat covers, coal-taking pipes in power plants, conveyor belts, pipes carrying pulverized coal in power plants, turbine blades, and low-cost domestic items [[101\]](#page-19-2). Shah et al. [[102\]](#page-19-3) fabricated acacia catechu particles reinforced epoxy composites and found an improvement of 14% and 94% in fexural and impact strength, respectively, with the insertion of 1 wt% acacia catechu particles. Rice husk reinforced epoxy composite was scrutinized by Rout et al. [[103](#page-19-4)] and they observed that rice husk particles enhanced the sliding wear resistance of the polymer composite. Narendar et al. [[104](#page-19-5)] noted that the chemical treatment increase crystallinity of coir pith resulting in improved compressive strength and storage modulus of the epoxy composite. Salasinska et al. [[105\]](#page-19-6) prepared a low-cost composite with enhanced mechanical and thermomechanical properties by reinforcing a large quantity of walnut shell powder into an epoxy matrix that could be used for low-performance application parts and applications. Shah et al. [\[106\]](#page-19-7), on the other hand, investigated alkalitreated walnut shell reinforced composites and discovered that the addition of treated walnut shells increased thermal stability, storage modulus, and crystallinity index. In another research, Owuamanam et al. [\[107](#page-19-8)] compared untreated and stearic acid (SA) treated eggshell reinforced bio-epoxy composite. They came up with a conclusion that increase in fller content afects tensile strength, bending strength, and impact toughness. However, the bending modulus increases with the increase in fller content. Shah et al. [[108\]](#page-19-9) observed that stearic acid-treated eggshell particles reinforced epoxy composites showed reduced brittleness, increase thermal stability, improved stifness, and greater glass transition temperature related to the pure matrix. Jabbar et al. [\[109\]](#page-19-10) analyzed the reinforcement of NaOH treated pulverized jute into the epoxy matrix. They found that the addition of alkalitreated pulverized jute increases glass transition temperature, storage modulus and lowers the height of the tangent delta peak of the composite.

Natural Fibers/Fillers Based Hybrid Epoxy Composites

Natural fibers and materials derived from renewable resources are afordable and readily available in large quantities, making them perfect for the production of low-cost composites. Each material possesses unique properties and, considerable advantages and disadvantages based on its structure and properties. Fiber hybridization is one of the most common and rapidly expanding technology for improving composite qualities. The advantages of one fber can balance the limitations of the other in a hybrid composite with diferent types of fllers. Hence, appropriate material design can yield a composite with a good balance in all attributes. Hybridization of bio-fllers with natural fbers is also a promising research direction $[110]$ $[110]$ $[110]$. Biofiber/filler hybrid reinforcements can modify material properties for the desired application and have the capacity to be used in every feld like automotive, building materials, packaging, electronics, and chemical resistance [\[53\]](#page-17-32). At the same time, environmental concerns have drawn researchers' attention of researchers to manufacture high-performance composites with the combination of diferent kinds of biofbers and fllers for [[111](#page-19-12)]. Considerable research has been done to reinforce natural fbers with other fllers to fabricate hybrid epoxy biocomposites. Some recent work done on natural materials hybrid epoxy composites is discussed below.

Hybridization of Natural Fiber Epoxy Composites

Hybridization of reinforcement fbers from natural resources with complementary characteristics, such as fiber/fiber or fiber/filler provides a set of properties that are difficult to achieve with a single material. Hybridization provides better properties as well as signifcant cost savings due to the use of low-priced raw materials. Sumesh et al. [[112](#page-19-13)[–114\]](#page-19-14) investigated and optimized the various factors stabilizing mechanical and wear properties by combining diferent natural fbers in diverse ratios. They found that adding of 30 wt% of sisal fber/pineapple fber enhanced the tensile strength from 26.19 to 33.48 MPa, fexural strength from 67.79 to 73.07 MPa, and impact strength from 55.31 to 64.88 J/m [\[112\]](#page-19-13). In another study using the grey relational approach, they noticed that the optimum ratios for sisal and banana fber in the epoxy matrix were 20% sisal, 15% banana, and 5% NaOH, under 10 MPa pressure, and 100 °C temperature. In addition, there was a strong relation between experimental and predicted grey correlational grades for analyzing factors infuencing mechanical properties [\[113](#page-19-15)]. Furthermore, they studied banana and coir fber reinforced hybrid epoxy composites and had recorded optimized mechanical properties for 20% banana /15% coir/5% alkali treatment/16 MPa pressure and 100 °C temperature [\[114](#page-19-14)]. The hybridization of hemp and fax fber with a total weight ratio of 30% in the epoxy matrix exhibited lower water absorption and higher mechanical properties with an increase in the percentage ratio of fax fber. The storage and loss modulus was found to be maximum for 15% flax + 15% hemp epoxy composites [[115\]](#page-19-16). The combination of flax and basalt fibers improved the impact properties of the fax fber-reinforced composite $[116]$ $[116]$ $[116]$. The hybridization of bamboo/kenaf fiber with a 50:50 mixing ratio in epoxy matrix presented good dynamic mechanical and thermal properties [\[117](#page-19-18)]. Kenaf/areca fber epoxy composite showed higher tensile, fexural, and hardness properties when kenaf fber was used as an outer layer while improving the impact and compressive strength as a core material in the hybrid composite [[118\]](#page-19-19). The addition of NaOH treated jute fber increases the mechanical properties of sisal fber reinforced epoxy composite [\[119](#page-19-20)]. Results demonstrate that adding about 50% sisal fber to banana epoxy composite decreases moisture absorption and increased the mechanical performance of the composite [[120\]](#page-19-21). The mechanical properties of hybrid composites were improved by adding woven jute fber to pure oil palm empty fruit bunches (EFB) composites. Hybrid composite had higher tensile and bending properties than those of EFB composite, but lower than jute fabric composite [[121](#page-19-22)]. The hybridization of Grewia optiva and Bauhinia vahlii fber in epoxy resin improves the physical and mechanical properties of the composite [[122](#page-19-23)]. The addition of lignin with hemp fber can increase the impact, tensile, and bending strength. The addition of lignin up to 2.5% w/w augments both bending and tensile modulus [\[123](#page-19-24)]. The mechanical and thermal properties of jute-epoxy composite increase while hygroscopicity reduces with the inclusion of banana fiber, up to 50% [[124\]](#page-19-25). The results obtained from dynamic mechanical analysis (DMA) and impact tests confrm that hybrid composite outperform non-hybrid composites in terms of residual bending strength and impact performance [[125\]](#page-19-26). Basaltfber reinforced hybrid composite can efectively upsurge the durability of biocomposite under salt fog environment conditions [\[126\]](#page-19-27). Gupta [[127](#page-19-28)] concluded that the hybridization of natural materials is a practical approach to escalate the dynamic mechanical properties of the composite. Further, the chemical treatment of fbers shows improved mechanical properties of epoxy hybrid composites than generally used fber composites [\[128\]](#page-19-29). Chaudhary et al. [[129\]](#page-19-30)

studied diferent combinations of natural fbers in epoxy polymer and observed better mechanical performance of hybrid composites. The hybrid composite containing jute, hemp, and flax fiber had the highest tensile strength, impact strength, and modulus. The maximum fexural strength for the hybrid composite was 86.6 MPa. Moreover, the chemical treatment increases the thermal stability of hybrid composites. Jute/curaua and jute composites presented better properties for the untreated conditions, while jute/sisal composite displayed better results under mixed treatment. However, the results of the untreated and mixed conditions jute/ramie composite depicted no significant difference [[130](#page-19-31)]. Hybrid composites possess better micro-hardness and impact strength [\[131\]](#page-19-32). Tensile, bending strength, and modulus increase by increasing the flax fiber content [\[132](#page-19-33)].

Bio‑fllers/Natural Fiber Reinforced Hybrid Epoxy Biocomposites

The use of organic and inorganic fillers in conjunction with natural fbers to make hybrid composites is becoming increasingly popular. The hybridization of bio-fllers with natural fber is an efective research methodology to modify the properties of the composite. The combination of biofllers with natural fber improves both mechanical and water absorption properties [[133](#page-19-34)]. Stocchi et al. [\[134\]](#page-19-35) prepared jute woven fabrics and fly ash particles reinforced hybrid composite. They determined that hybrid composite gave the best mechanical and fracture characteristics. Combining Azadirachta indica seed powder and spent Camellia Sinensis powder with jute fabric in an epoxy matrix increased the mechanical and thermal properties of the epoxy composite [[135\]](#page-20-0). Calcined eggshell particles and alkali-treated sisal fber reinforced hybrid epoxy composite can be used to make green composites because they have better insulation, water resistance, and mechanical properties than unreinforced epoxy [\[136\]](#page-20-1). Narendar et al. [\[104\]](#page-19-5) prepared epoxy hybrid composite employing coir pith and nylon fabric. They concluded that the incorporation of chemically treated coir pith into epoxy resin improved its storage modulus. Furthermore, the combination of coir pith with nylon fabric improved the storage modulus. Chemical fller modifcation and hybridization improve the composite's mechanical properties and water resistance [[137,](#page-20-2) [138](#page-20-3)]. Sumesh et al. [\[139](#page-20-4)] used banana fly ash, pineapple fly ash, and coir fly ash fillers $(1-4 \text{ wt\%})$ with 30 wt% of sisal $(S)/$ pineapple (P) hybrid fiber composites as a reinforcement in an epoxy matrix. They had recorded tensile strength of 23.78–33.79 MPa by the substitution of Banana fy ash, pineapple fy ash, and coir fy ash fillers, compared to hybrid natural fiber composites (30 wt) sisal/pineapple), which showed a strength of 20.45 MPa. Similarly, by adding biowaste fllers, the impact and fexural properties were improved up to 21.77% and 22.11%.

The addition of calcium carbonate powder to a bagasse fber reinforced epoxy composite improves the composite's bending and compression strength. Comparative analysis of the outcomes of chemically treated and untreated fbers reveals that the mechanical properties of chemically treated fbers reinforced hybrid composites are superior to those of untreated fber-reinforced composites [[140](#page-20-5)]. The bending, tensile, impact strength, and thermal stability of banana/fax, banana/kenaf, sisal/fax, and sisal/kenaf composites had signifcantly improved by diferent bagasse ash (BGA) content [[141](#page-20-6)]. Hybrid composites with various ratios of eggshell particles with jute fber epoxy matrix have been studied. The fndings indicate that 10% eggshell powder composite gives higher impact, tensile and hardness properties [\[142\]](#page-20-7). Various mechanical and water absorption characteristics of groundnut and lufa fber reinforced epoxy hybrid composites with diferent ratios of fber content ranging from 10% to 50% have been studied. The results show that alkali-treated luffa fber composite having 40% fber content outperforms other fiber content composites. Luffa and groundnut reinforced hybrid composite exhibits higher mechanical properties than composite reinforced with only luffa fiber $[143]$. The mechanical and morphological properties of the composite improve signifcantly by adding palm oil nanofller into kenaf fber reinforced epoxy composite [[144\]](#page-20-9). The addition of fy ash particles with natural cellulosic fber improves the composite's tensile characteristics and surface quality. Fly ash particles behave as fllers, additives, surface treatment media, and cellulosic fber as reinforcement elements to create a sustained smooth treatment of composite material. The produced hybrid composite is stronger than wood and plastic composites and can perform multiple functions [\[145](#page-20-10)].

Applications of Natural Fibers/Fillers Based Epoxy Composites

Automotive Industry

The use of natural fbers composites is mostly limited to nonstructural applications due to their brittle nature and are therefore primarily used in interior parts of the auto motives, door panels, packaging, cases, chairs, tables, and ironing boards. In 2012, the automobiles sector used more than 98% of the natural fber-based composites developed in the European Union [\[146](#page-20-11)]. Rieter Company replaced glass fbers with abaca fber and began commercial production of spare wheel pan covers for the Mercedes A-class car with reduced cost and weight, having a negligible effect on quality [\[147\]](#page-20-12). Rieter is also committed to developing the most advanced lightweight and eco-friendly sound packaging to enhance the recycling rate of individual components and minimize energy consumption and environmental effects.

Daimler [\[148\]](#page-20-13) manufactured sisal and flax fiber-reinforced epoxy composite with augmented mechanical properties and 20% lighter weight for the door panels of the Mercedes Benz E class. Natural cellulose fabric reinforced epoxy composites with an average strength higher than required for automotive threshold strength (25 MPa) have developed. Hence, bark fabric reinforced epoxy composites can be used for the interior parts of automobiles [[149\]](#page-20-14). Mahdi et al. [\[150\]](#page-20-15) studied date palm leaf fber (DPLF) reinforced epoxy composite. They noted that date palm leaf fiber reinforced epoxy composites had a high energy absorbing capacity and proved to be a potent candidate as environmentally friendly materials for automotive applications. Jute fber epoxy composites gave a better mechanical performance than polyester fberreinforced composites and are preferable for application in the automobile industry [[151\]](#page-20-16). Sumesh et al. [\[152\]](#page-20-17) prepared pineapple/Flax epoxy composites with outstanding mechanical properties. The Pineapple/Flax natural fbers with peanut oil cake fller might be employed as a reinforcement material for making friction composites for brake pad applications. Calotropis gigantea fbers reinforced epoxy composites have demonstrated tribological performance in various engineering sectors, such as brake pads and brake discs [[153\]](#page-20-18).

Building Materials

Growing research in biocomposites demonstrates the signifcance of bio-fbers and fllers, which have applications in nearly every feld. Biocomposites developed with natural fbers are broadly used in building components, transport sectors, furniture, packaging, marine, sports, and aerospace industries [\[56](#page-17-35), [154](#page-20-19)]. Natural fiber- reinforced structural insulated panels (NSIPs) have shown better mechanical performance than oriented strand board structural insulated panels (OSB SIPs) and can be used to replace OSB SIPs [\[155\]](#page-20-20). Coir fbers are primarily employed to make a variety of floor decoration materials but the automotive industry also uses coir fber for seat covers and to support seats [[156](#page-20-21)]. Bagasse fber composites can make medium-density fiberboard, that can be used in train cars [\[157,](#page-20-22) [158](#page-20-23)]. Natural fber composite sheets outperform commonly used concrete and steel-made sheet fles [[159](#page-20-24)]. Likewise, natural fber hybrid composites can be used specifcally for cost-efective residential and interior parts [[160](#page-20-25)]. Banana fberreinforced composites are better alternatives and new materials used in low-cost household furniture, replacing wood, plastic, and conventional metallic, and non-metallic materials. Banana fber-reinforced epoxy resin composites have been used in the design and manufacturing of multifunctional work tables. Woven fabric made from the natural banana fber can be successfully reinforced in epoxy resin to develop a household stand for telephone $[161]$ $[161]$ $[161]$. Alkali-treated Teff straw fiber incorporated epoxy composites can be used for lightweight and semi-structural applications [\[162](#page-20-27)].

Medical Applications

Biocomposites materials also have a wide range of applications in the medical feld. They can be used as an alternative to titanium, stainless steel, zirconium, and cobalt chrome, for fracture fxation in orthopedics [[163\]](#page-20-28). Woven natural fberreinforced epoxy materials are highly recommended in applications like patient positioning and support devices. Magnetic resonance imaging (MRI), X-ray computed tomography (CT) and radiotherapy results reveal that cotton and bamboo fber reinforced epoxy composites have minimum impact on resulting images and are suitable for use in these applications [\[164\]](#page-20-29). Flax and ramie fber reinforced epoxy composites provide comparable properties to the human femur and tibia. Moreover, when compared to other accessible materials for orthopedic implants, such as ceramic and metallic plates, these natural fber-based composites are biocompatible, biostable, and ecofriendly. Therefore these hybrid composites are recommended in orthopedic implant applications [\[165](#page-20-30)].

Other Applications

Although natural materials-based epoxy composites are most commonly used in the automotive and construction industries, they have proven potential in almost any feld. In a recent study, fax fber-reinforced epoxy made pipes have demonstrated signifcantly higher internal pressure than the conventional pipe pressure range for domestic water distribution, and long-distance water transfer. It suggests a signifcant potential for application of fax fber-incorporated epoxy pipes [[166\]](#page-20-31). The fshing rod is made of materials that remove nanocellulose from vegetable roots [\[158](#page-20-23)]. The thermal decomposition property of coir particle composites is high enough to improve the conductivity of the sample, making it a preferable choice to be used in applications such as electrical and sensor devices [[167\]](#page-20-32). Natural materials-based composites also have the ability to be used in musical instruments [\[168\]](#page-20-33). Natural fber-based hybrid composites can be employed to prepare water and chemical tanks as they show chemical resistance [\[169](#page-21-0), [170](#page-21-1)]. Epoxy composite reinforced with mallow fiber gives the same ballistic efficiency as Kevlar fber and is used in military applications for personal ballistic protection [\[62](#page-18-14)].

Conclusion

This review emphasizes the importance of natural materials in high-performance composites over synthetic materials. The physical and chemical properties of these fbers, as well as the difficulty of making composite materials, are briefy discussed. Natural fbers show high capacity in producing new composites with high performance having a minimal environmental impact. However, more research is needed to eliminate variations in natural fber performance attributes and improve the interface between natural fbers and polymer matrix. Furthermore, because natural fber composites have low mechanical properties compared to synthetic fbers, they are used for nonstructural applications. Hence, for structural applications, hybridization of more than one material from natural resources in polymer composites can increases the mechanical and thermomechanical properties that meet the required standards of the civil engineering materials feld.

Furthermore, the physical, thermal, and mechanical properties of natural fber/fllers reinforced epoxy composite/hybrid composite are ideal for the fabrication of highperformance engineering materials. The reinforcement of natural fbers and fllers is an impressive way to improve the mechanical properties of epoxy composites. Bio-fllers reinforced epoxy composites can be a better substitute synthetic ones in engineering applications requiring low to medium structural strength.

To overcome the variation in fbers advanced retting techniques should be applied to produce high-quality fbers and a better classifcation of the fbers; in terms of their diameter and length based on their end-use to minimize irregularity. Moreover, fber surface sizing with diferent nanomaterials can enhance the interfacial properties of the composites and needs further exploration. Similarly, a combination of various materials (dual coating) on the surface of natural fbers can improve the interfacial adhesion of the composite and hence can provide better strength and toughness, allowing it to be used in structural applications. Furthermore, adding hybrid materials and surface coating with functional materials can improve fame retardancy, electromagnetic shielding performance, water-resistance, tribology, and thermal properties and could serve as a substitute for conventional materials. In addition, diferent natural fbers, which have yet to be studied as a reinforcement, must also be taken into account.

Acknowledgements This work was supported by the National Natural Science Foundation of China (Project No.51773048), Natural Science Foundation of Heilongjiang Province (Project No. E2017022).

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no confict of interest.

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