Review Article

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Effect of fiber treatment on physical and mechanical properties of natural fiber-reinforced composites: A review

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Abstract: Due to environmental and financial concerns, there is a growing demand for composite materials in a wide range of industries, including construction and automotive industries. In 2020, the market for wood plastic composites was estimated to be worth $5.4 billion. By 2030, it is expected to have grown to $12.6 billion, with a compound annual growth rate of 8.9% between 2021 and 2030. The fundamental disadvantage of reinforced composites by natural fibers is the different nature of the hydrophilic lignocellulosic and the hydrophobic thermoplastic polymers, although natural fibers would lower total costs. These composites typically fail mechanically as a result of fiber debonding, breaking, and pull-out. In a fiber-reinforced composite, the matrix's function could be described as distributing the force to the added fibers using interfacial shear stresses. A strong connection between the polymeric matrix and the fibers is necessary for this procedure. Weak adhesion at the interface prevents the composite from being used to its maximum potential and leaves it open to attacks from the environment that could damage it and shorten its lifespan. Poor mechanical performance is caused by insufficient adhesion between hydrophobic polymers and hydrophilic fibers in natural fiber-reinforced polymer composites. Consequently, during the past 20 years, a variety of chemical, thermal, and physical methods have been employed to address these issues. These methods largely concentrated on the grafting of chemical groups that could enhance the interfacial contacts between the matrix and natural fibers. This review article aimed to give information on several types of fiber treatments and natural fiber-treated composites with a specific focus on their physical and mechanical properties.

Keywords: treatment, natural fiber, composite, polymer, physical properties, mechanical properties

1 Introduction

Wood plastic composite (WPC) is a synthetic composite made from a polymer matrix imbedded with wood fiber/wood flour. Thermosettings and thermoplastics are the two main categories of polymers. High-density polyethylene (HDPE), polypropylene (PP), and polyvinyl chloride (PVC) are the most often used thermoplastic polymers as a matrix for natural fibers (Table 1), whereas polyester, epoxy, phenolic, and resins are the most widely employed thermoset materials (Table 2) [1]. The fact that thermoplastic polymers can be cut, screwed, and nailed using tools previously used for wood construction and their processing temperatures, which are often lower than wood's thermal degradation temperature (180–200°C), make them appealing for WPCs, where HDPE (83%), PP (9%), and PVC (7%) are the most common thermoplastic polymers used in WPCs [2].

In response to the rising demand for biodegradable, renewable, and sustainable materials, this class of composite material, also known as "green composites," has emerged as a significant group of engineering materials. Due to the existence of wood as an organic material, WPCs have a sustainable nature that allows for environmental conservation and waste reduction. Such materials also offer substantial benefits that justifiably support their use [3]. Although the majority of polymers are non-biodegradable, wood fibers (as well as the other main natural-organic fillers including waster

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Table 1: Properties of typical thermoplastic polymers used in natural fiber composite fabrication (adapted from ref. [1])

Property	PP[a]	LDPE	HDPE	PS	Nylon 6	Nylon 6,6
Density ($g \cdot cm^{-3}$)	0.899–0.920	0.910–0.925	0.94–0.96	10.4–1.06	1.12–1.14	1.13–1.15
WA-24 h (%)	0.01–0.02	<0.015	0.01–0.2	0.03–0.10	1.3–1.8	1.0–1.6
T_g (°C)	−10 to −23	−125	−133 to −100	N/A	48	80
T_m (°C)	160–176	105–116	120–140	110–135	215	250–269
Heat deflection Temp (°C)	50–63	32–50	43–60	Max. 220	56–80	75–90
Coefficient of thermal expansion ($mm^{-1} \cdot mm^{-1} \cdot °C \times 10^5) \cdot cm^{-1}$	6.8–13.5	10	12–13	6–8	8–8.86	7.2–9
Tensile strength (MPa)	26–41.4	40–78	14.5–38	25–69	43–79	12.4–94
Elastic modulus (GPa)	0.95–1.77	0.055–0.38	0.4–1.5	4–5	2.9	2.5–3.9
Elongation (%)	15–700	90–800	2.0–130	1–2.5	20–150	35–>300
Izod impact strength ($J \cdot m^{-1}$)	21.4–267	>854	26.7–1,068	1.1	42.7–160	16–654

[a]PP = polypropylene, LDPE = low-density polyethylene, HDPE = high-density polyethylene and PS = polystyrene.

Table 2: Properties of typical thermoset polymers used in natural fiber composites (adapted from ref. [1])

Property	Polyester resin	Vinyl ester resin	Epoxy
Density ($g \cdot cm^{-3}$)	1.2–1.5	1.2–1.4	1.1–1.4
Elastic modulus (GPa)	2–4.5	3.1–3.8	3–6
Tensile strength (MPa)	40–90	69–83	35–100
Compressive strength (MPa)	90–250	100	100–200
Elongation (%)	2	4–7	1–6
Cure shrinkage (%)	4–8	N/A	1–2
WA (24 h@20°C)	0.1–0.3	0.1	0.1–0.4
Izod impact strength ($J \cdot m^{-1}$)	0.15–3.2	2.5	0.3

agriculture fibers) are usually added to the polymer matrix up to 40–70% by weight and reduce non-biodegradable proportion of WPC significantly [4]. Furthermore, it is possible to use waste or recycled polymer materials to produce WPCs with the minimum negative effect on the environment. Finally, the thermoplastic-based green composites are recyclable and more biodegradable due to their polymer features and natural fiber content, respectively.

The benefits of wood flour include its low cost, lightweight, and accessibility, making it an economically friendly material that can be used in composites [5–8]. The construction and automotive industries are where WPCs are most commonly used, but they are also employed in packaging, the creation of different home furnishings, office equipment, and other goods [5,6,9]. Salemane and Luyt [10] investigated how adding wood flour improved the mechanical properties of a composite made of wood and PP. Since fibers have substantially greater strength and stiffness amounts than matrices, adding fibers to a polymer matrix usually significantly improves the tensile properties of composites.

It is imperative to create alternative, affordable, and ecologically friendly natural fiber sources for plastic composites due to rising wood pricing and demand for traditional wood sectors. The composite's mechanical, thermal, and other properties are quickly enhanced by the plant fiber utilized as a reinforcing material [11]. For their persuasive characteristics, including cheap prices, lightweight, low density, biodegradability, recyclability, high stiffness and strength, and renewable features, natural fibers are favored against synthetic fibers (Table 3) [12]. Plant-based natural fibers including flax [13,14], hemp [15,16], jute [17,18], sisal [19,20], kenaf [21,22], bagasse [11,23,24], banana [25,26] coir [27,28], plantain [29–31], and pineapple fibers [32,33] have been adopted by composite sectors as replacements for synthetic fibers in an attempt to substitute them due to their numerous benefits during the recent decades [34].

Natural fibers have many inherent drawbacks, including weak compatibility with polymer materials. The hydrophilic feature of the natural fiber that led to weak adhesion and moisture absorption causes weak compatibility in biocomposites. As a result, natural fibers must be pretreated in order to increase the biocompatibility between the fibers and matrix by the activation of fiber's hydroxyl groups [35]. Pre-processing modifications of composite elements are performed to prevent the fibers' hydrophilicity and to improve the interfacial characteristics of polymer and natural fibers. Chemical, mechanical, and thermal treatment methods represent the three basic types. Although chemical treatment is frequently employed, it has negative environmental effects. Hence, environmentally acceptable alternatives to chemical treatment including mechanical and thermal methods were recommended [36]. The fiber modification aims to increase the fibers' hydrophobicity, interfacial connection between the fiber and matrix, roughness, and wettability as well as to reduce

Table 3: Physical and tensile properties of natural fibers and glass fibers (adapted from ref. [56])

Fiber type	Diameter (μm)	Relative density (g·cm^{-3})	Tensile strength (MPa)	Elastic modulus (GPa)	Specific modulus (GPa × cm^3·g^{-1})	Elongation at failure (%)
E-glass	<17	2.5–2.6	2,000–3,500	70–76	29	1.8–4.8
Abaca	—	1.5	400–980	6.2–20	9	1.0–10
Alfa	—	0.89	35	22	25	5.8
Bagasse	10–34	1.25	222–290	17–27.1	18	1.1
Bamboo	25–40	0.6–1.1	140–800	11–32	25	2.5–3.7
Banana	12–30	1.35	500	12	9	1.5–9
Coir	10–460	1.15–1.46	95–230	2.8–6	4	15–51.4
Cotton	10–45	1.5–1.6	287–800	5.5–12.6	6	3–10
Curaua	7–10	1.4	87–1,150	11.8–96	39	1.3–4.9
Flax	12–600	1.4–1.5	343–2,000	27.6–103	45	1.2–3.3
Hemp	25–600	1.4–1.5	270–900	23.5–90	40	1–3.5
Henequen	—	1.2	430–570	10.1–16.3	11	3.7–5.9
Isora	—	1.2–1.3	500–600	—	—	5–6
Jute	20–200	1.3–1.49	320–800	30	30	1–1.8
Kenaf	—	1.4	223–930	14.5–53	24	1.5–2.7
Nettle	—	—	650	38	—	1.7
Oil palm	—	0.7–1.55	150–500	80–248	0.5–3.2	17–25
Piassava	—	1.4	134–143	1.07–4.59	2	7.8–21.9
PALF	20–80	0.8–1.6	180–1,627	1.44–82.5	35	1.6–14.5
Ramie	20–80	1.0–1.55	400–1,000	24.5–128	60	1.2–4.0
Sisal	8–200	1.33–1.5	363–700	9.0–38	17	2.0–7.0

moisture absorption, which will improve the composites' mechanical properties [37–41].

An overview of current developments in the field of natural fiber-treated composites is given in this study, and the effect of various treatments on the mechanical and physical characteristics of composites reinforced by natural fibers is highlighted.

2 Thermal treatment

Thermal treatment of wood, which significantly alters its physical and mechanical properties, is one of the most significant wood treatment techniques [42]. In recent years, the application of thermal modification has accelerated, and it is still expanding as an industrial technique [43]. Normal operating temperatures for thermal treatment are 180 to 260°C. The wood structure is not considerably affected by temperatures below 140°C, while unfavorable degradation occurs at temperatures above 260°C [44]. Wood's chemical compounds are impacted by heat treatment. The temperature of the treatment has been discovered to have a greater impact on chemical modifications than duration [45].

The chemical modifications that take place in wood by thermal treatment have a number of positive effects on the physical characteristics of wood, including less swelling and shrinkage, improved durability to biological deterioration, alteration in color, and lower equilibrium moisture content. Thermally treated timber has a significant and substantial drawback because its mechanical features deteriorate, limiting its utilization for strength-bearing purposes, and its MOE decreases [46].

Ayrilmis *et al.* [47] studied how thermal treatment affected the mechanical futures and dimensional endurance of WPCs produced by the flat press method. The dry mixture of *Eucalyptus camaldulensis* and 50% PP powder was used to make the WPC samples in an autoclave for 20 and 40 min at three different temperatures (120, 150, and 180°C) by employing a standard flat press procedure in a laboratory environment. The study confirmed that depending on the temperature and time circumstances, the thermal treatment of the wood fibers resulted in decreases of 60 and 31% in the TS and water absorption (WA) after 28 days of water soaking, respectively (Table 4). This was mostly due to the thermal treatment's hydrolysis of hemicelluloses. The TS and WA of the WPC panels were significantly affected by the thermal treatment of the wood fibers, especially over 150°C for 40 min. However, the treatment temperature had a greater impact on the TS and WA than the treatment duration (Table 4) [47]. Based on the duration and temperature circumstances, the modulus of elasticity, rupture, and the internal bond (IB) strength of the wood fibers treated by this method fell by 5 to 19%, 7 to 22%, and 9 to 28%, respectively, while the screw withdrawal resistance reduced from 3

Table 4: The relationship between the WPC panels' physical characteristics and the thermal treatment of the wood fibers (the ratio of wood fiber to PP: 50:50 by wt) (adapted from ref. [47])

Thermal-treatment level (°C, min)	Physical properties							
	Density ($g \cdot cm^{-3}$)	Thickness swelling (%)				WA (%)			
		1-day	7-days	14-days	28-days	1-day	7-days	14-days	28-days
Untreated reference	0.82	3.98A[a]	5.06A	7.82A	8.15A	6.45A	16.53A	24.12A	25.94A
120,20	0.80	2.94B	3.82B	4.97B	5.20B	4.82B	14.72B	21.59B	23.18B
120,40	0.82	2.64BC	3.48BC	4.59B	4.84B	4.39B	14.09B	20.96B	21.46B
150,20	0.81	2.28C	3.15CD	4.27B	4.51C	3.85C	13.37BC	19.77C	21.12C
150,40	0.82	2.05CD	3.05D	3.81C	4.02C	3.47CD	12.96C	19.25C	20.58D
180,20	0.83	1.84D	2.28E	3.17D	3.39D	3.15D	10.88D	17.92D	18.64E
180,40	0.81	1.72D	2.05E	3.06D	3.25D	2.91E	9.79E	16.95E	17.84E

[a] Letters show Duncan's multiply range test ($p < 0.01$).

to 11% (Table 5). The IB outcomes depicted that raising the intensity of the thermal treatment had a negative impact on the mechanical interlocking between the polymer and wood fibers [47].

The creation of soluble acidic compounds, including acetic acid and formic acid, from the decomposition of hemicellulose may be responsible for the WPC panels' loss of mechanical features [48]. The crystalline structure of cellulose (long chain) is broken down into shorter pieces by these acids, which speed up the cellulose depolymerization. Additionally, as the temperature and length of the treatment increase, C–C and C–O links cleave within the polymer itself. As a result, the copolymer system of lignin–-hemicellulose separates, and hemicelluloses and amorphous cellulose are depolymerized [49]. The modulus of rupture (MOR) and modulus of elasticity (MOE) of the wood may be influenced by cellulose depolymerization and the decrease of its length chain [50]. Other studies also showed similar outcomes [51–54].

According to Hu *et al.* [55], the mechanical and physical characteristics of the bamboo plastic composite were examined in relation to the influence of bamboo fibers that had been modified by utilizing a vacuum-heated method. The outcomes showed that the bamboo fibers' surface polarity and hemicellulose content could be decreased after the vacuum heat treatment, enhancing the interface compatibility between the bamboo fiber and polyethylene. The composite's WA after 24 h was reduced by 73.01% when the temperature was 160°C in comparison with the control group, and the composite's thickness swelling after 24 h was the lowest point at 180°C, which, compared to the control group, was 71.47% lower. The mechanical strength of the composites also exhibited a pattern of raising and reducing as a function of vacuum heat treatment temperature. The bending strength and modulus of the composites reached the highest amount at 180°C, increasing by 39.91 and 21.77%, respectively, compared to the control group. Thus, the findings showed that adding a compatibilizer and

Table 5: The relationship between the WPC panels' mechanical characteristics and the thermal treatment of the wood fibers (the ratio of wood fiber to PP: 50:50 by wt) (adapted from ref. [47])

Thermal-treatment level (°C, min)	Mechanical properties			
	Modulus of rupture ($N \cdot mm^{-2}$)	Modulus of elasticity ($N \cdot mm^{-2}$)	IB strength ($N \cdot mm^{-2}$)	Surface SWR (N)
Untreated reference	25.9A[a]	2682.4A	0.95A	1,010A
120,20	24.7B	2483.8B	0.86B	982AB
120,40	24.2BC	2434.5B	0.83BC	976AB
150,20	23.7CD	2358.3C	0.77CD	951BC
150,40	23.0D	2264.2D	0.75D	924CD
180,20	21.7 F	2225.8D	0.71DE	915CD
180,40	21.0 F	2102.5E	0.68E	894D

[a]Letters show Duncan's multiply range test ($p < 0.01$).

thermal treatment of the composites could greatly increase their bending strength and water resistance, but it may also somewhat diminish their impact strength [55].

2.1 NaOH

Alkali modification is advantageous for cleaning the surface of the fiber, altering the surface chemistry, reducing moisture absorption, and raising the roughness of the surface. The process helps clean away contaminants and waxy particles from the surface of the fiber and produces a smoother texture that makes mechanical interlocking easier. The chemical connection between the reinforcement and resin is also strengthened by the cleaned fiber surface [56].

Bharath and Basavarajappa [57] evaluated phenol-formaldehyde (PF) composites containing coconut tree leaf sheath (CLS) for their fire resistance capability. To create CLS samples, a conventional hot press was employed with 60 wt% untreated and treated CLS fibers with 5% NaOH. The results of the flammability test showed that the treated composites' mass loss rate and flame propagation rate had been reduced, while their flame resistances had been increased. The limiting oxygen index test also showed that alkali-treated composites needed more oxygen to burn than untreated composite material [57].

Guo *et al.* [58] reported that following alkalization, some pectin, hemicellulose, lignin, and other low-molecule contaminants dissolve and are eliminated, along with a significant number of hydrogen bonds. The fiber's surface gets rougher, which improves the fiber's ability to bind to the resin at the interface. The remaining material is primarily composed of wood fiber. The hydroxyl of the crystalline wood fiber is exposed to the alkaline solution, which causes the wood fiber to become fluffy and better able to attach to the coupling agent. This reduces the hydrophilicity of the wood fiber and enhances the interface's adhesion properties and wettability. The alkalization process reduces the fiber split, decreasing its diameter, while enhancing its aspect ratio, resulting in an expansion of the fiber's interface with the matrix [58].

Li *et al.* [59] investigated the possibilities of adopting several kenaf core pre-treatment using HDPE. The study found that pre-treatment had little impact on fungus resistance and that the duration of pectinase or cellulase enzyme pre-treatment should be reduced to 30 or 60 min, respectively. The mechanical performance of a 60/40 kenaf/HDPE mixture was significantly enhanced by pre-treating the core particles with 1% NaOH or 1% HCl for 60 or 30 min, respectively. Pre-treating the kenaf fibers for 1 h with NaOH resulted in increases in MOE and MOR of approximately 50 and 82%, respectively [59].

Corn husk fiber (CHF) and recycled polystyrene foam (rPS) composites' tensile, thermal, WA, and morphologies were studied by Chun *et al.* [60] in relation to their fiber loading and alkaline treatment. According to the tensile test results, rPS/CHF composites' strength and modulus have increased by an average of 26 and 13%, respectively. A portion of the hemicellulose, lignin, and other compounds were eliminated from CHF after the alkaline treatment, and the fibers' surface roughness was raised. The interfacial interlock between the treated fibers and the rPS polymer was enhanced. Because of this, there was an improvement in the stress transmission between the fiber and matrix, leading to composites with greater strength and modulus. Similar findings were observed in other works as well [61,62]. Compared to untreated rPS/CHF composites, the average WA is 29% lower in treated rPS/CHF composites. Additionally, Chun *et al.* [63,64] discovered that composite materials with treated natural filler will absorb less water.

Baffour-Awuah *et al.* [65] investigated the effects of treating wood flour with alkali and ultrasound methods on the mechanical characteristics of wood polypropylene composites. The findings demonstrated that lignin and hemicellulose were eliminated from the wood by the alkali treatment, and the amount of hydroxyl groups on the surface of the cellulose increased. This technique was made more effective by ultrasonic modification. When PP grafted with maleic acid was utilized as a compatibilizer, mechanical features of WPC specimens demonstrated that alkali treatment increased both composite strength and modulus. While stronger modulus results from the elimination of hemicellulose and lignin, which are less stiff than cellulose, the strength improvement is caused by enhanced adhesion between the matrix and the fiber. Composites having 1 and 3% NaOH-modified wood showed improvements in average tensile modulus of 13% (from 4.27 to 4.83 GPa) and 20% (from 4.27 to 5.13 GPa), respectively, as comparison to composites manufactured with untreated wood. It has been definitively proven that chemical treatment of wood combined with ultrasonic improves the mechanical properties of composites more effectively than chemical treatment [65]. The alkali solution penetrates the wood flour by utilizing ultrasound, which also gives each particle uniform vibrations. By combining with alkali treatment, ultrasound expedites the elimination of lignin from wood and boosts the amount of hydroxyl groups on the cellulose's surface. The particle size of the wood is also decreased by the application of ultrasound. Thus, all of the mentioned processes have an impact

on the stiffness and strength of WPCs produced by alkali-treated wood [65].

Cui *et al.* [66] investigated the role of weight fraction and wood fiber length, as well as three surface modifications, including silane, alkaline, and alkaline + silane methods, on the mechanical features of wood recycled plastic composites (WRPCs). The research showed that as the amount of wood fiber rose, flexural strengths increased and impact strengths decreased. Although the flexural modulus improved with longer fibers, the impact and flexural strengths reduced. The strongest mechanical features were observed in a composite composed of wood fibers treated by alkaline + silane methods along with MAPP. According to Figure 1, all three surface treatments enhanced the WRPC materials' flexural strength in comparison with unreinforced samples. Although the findings for the silane treatment are relatively close to those for the combined treatment, the alkaline + silane-treated composites demonstrated the greatest flexural strength when compared to the other two types of composites. The greatest improvement was about 27% (50 wt%). Because of the complex microstructure of wood fiber, a surface treatment method may only be able to slightly alter the surface properties, which results in a limited increase in flexural strength [67]. The flexural modulus and surface treatment trends are quite close to flexural strengths, where 50 wt% treated wood fiber by alkaline + silane had the highest flexural modulus (Figure 2) [66].

Nam *et al.* [68] produced composite material using treated coconut shell fiber and polylactic acid after soaking it in a 5% NaOH solution for 72 h. In comparison to untreated coconut shell fiber, the research revealed that the shear strength and tensile strength had improved by 72.8%.

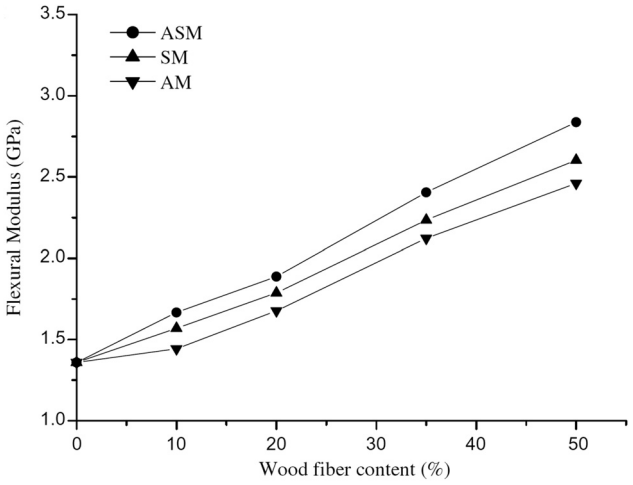

Figure 2: Flexural modulus of WRPC material as a function of wood fiber fraction with different surface treatment methods (adapted from ref. [66]).

Rajeshkumar *et al.* [69] evaluated the effects of four levels of NaOH concentrations (5–20%) on the mechanical characteristics of epoxy composites reinforced by *Phoenix* sp. fibers. In comparison to untreated fiber composites, the results showed that the treated fibers had better interfacial adhesion with polymeric matrix, decreased mechanisms of failure (including fiber debonding and pull-outs), and had improved mechanical characteristics [69]. Figure 3 shows that reinforced specimens using 15% treated fiber have the highest tensile modulus (589.12 MPa), which is 18.25% higher than the untreated composite. According to the study's findings, composites with 15% treated fiber have the best flexural characteristics (Figure 4), and their flexural strength (143.65 MPa) is much higher than that of the control samples

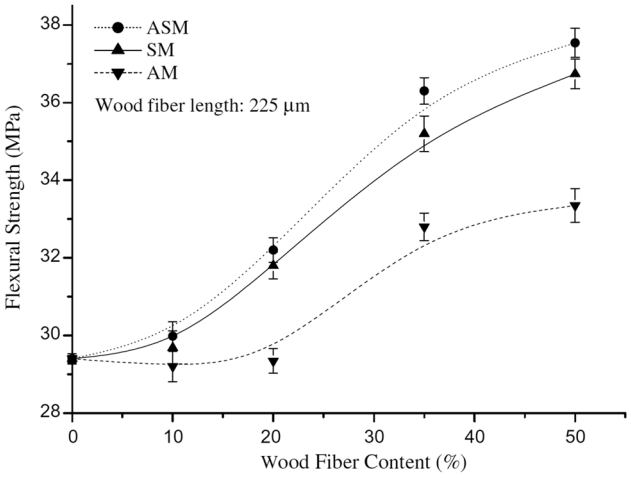

Figure 1: Flexural strengths of wood fiber composites as a function of wood fiber content with different surface treatment methods (adapted from ref. [66]).

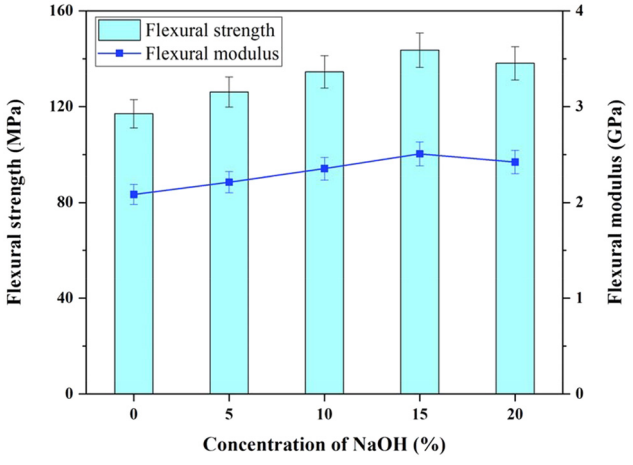

Figure 3: The effect of NaOH treatment on tensile properties of reinforced composites (adapted from ref. [69]).

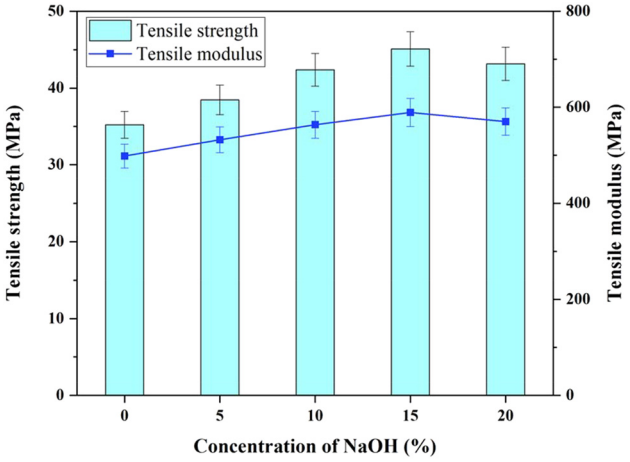

Figure 4: The effect of NaOH treatment on flexural properties of reinforced composites (adapted from ref. [69]).

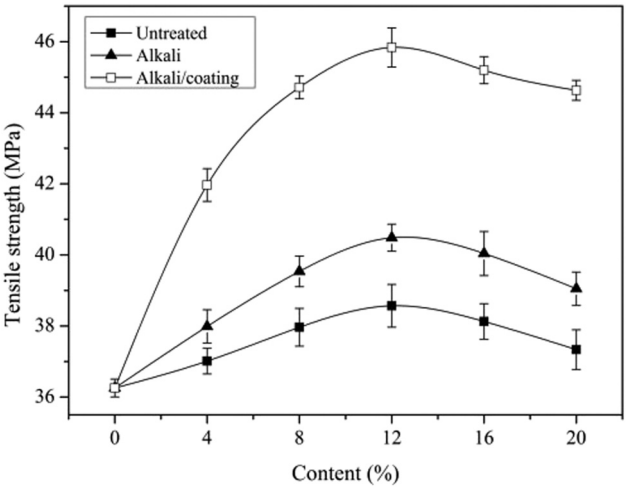

Figure 6: Tensile strength of composites (adapted from ref. [70]).

(117.05 MPa). It was determined that the qualities of this composite material complied with the requirements of EN standards 312-2 and 312-3 and, therefore, could be utilized for industrial purposes such as car panels [69].

2.2 Polyvinyl alcohol (PVA)

A type of secure, eco-friendly, and water-soluble material that can be employed as fiber modification method is PVA. PVA units that have been somewhat alcoholic have both hydrophilic and hydrophobic OH groups and molecular chains, respectively, that are well compatible with the hydrophilic nature of cellulose. PVA's hydroxyl groups can be bonded by borax, formaldehyde, and other chemicals. In order to increase the interfacial adhesion between natural fiber and polymer matrix, PVA can be employed to treat the fiber [70].

To assess the mechanical characteristics and water uptake of the composites, Hu *et al.* [70] coated the sisal fibers with an alkali + PVA using an ultrasonic process. The sisal-reinforced composites were manufactured using a twin-screw extruder. The study demonstrates that sisal fiber and HDPE can effectively be treated with an alkali/PVA coating compound to increase the interfacial adhesion, enhance the mechanical characteristics of the composite, and decrease WA. The interfacial adhesion can be clearly seen in the micromorphology of the composites' cross sections. The interfacial adhesion between sisal fibers and the HDPE polymer has been considerably enhanced following surface modification, which is also supported by the scanning electron microscope (SEM) photos of the cross section and other scientists [71–75]. As depicted in Figure 5(a), weak mechanical property is often caused by the insufficient adhesion of sisal fibers to HDPE polymer. Figure 5(b) illustrates that the interfacial bonding enhances as the space between the alkali-treated fibers and polymer reduces. As shown in

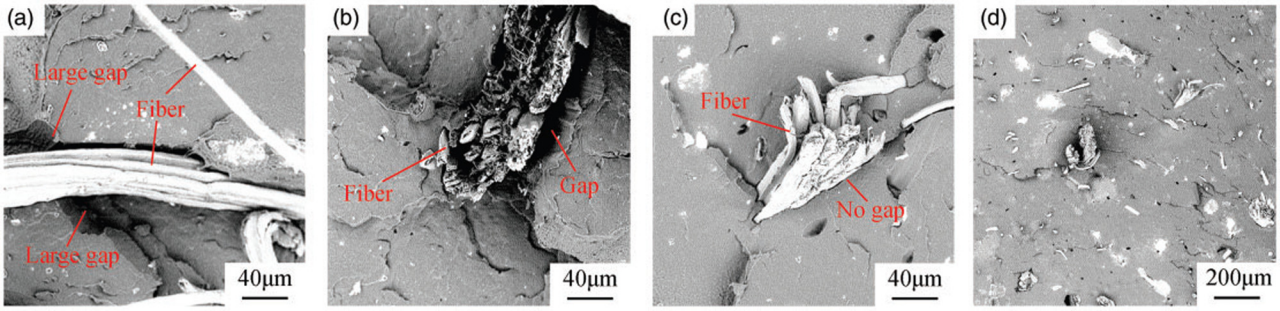

Figure 5: SEM images of sections of untreated sisal composites (a), alkali-treated sisal composites (b), and alkali/PVA coating compound-treated sisal composites (c) and (d). SEM: scanning electron microscope; PVA: polyvinyl alcohol (adapted from ref. [70]).

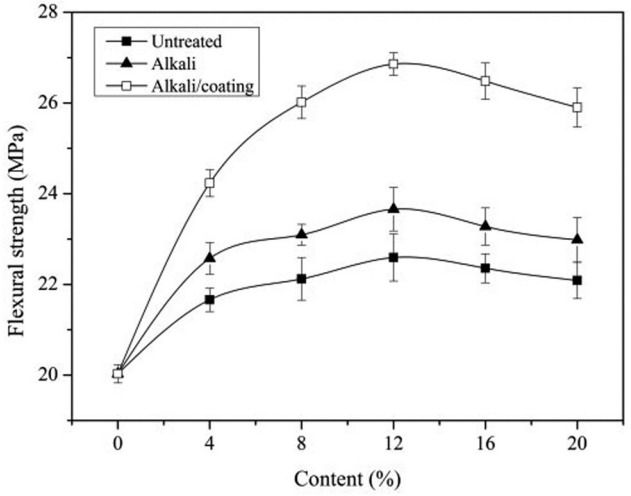

Figure 7: Flexural strength of composites (adapted from ref. [70]).

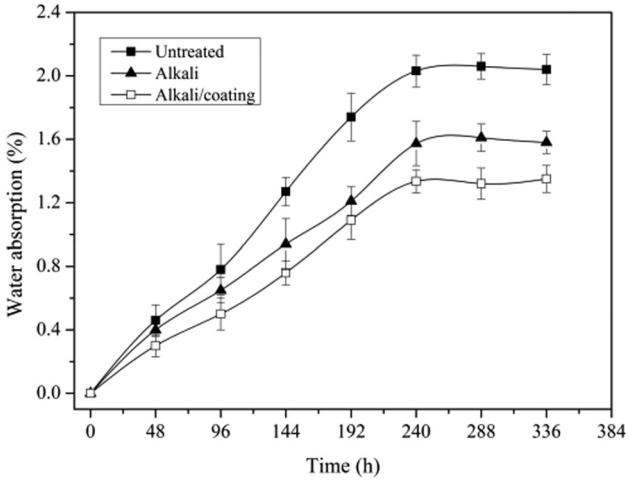

Figure 8: WA of composites (adapted from ref. [70]).

Figure 5(c), there is not much space between alkali + PVA-treated sisal fibers and HDPE. According to the observations, the sisal was only fractured at the cross section and not pulled out. Both tensile and flexural strengths increased initially and then declined when the sisal fiber amount was raised (Figures 6 and 7). The flexural and tensile strengths of the alkali/PVA-coated sisal fiber composites attained their highest amounts of 45.84 and 26.86 MPa, respectively, when the sisal fiber content reached 12%. The alkali/PVA coating treatment increased the tensile and flexural strength of composites by up to 18.85 and 18.90%, respectively, in comparison to untreated sisal fiber composite. However, once sisal fiber levels rose beyond 12%, fiber aggregation took place and led to weak distribution. The WA of sisal fiber/HDPE composites with a 12% filling content is shown in Figure 8. By sisal fiber's surface treatment, the composite materials' capacity to absorb water was drastically reduced. Untreated sisal composite absorbed 2.03% of water, but alkali-treated sisal composite absorbed 1.57% of water, 22.67% less than untreated sisal composite. The alkali/PVA coating treated sisal fiber composite absorbed 1.33% of water, 34.48% less than untreated sisal fiber composite [69].

2.3 Steam-exploded (SE) treatment

The impact of adding SE wood flour to a wood flour/plastic composite was studied by utilizing *Cryptomeria japonica* (Japanese cedar), *Shorea negrosensis* (red meranti), and *Fagus crenata* (beech) and three types of thermoplastic polymers including PVC, polymethylmethacrylate, and polystyrene [76]. The substitution of SE for wood flour often enhanced the wood plastic board's modulus of elasticity and rupture, and resistance to water. However, depending on the plastic polymer and partially the wood species of SE, the best-performed composition of the board was varied. Despite plastic polymer, water durability was superior for all composites utilizing SE compared to untreated composites, and the SE wood species had minimal effects on the final board's qualities [76].

2.4 Fungicide solutions

Ashori *et al.* [77] studied the impact of two chemical preservatives including "3-iodo-2-propynyl butylcarbamate (IPBC)" and "2 thiazol-4-yl-1*H*-benzoimidazole (TBZ)" on endurance against the *Coriolus versicolor* (white-rot fungus) as well as physical and mechanical characteristics of HDPE-reinforced composites by poplar wood flour (PF). The study revealed that treated composites had much lower strength losses and higher resistance against decay than untreated samples. Fungicide treatments enhanced TS and WA, but there was no obvious difference between the modified samples (Table 6). The weight loss of different treated composites varied from 1.1 to 4.5%, and the control had the greatest weight loss. Additionally, compared to the treated composites with TBZ, samples that had undergone IPBC treatment had slightly less weight loss. As a result, reinforced composites can be properly protected with IPBC and TBZ. Therefore, IPBC demonstrated better results than TBZ, and it is advised to protect WPCs [77].

The thermal decomposition, dimensional stability, and biological and mechanical properties of HDPE-reinforced composites by CCA-treated wood, which were collected after serving for 20 years, were examined by Tascioglu *et al.* [78]. With the exception of the Izod impact strength,

Table 6: Average mechanical and physical properties before and after fungal incubation (adapted from ref. 77)

Treatment	Property									
	MOR (MPa)		MOE (MPa)		NI (kJ·m^{-2})		WA (%)		TS (%)	
	Before	After	Before	After	Before	After	Before	After	Before	After
Control	47.4 (1.2)	31.7 (3.4)	4,424 (76)	2,466 (221)	0.43 (0.08)	0.34 (0.08)	11.4 (1)	14.9 (3.3)	4.1 (1.2)	5.0 (3.2)
A_1	49.1 (0.9)	34.0 (2.2)	4,046 (115)	2,730 (154)	0.37 (0.09)	0.31 (0.08)	9.4 (1.1)	10.8 (2.1)	4.0 (0.9)	4.5 (1.4)
A_2	44.3 (0.8)	35.9 (2.1)	4,360 (97)	2,877 (165)	0.37 (0.09)	0.35 (0.09)	8.6 (0.5)	10.0 (3.1)	3.9 (1.4)	4.4 (2.7)
A_3	43.9 (1.4)	39.6 (2.5)	4,250 (136)	3,192 (111)	0.38 (0.09)	0.37 (0.1)	10.4 (2.2)	11.3 (2.2)	4.3 (1.3)	4.7 (2.1)
B_1	37.4 (1.9)	26.8 (3.0)	3,951 (117)	2,339 (89)	0.41 (0.07)	0.31 (0.09)	12.7 (0.9)	14.7 (3.4)	3.8 (0.5)	4.1 (1)
B_2	34.4 (2.4)	27.8 (2.8)	3,686 (112)	2,395 (99)	0.37 (0.09)	0.34 (0.08)	12.1 (1.3)	14.1 (1.9)	3.7 (3)	4.2 (2.1)
B_3	32.8 (1.9)	27.2 (1.2)	3,723 (80)	2,627 (146)	0.37 (0.08)	0.35 (0.03)	13.4 (1.4)	14.5 (2.9)	4.5 (1)	4.9 (1.2)
A_1B_1	41.8 (2)	37.8 (1.0)	4,179 (116)	3,080 (79)	0.37 (0)	0.36 (0.08)	11.8 (1.3)	13.8 (1.9)	5.2 (4.1)	5.8 (2.3)
A_3B_3	43.0 (1.7)	40.8 (1.0)	4,282 (89)	3,437 (100)	0.39 (0.02)	0.37 (0.03)	9.3 (1.5)	10.2 (2.3)	4.6 (3.2)	5.0 (2.3)

Note: Values listed in parentheses are the standard deviations based on four specimens.

the WPC's dimensional stability and mechanical properties were enhanced by the recycled CCA-treated wood flour and coupling agent. The biological analysis confirmed that the treatment enhanced resistance to fungus and termites. Instead of treating wood with chemicals, this procedure could be utilized as a substitute recycling method.

Because the majority of WA occurs in the wood, increasing the wood flour percentage made the material more susceptible to decay and mass reductions. Several borate-based biocides, including zinc borate (ZB), disodium octaborate tetrahydrate, calcium/sodium borate, and boric acid, can stop the fungi and other biodeteriorates [79]. ZB, which has a high level of leaching resistance, broad efficacy against insects and fungi as well as very low mammalian toxicity and cost, is widely utilized commercially in a variety of wood composites, such as wood plastic and particleboard [80].

Hosseinihashemi and Badritala [79] studied the long-duration WA of wood flour prepared by ZB in a PP matrix as influenced by "*Trametes versicolor*" after 98 days of

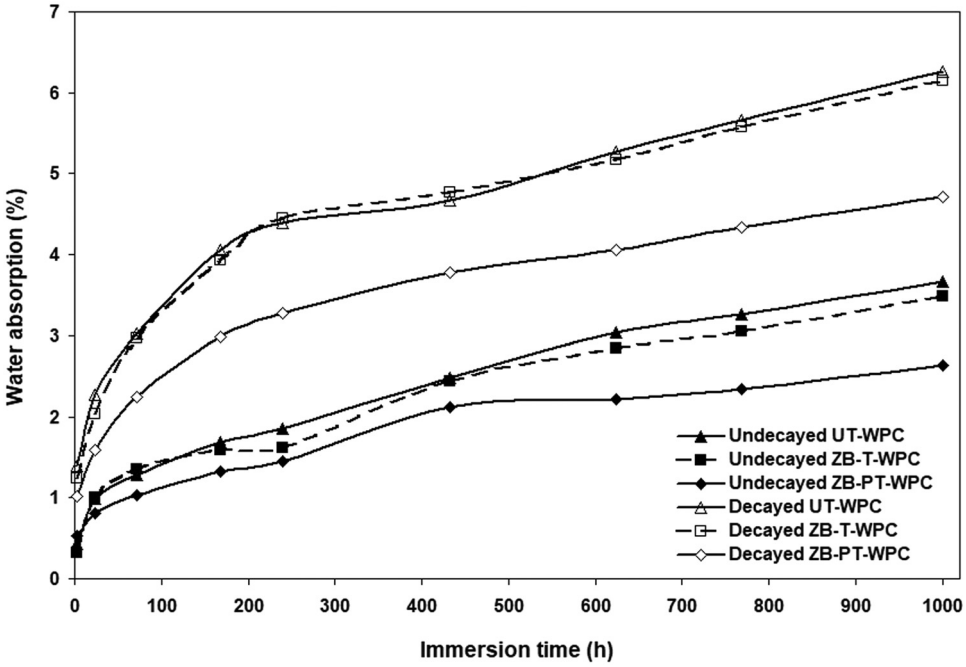

Figure 9: Typical WA curves for selected composite formulations. UT-WPC: untreated wood flour/plastic composite; ZB-T-WPC: zinc borate treated wood flour in manufacturing process/plastic composite; ZB-PT-WPC: zinc borate pretreated wood flour/plastic composite (adapted from ref. [79]).

incubation. Since the WA and diffusion coefficients differed depending on the sample formulation, it was clear that the ZB pretreatment employed in the current investigation had a beneficial impact on the WA of WPCs. Figure 9 illustrates how the ZB-containing specimens had less WA than the untreated ones. In comparison to the untreated composites, the WA of the ZB-T-WPC and ZB-PT-WPC samples reduced by 4.6 and 27.9%, respectively. This is mostly due to ZB's lower water solubility. Additionally, the average WA was higher in the ZB-T-WPC samples compared to the ZB-PT-WPC samples. It might be anticipated that ZB precipitation in cell lumens and cell walls will eventually decrease in WA [79].

Badritala *et al.* [80] investigated how ZB treatment affected the mechanical and morphological characteristics of wood flour in the PP matrix. Except for tensile strength, the ZB treatment had no effect on the composite's mechanical features. In comparison to the untreated samples, ZB-containing samples had reduced flexural, tensile, and impact strengths. However, the hardness was only slightly enhanced by the ZB modification. According to SEM results, certain crystalline deposits of ZB were deposited on the outer surface of the wood fibers, increasing the surface area of the solids inside the WPC and decreasing the bond effectiveness of the polymer [80].

Gnatowski [81] investigated how zinc borate (ZnB) affected WPCs' ability to absorb water. The findings demonstrated that two industrial WPCs with ZnB absorbed lower water in exposed situations. The moisture diffusion coefficient of WPCs made of HDPE and ZnB was studied by Jahadi *et al.* [82]. The findings indicate that the diffusion constant of the composites was reduced by the addition of ZnB (1%).

2.5 Chemical coupling agents

One of the major chemical techniques for enhancing interfacial adhesion is chemical coupling. In this technique, a substance is applied to the fiber surface to create a chemical link between the matrix and the fiber. The majority of researchers discovered that these medications were successful and improved interfacial adhesion [37]. In order to treat wood fibers, more than 40 different compatibilizers have been studied. The most important types include anhydrides, silanes, isocyanates, and anhydride-modified copolymers [83]. In general, the literature claims that using maleic anhydride (MA) grafted matrices as a coupling agent increases tensile strength and elongation at break [39]. The load of the coupling agent is a key factor in the treatment of fibers. Insufficient bonding between the fibers and matrix results from low values of coupling agents. In contrast, taking too much might cause substantial fiber's agglomeration [84,85]. As a result, the best performance of the composite materials would need the appropriate amount of compatibilizers [86].

Cui *et al.* [87] treated the wood fibers using compatibilizers, including MA, methyl methacrylate (MMA), and KH550, in order to decrease the water uptake of composites. The outcomes showed that the three surface modifications could successfully reduce the TS and WA of WRPCs. The WA ratios of WRPCs with treated wood fibers by KH550, MMA, and MA were decreased by 10.0, 21.7, and 27.6%, respectively, compared to the untreated samples, and the impact toughness was improved by 18.4, 24.0, and 2.1%, respectively, after 4 weeks of soaking in hot water. When WRPC was treated with KH550 and MA, the tensile strength improved by 26.0 and 11.1%, respectively, but it reduced by 6.5% when treated with MMA. The effective approach for decreasing the WA of composites was treated wood fibers by coupling agents, and this type of composite had the best overall features [87].

In the study conducted by Khamtree *et al.* [88], alkaline, silane, and alkaline–silane treatments were employed to rubber wood flour (RWF) to create reinforced recycled polypropylene (rPP) composites. To assess the role of treatments in WA, morphological characterization, mechanical and thermal properties of WPCs, silane treatment was employed at different proportions and durations. In comparison to silane or alkaline alone, the results showed that RWF treated with an alkaline–silane mixture demonstrates superior properties. Additionally, treatment times mostly unchanged the mechanical strength, hardness, and WA, although silane amounts had a considerable impact on these properties. The highest water resistance, mechanical strength, and hardness of WPCs were achieved by an alkaline-silane treatment (5% silane for 2 h). This procedure also raised the crystallinity of the WPCs and enhanced the interfacial bonding of RWF and rPP [88].

According to Hou *et al.* [89], incorporating the two treatments for composites will improve bonding and might even have an unanticipated synergetic effect.

Ramlee *et al.* [90] studied the different characteristics of sugarcane bagasse (SCB) and oil palm empty fruit bunch (OPEFB) composites treated with 4% H_2O_2 and 2% silane. In accordance with the findings, a 24-h examination revealed that the silane-treated sample had the best mechanical characteristics and the least amount of thickness swelling and water uptake. Flexural strength (Figure 10) and flexural modulus (Figure 11) of reinforced composites were significantly improved with the addition of 2% silane-treated fibers compared to H_2O_2 and untreated composites.

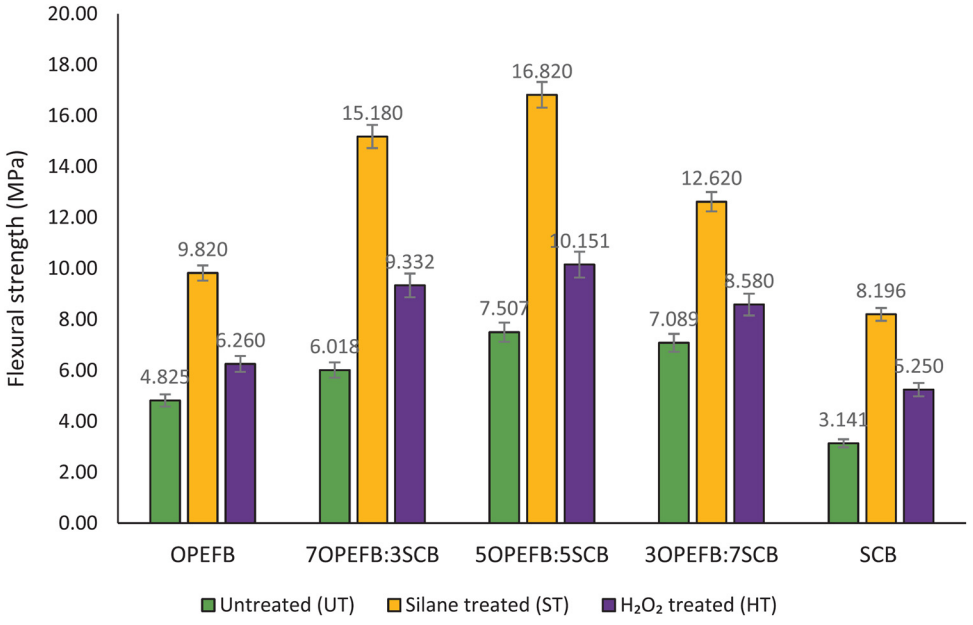

Figure 10: Flexural strength of treated reinforced composites (adapted from ref. [90]).

The research revealed that silane treatment enhanced the functionality of waste natural fibers, and green composites containing a mixture of different fibers meet both the physical and mechanical requirements for insulating board standards to create new classes of sustainable and eco-friendly construction materials such as thermal insulation [90].

Vinod *et al.* [91] assessed the role of NaOH, oxalic acid, and silane as treatments of soy stem fibers in the physical and mechanical features of reinforced composites. The study's findings demonstrated that silane treatment of composites improved their mechanical performance when compared to NaOH, oxalic acid, and untreated fibers. The soy fibers' thermal stability was enhanced after the silane treatment, and the silane-treated composite had a lower coefficient of thermal expansion [91]. The significant improvement in mechanical and physical properties of silane-treated composites was also confirmed by Ramesh *et al.* [92], where the tensile strength of silane-treated fiber composites varies in the range of 35–40 MPa, which is two times higher than that of untreated fiber composites.

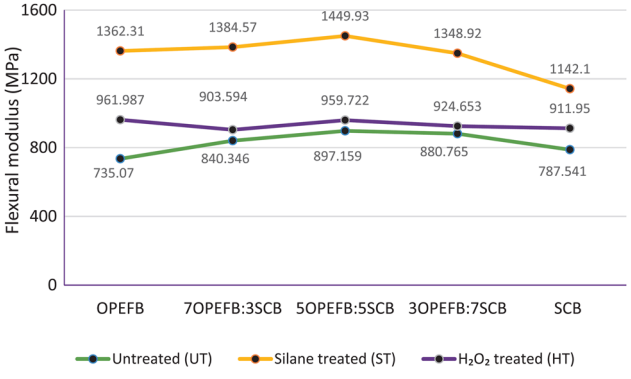

Figure 11: Flexural modulus of treated reinforced composites (adapted from ref. [90]).

2.6 Plasma treatment

By inserting functional groups like sulfonates and amine into wood fibers, plasma treatment is an environmentally friendly mechanical treatment that is applied to enhance the wood fibers' surface features. It could be applied to remove contaminants from the fiber surface, increase fiber porosity, and the wood fiber defibrillation, which would then improve the mechanical interlocking of polymer on the surface of fibers. Along with the advantages that plasma treatment has over chemical-based treatments in terms of the environment, it also takes much less time (1–3 min) than traditional chemical processes, which need several days to treat fibers appropriately [36]. For the fibers of Spanish broom and beech veneer, plasma modification transforms the soft surface of the fibers into a cross-linked one (Figure 12) [93,94]. High plasma discharge power results in greater surface etching, which increases the degree of inhomogeneity of the fibers by creating deeper and wider cracks. On the fiber surface, which was peeled off, threads were produced [36].

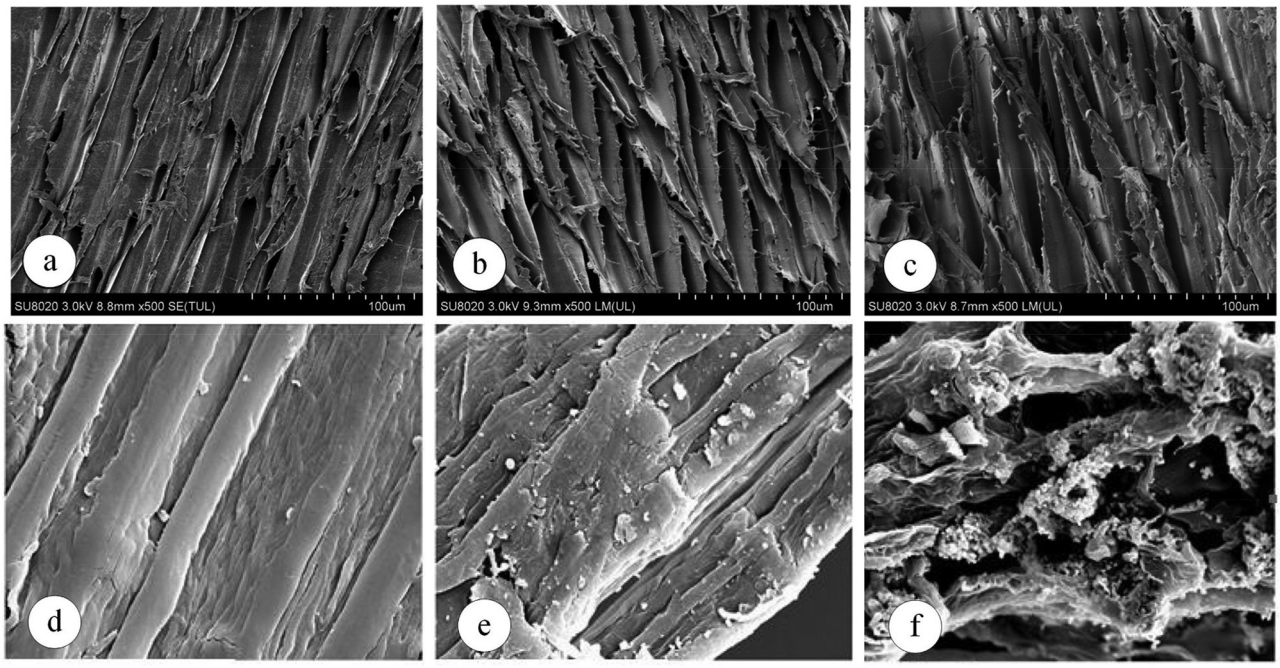

Figure 12: SEM pictures of: beech veneer's surface (a) untreated, (b) 1 kW plasma treated fibers, (c) 2 kW plasma treated fibers; Spanish Broom fibers (d) untreated (e) 10 W plasma treated fibers (f) 70 W plasma treated fibers (adapted from ref. [36]).

It has been shown to be beneficial to treat WPC by low-pressure plasma to increase adhesion. Although treating WPCs with low-pressure plasmas is efficient, it is challenging to implement on an industrial scale since a vacuum is required, the treatment is not continuous, and it can only be applied to small pieces [95].

Hämäläinen and Kärki [96] used contact angle measurement with sessile drop technique and tensile strength testing of glued samples to examine the impact of atmospheric plasma treatment on PP and spruce (*Picea abies*) wood–plastic composite surfaces. WPC profiles that have been extruded are subjected to a plasma treatment. The findings demonstrate an enhancement in the tensile strength of glued specimens after plasma treatment as well as an improvement in the contact angle of WPC materials treated with plasma. According to the research findings, it is evident that atmospheric plasma processing enhances the bonding of WPCs, as proved by the results of sessile drop and tensile strength tests of glued specimens. An increase in polar groups in the Raman spectra could serve as a sign of the plasma treatment's effectiveness [96].

2.7 Distillate treatment

Väisänen *et al.* [97] investigated how hardwood distillate (HWD) affected the characteristics of a commercial WPC

Table 7: Densities and mechanical properties of the studied materials (adapted from ref. [98])

Material	Density ($g \cdot cm^{-3}$)	Tensile strength (MPa)	Tensile modulus (GPa)	Strain (mm)	Flexural strength (MPa)	Modulus of elasticity (GPa)	Bending (mm)	Charpy's impact strength ($kJ \cdot m^{-2}$)
LG	1.130 ± 0.002	22.41 ± 0.82	2.09 ± 0.20	1.86 ± 0.26	44.09 ± 2.00	3.05 ± 0.16	4.54 ± 0.28	11.57 ± 2.02
LG + HWD1	1.131 ± 0.002	22.85 ± 0.31	2.33 ± 0.09[a]	1.81 ± 0.12	45.27 ± 1.01	3.21 ± 0.08[b]	4.42 ± 0.28	10.61 ± 1.27
LG + HWD2	1.132 ± 0.002	22.26 ± 0.30	2.24 ± 0.06	1.83 ± 0.15	43.09 ± 0.83	3.11 ± 0.12	4.37 ± 0.30	11.17 ± 0.81
LG + HWD4	1.138 ± 0.004	22.05 ± 0.44	2.16 ± 0.06	1.92 ± 0.17	42.86 ± 1.17	2.89 ± 0.09[b]	4.72 ± 0.29	10.89 ± 1.13
LG + HWD8	1.141 ± 0.003	19.67 ± 0.52[a]	1.86 ± 0.11[a]	1.85 ± 0.13	39.36 ± 0.63[a]	2.73 ± 0.09[a]	4.67 ± 0.27	9.42 ± 0.69[b]

WPC: wood–plastic composites; LG: LunaGrain; HWD: hardwood distillate.
[a]$p < 0.01$ (highly significant difference compared with the unmodified WPC).

made of thermally treated sawdust (Scots pine) in a polymeric matrix by applying several levels (1–8 wt%) of distillate to the mixture. The results of the mechanical experiments revealed that adding 1 wt% of HWD to WPCs improves their flexural and tensile properties but not their impact strength (Table 7). Incorporating up to 4 wt% HWD greatly reduced the WA of WPCs without decreasing the observed mechanical features, which is another major result from this study. Table 7 shows a substantial tensile modulus improvement occurred by incorporation of HWD (1 wt%). Additionally, improvements in other mechanical characteristics were seen by utilizing distillate modification for produced composites. The reason why composites are more rigid is that HWD occupies the cracks and spaces in the composites, which is also demonstrated by the fact that bending and strain were reduced on greater distillate amounts (above 4 wt%) [97].

Väisänen *et al.* [98] examined the effect of several treatments (alkali, enzymatic, steam, and wood distillate) on the properties of hemp fibers and their composites. Utilizing vacuum-assisted resin transfer molding, long (aligned) hemp fiber (*Cannabis sativa* L.) reinforced epoxy composites were manufactured. The findings demonstrate that the fibers' tensile strength, Young's modulus, and toughness all massively enhanced after steam treatment. Water uptake of the reinforced samples was dramatically reduced as a result of the treated hemp fibers (Figure 13). The fiber modification by the alkali method depicted the minimum WA levels among other processes.

According to Baghaei *et al.* [99], an improvement in the interfacial adhesion between the polymer matrix and fibers may be utilized to explain alkali treatment led to a reduction in the WA. Another factor is that the elimination of hemicelluloses and other hydrophilic materials reduced fibers' hydrophilicity. Another effective strategy to reduce the composite's WA is enzyme treatment. Compared to other treatments, the treated fibers by wood distillate indicated low efficiency in water up taking properties. However, untreated fibers still differ significantly from fibers that have been treated using wood distillate [98].

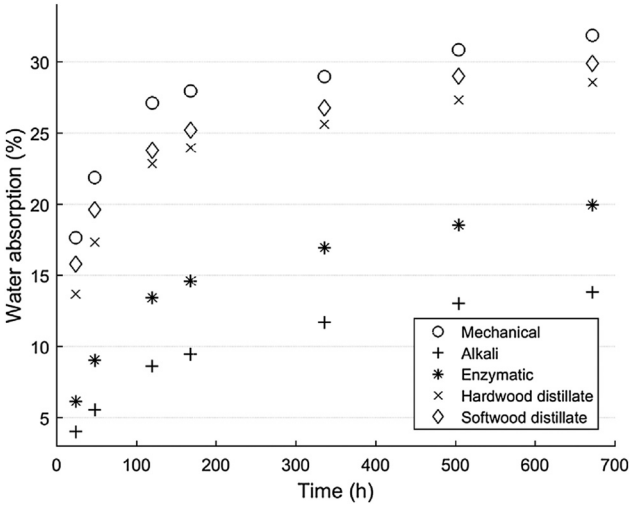

Figure 13: WA of different epoxy-hemp composites during 28 days of immersion (adapted from ref. [99]).

3 Conclusion and future scope

Natural fibers have many benefits to utilize in composite materials including high elastic modulus, flexural strength, flexibility, low density, renewability, biodegradability, and recyclability. These qualities have made natural fiber-reinforced composites more desirable. The hydroxyl groups of hemicellulose, cellulose, and lignin, which compose natural fiber, form many hydrogen bonds in the wood's structure. The hydroxyl groups may create new hydrogen bonds with water molecules, causing water uptake, fiber expansion, and the development of small cracks in the specimen, leading to the fiber debonding and the deterioration of the interface between polymer and fiber. Due to the hydrophobicity of the polymer and the hydrophilicity of the fiber, polymer matrix containing natural fibers typically exhibit weak interfacial adhesion and hence have a limited capability to transfer stress from the matrix to the reinforcement materials. The fiber surface may experience a variety of treatments, which are mainly categorized as chemical, physical, and thermal treatments, to enhance bonding properties, decrease WA, enhance weak wettability, and improve both structural and mechanical properties. The utilization of a wide range of fiber treatments on different properties of reinforced composites with natural fibers was discussed in this research. Generally, physical and thermal treatments compared to chemical methods are more environmentally friendly, but they require higher energy consumption, are relatively newer, and also have the appropriate potential to develop in the future. According to the results of the studies, the physical and mechanical properties of the fiber-treated composites were typically improved by using fiber treatments, and combined treatment methods may exhibit an unanticipated synergetic activity.

Future research should concentrate on creating fully bio-composites utilizing biodegradable or recycled polymers and a higher percentage of natural fibers, as well as focus on a better understanding of the natural fiber's interaction with polymers through fiber modification to create environmentally friendly innovative products in

all potential industries, including building construction, automotive, and outdoor and indoor applications. Natural fiber surfaces can be treated using traditional treatments (mainly chemical methods) and eco-friendly approaches to improve the interaction of fiber and polymer for a variety of purposes. Natural fiber modification using chemical methods, including benzoylation, peroxide, mercerization, acetylation, compatibilizers, and polymer grafting, requires the use of a large number of dangerous compounds. The manufacturing costs of natural fiber-reinforced composites may rise due to appropriate management of chemical waste. In order to treat the natural fibers, eco-friendly approaches such as bacteria, fungi, enzymes, plasma, cellulose, nanocellulose coating, and supercritical carbon dioxide should be employed. These methods are a great replacement for traditional techniques. Researchers should focus on two major areas of interest in future research projects based on the reviewed articles in the current study: firstly, moving toward environmentally friendly methods for fiber treatment, and secondly, the incorporation of two or more treatment methods simultaneously with the evaluation of different levels, ratios, durations, and other experimental situations to develop eco-friendly fiber modification techniques for the manufacturing of reinforced composites.

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Data availability statement: Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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