Advances in Materials Science and Engineering

Advances in Wood Composites

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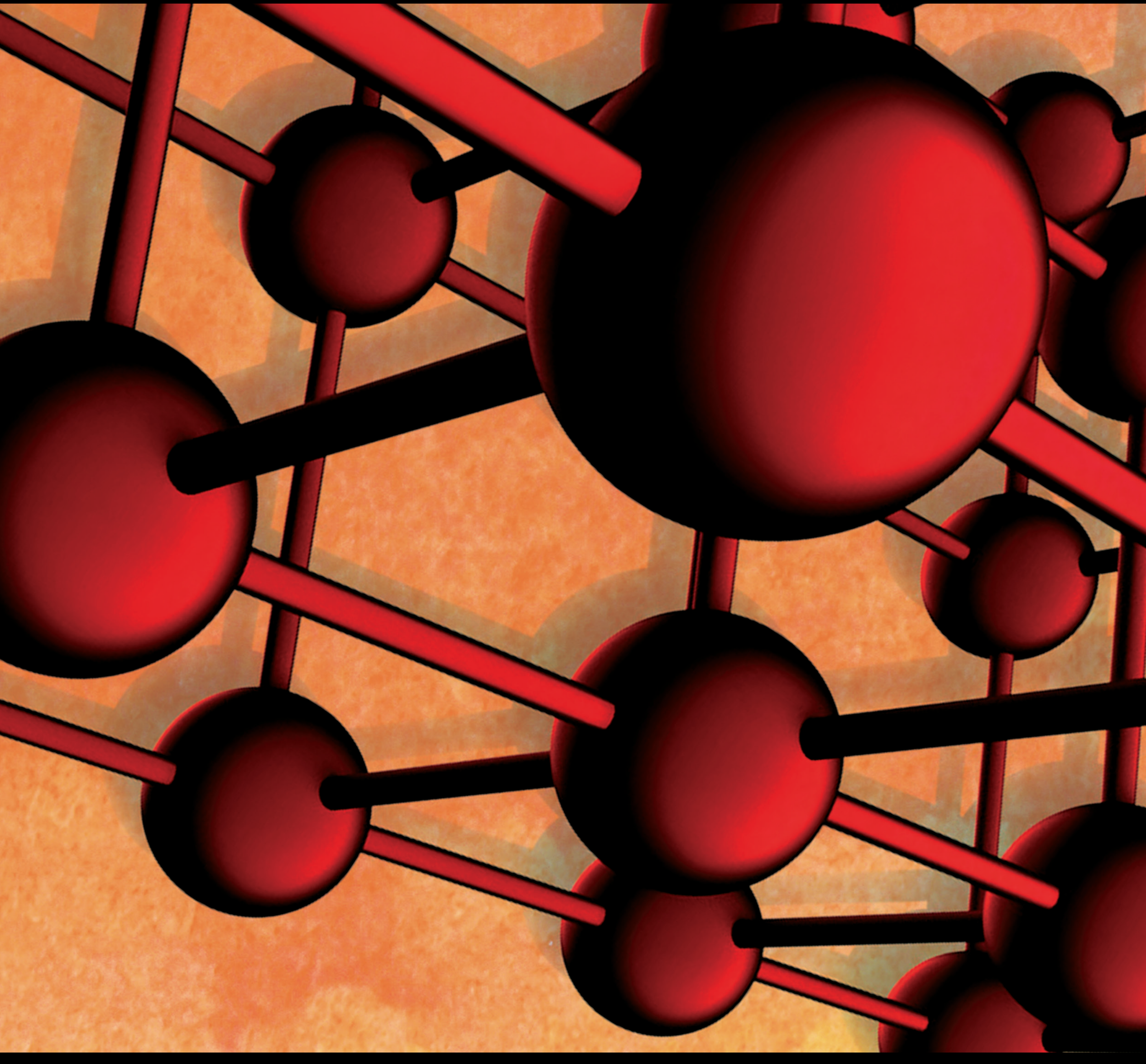
Guest Editors: Ľuboš Krišťák, Richard Kminiak, and Tomasz Rogozinski



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Review Article

Lignin as Green Filler in Polymer Composites: Development Methods, Characteristics, and Potential Applications

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After cellulose, lignin is the most commonly used natural polymer in green biomaterials. Pulp and paper mills and emerging cellulosic biorefineries are the main sources of technical lignin. However, only 2–5% of lignin has been converted into biomaterials. Making lignin-based polymer biocomposites to replace petroleum-based composites has piqued the interest of many researchers worldwide due to the positive environmental impact of traditional composites over time. In composite development, lignin is being used as a filler in commercial polymers to improve biodegradability and possibly lower production costs. As a natural polymer, lignin may have different properties depending on the isolation method and source, affecting polymer-based composites. The application has been affected by the characteristics of lignin and the uniform distribution of lignin in polymers. The review's goal was to provide an overview of technical lignin extraction, properties, and its potential appropriate utilization. It was also planned to revisit the lignin-based composites' preparation procedure as well as their composite characteristics. Solvent casting and extrusion methods are used to fabricate lignin from polymeric matrices such as polypropylene, epoxy, polyvinyl alcohol, polylactic acid, starch, wood fiber, natural rubber, and chitosan. Packaging, biomedical materials, automotive, advanced biocomposites, flame retardant, and other applications for lignin-based composites has existed. As a result, the technology is still being refined to increase the performance of lignin-based biocomposites in several applications. This review could assist explain lignin's position as a composite additive, which could lead to more efficient processing and application strategies.

1. Introduction

Approximately 50–70 billion tons of lignin are produced per year [1, 2] by the pulp and paper industry and emerging cellulosic biorefineries, with 98% being burned for energy or simply dumped in landfills, posing a major disposal issue [3]. In biomass cell walls, lignin is the most available non-carbohydrate polymer. It has several impressive properties, including good mechanical and physicochemical properties, low weight with antioxidant and antimicrobial properties, diverse possible modifications, and excellent thermal stability. Its biodegradability, environmental friendliness, wide ecological adaptability, and reinforcing ability create it as an excellent alternative for developing novel green materials, including polymer composite materials [3–5]. Many potential applications for lignin can be developed and implemented in industries such as adhesives, biosurfactants, antimicrobial agents in packaging and textiles, antioxidants, adhesives, anticorrosion, carbon fiber or carbon black, cosmetics, reinforcing agents, hydrogel, phenolic resin, flame retardant, polyurethane, foam composite, compatibilizers, and new biomedical materials [6–11]. However, lignin-based composites' product commercialization and manufacturing processes need to investigate because some had not yet been commercialized in the market.

Making a lignin-based polymer composite to replace petroleum-based composites has become of great interest to many researchers worldwide, considering its negative environmental impact in the long term on typical composites. Introducing lignin into commercial polymers can create green composite or eco-friendly composite as lignin contributes to improving the product's biodegradability. For example, the presence of lignin in polypropylene-reinforced kenaf core hybrid polymer nanocomposites increases the thermal stability and fire retardant properties of bionanocomposite. However, there is still a great challenge before obtaining lignin-derived biocomposites comparable to those found in the petroleum industry. The composite properties were influenced by the chemical structure of lignin [12]. Lignin can function as a filler, stabilizer, compatibilizer, and reinforcement in composites with its distinctive chemical structure. Its aromatic and cross-linked functional groups make it suitable for a variety of polymeric matrices with improved wettability, mechanical, and fire-retardant properties [13]. Due to the intricacy of its structure and reactivity, lignin is frequently difficult to combine properly. When used as an extender, lignin can lower the cost of polymer composite production while also improving biodegradability. Lignin contains inter- and intramolecular hydrogen bonds, making it thermoplastic and thermally stable at high temperatures [14]. Adding lignin to polylactic acid (PLA) or polypropylene (PP) has been a hot topic over the last decade [15]. The different characteristics between lignin-containing hydroxyl groups and polymers such as aliphatic polyester become a challenge to make a uniform distribution for lignin and polymer such as polylactic acid (PLA) mixture [15].

Furthermore, lignin matrix compatibility and homogeneity are big challenges in most research [16]. Besides that, high mineral content in lignin and other impurities can interfere with the uniform mixing of the composite. Lignin has different polarity and immiscibility from nonpolar polymers. It is also not easy to dissolve in common solvents. Therefore, some lignin modification is required to modify the lignin properties, such as acetylation, esterification, methylation, and phenolation [17, 18]. Chemical modifications of lignin such as phenolation and acetylation have become prominent in recent years. Modified lignin can be used as a compatibilizer in bi- or multiphase composites. Esterification is one method of modifying lignin when used as a compatibilizer [16]. Previously, some reviews have been reported in lignin-based biocomposites; however, to the best, our acknowledgment is not in deep discussion especially on the method to prepare lignin as filler incorporated with some matrices. Besides that, this review brings detailed information on the characteristic of lignin-based biocomposites. The goal of this review was to present an overview of the application of lignin as filler in composites including the development methods and composite properties such as thermal, morphological, mechanical, thermal, and other properties.

2. Lignin Structure, Extraction Method, and Its Properties

Lignin contains an aromatic structure with an amorphous macromolecule of repeating phenylpropane units, making it a potential precursor to produce materials, fuels, and chemicals. Lignin is the second major component of wood cell walls (20–30%) that serves to cover structural supports and transport water and nutrients, as cement between wood fibers, as a hardening agent in fibers, and as a barrier against chemical, biological, and enzymatic degradation of cell walls [19]. It is a three-dimensional, irregular, and highly branched phenolic polymer with a highly complex chemical structure and formed through an irregular biosynthetic process. Figure 1(a) presents a lignin structure consisting of basic phenylpropanoid monomers, guaiacyl (G), syringyl (S), and p-hydroxyphenyl (H) units, which are derived from coniferyl, synapyl alcohol, and p-coumaryl precursors, respectively [2, 13]. These macromolecules contain large amounts of C-C and ether-related compounds with the aryl glycerol-β-aryl ether (β-O-4) link predominating so far, followed by the biphenyl (5–5) and phenyl coumarin (β-5) bonds [21, 22]. Species and environment influence the content and composition of lignin. Hardwood lignin consists mainly of G and S units and has traces of H units

[23, 24], while grasses contain G, S, and H units. Hardwoods have more lignin composition variability than softwoods. In contrast, softwood lignin consists mainly of G units, with low levels of H units [25].

The proportions of S, G, and H units were 1:94:5 in spruce lignin, 56:40:4 in beech lignin [26], and 1:1:1 in grass lignin. Even though there is variation in the content contingent on the sources, the typical lignin contents measured by the Klason method are 25–35% in softwoods, 20–25% in hardwoods, and 15–25% in herbaceous plants [27]. Precipitation of lignin begins in the primary wall's middle lamella and the secondary wall's S1 layer. Lignin is also found in S2 and S3 layers [28, 29]. Its composition and structure vary depending on the amount and proportion of functional groups linked to aromatic or aliphatic moieties [2, 30, 31] that affect the reactivity of lignin. The structural schematic of lignin in plants is shown in Figure 1(a), while the linkages of lignin are presented in Figure 1(b).

2.1. The Extraction Method and Its Properties. Many conversion pathways for lignin utilization have been proposed and studied. In some cases, the structure and properties of the extracted lignin (technical lignin) change due to the method of processing lignocellulosic biomass into pulp. In commercial applications such as fillers in composites, technical lignin can be extracted from biomass physically with or without chemical or biochemical treatments. However, controlling the physical properties, reactivity, and chemistry of the lignin surface should be performed [32–34]. Processes with sulfur such as kraft and sulfite pulping are discussed below, as are sulfur-free processes such as alkaline pulping (soda lignin) and solvent pulping (organosolv lignin). Sulfur-free lignin is lignin that has a low macromolecular size upon fractionation. In addition to low molar mass phenols or aromatic compounds, they have other desirable properties. Table 1 presents some key features of lignins related to their utilization with a detailed description of each lignin in the discussion.

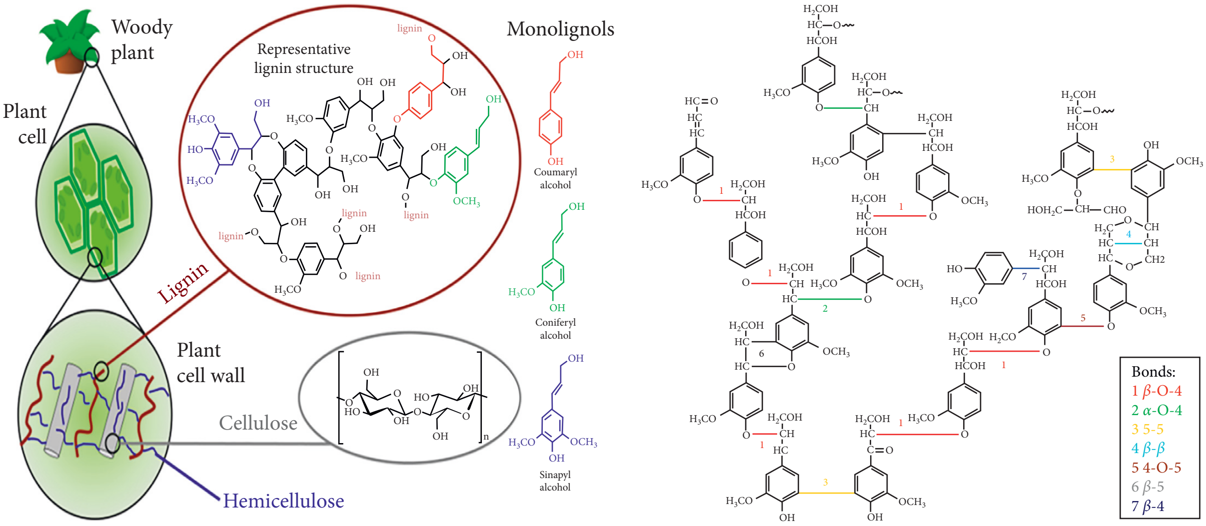
2.2. Sulfur Bearing Process

2.2.1. Kraft Processes. Kraft lignin (KL) is produced by kraft pulping with sodium sulfide and sodium hydroxide chemicals. The KL is the world's largest source of technical lignin at around 85% [53]. The β-O-4 linkages between the phenyl propane (C9) units in lignin are cleaved by sulfide at high temperatures up to 170°C [54], lowering the molecular weight of the polymer [34]. KL has a high sulfur content resulting in the formation of aliphatic thiol groups as a result of this mechanism and is hydrophobic [25]. During the pulping process, lignin reacts with chemicals to form fragments that are soluble in water or alkali, according to Dias et al. [55]. The phenylpropane group is also broken, resulting in a phenol-free hydroxyl group, making the lignin hydrophilic.

The process of lignin precipitation completes the lignin separation. LignoBoost is a precipitation and separation process to obtain a pure KL. Acidification with carbon dioxide precipitates and filters out the lignin. However, compared to previously published methods, an improved washing procedure is used by the recent publication of Solihat et al. [7]; they isolated *Acacia mangium* KL by single step and ethanol fractionated step that successfully recovered KL of ~35 and ~16%, respectively. The single step has higher acid-insoluble lignin (AIL) than the ethanol fractionated step. KL produces a complex mixture of breakdown products ranging in molecular mass from low-molecular-mass phenolic compounds to large macromolecules [37]. Acidification can precipitate most of the KL in black liquor. Lignin with a low molecular mass, on the other hand, is still present in the solution. The output of precipitation is primarily determined by the liquid's final pH. Because no corrosive salts are formed when carbon dioxide is used in technical applications, it is advantageous. However, it is impossible to achieve a pH of less than 8.5. If the liquid is acidified with a strong mineral acid, more lignin can be precipitated. The phenolic and hydroxyl groups of lignin can be separated from the weak acid by carbon dioxide but not the stronger carboxylic acid group [39].

KL can be used in the same applications as lignosulfonates (LS), despite having different inherent properties such as limited solubility. In rubber resins and plastics, KL or modified forms of KL can be used as dispersing agents and additives. These polymers can be used to make adhesives for a variety of products, including laminated boards and plywood. Lignin can also be broken down into chemicals with a low molecular mass [39]. The average molecular weight (Mw) of softwood KL is 3,900. KL has a low molar mass (Mn), ranging between 1,000 and 3,000 g·mol^{-1} [56]. The molecular weight of KL, on the other hand, can vary depending on the type of wood, analytical method, and isolation technique. Using methanol fractionation, Saito et al. investigated the properties of high molecular weight (HMW) and low molecular weight (LMW) kraft softwood lignin (KSL). HMW of KSL has been fruitfully separated from LMW, which is soluble in methanol and has Mw in the range of 3,000 to 86,600 g·mol^{-1}. HMW of KSL is insoluble in methanol and has Mw in the range of 14,900 to 188,000 g·mol^{-1} [38].

2.2.2. Sulfite Process. Sulfite lignin is traditionally the most widely used type of pulping process and the most widely available commercial type of lignin. A base of calcium, sodium, magnesium, or ammonium and aqueous sulfur dioxide (SO_2) is used in the pulping process. After sulfite pulping at 140°C with chemicals such as SO_2, Na+/Ca+/Mg+, and NH4+, LS results were obtained [54]. It is made through sulfite pulping, which involves incorporating a sulfonate group into partially hydrolyzed lignin and then converting it into sulfonated groups with various functional groups that give them unique colloidal properties [36] and a high molecular weight than KL [40]. It dissolves in a variety of aqueous solutions with varying pH levels but not in organic solvents such as ethanol or acetone. Because of its low molecular weight, LS is extremely compatible with other polymeric materials. During alkaline cooking at high



(a) (b)

Figure 1: (a) The lignin structure and three main lignin precursors [20] (Copyright @ 2017 Science and Education Publishing CC under 4.0) and (b) typical interunit linkages present in softwood lignin [6] (Copyright @ 2018 Springer CC under 4.0).

Table 1: Different characteristics of lignins and the appropriate utilization.

Lignin	Some key lignin characteristics
Kraft lignin (KL)	(i) Containing ash of 1–2% and suitable for low-value-added applications [25] (ii) Having a high concentration of condensed structures with strong ether bonds and phenolic hydroxyl groups [35, 36] (iii) Containing the organic matter such as polysaccharides and a few extractive fractions [37] (iv) Molecular weight variability from low to the high molecular weight of 3,000–188,000 $g{\cdot}mol^{-1}$ [38] (v) Solubility is limited but can be increased by sulfonation [39] (vi) KL or modified KL can be used as dispersing agents, additives, and thermoset polymers mainly in low molecular weight [39] (vii) KL can produce thermoset polymers when it is condensed with formaldehyde and cross-linked with phenol [39] (viii) KL with higher molecular weight can be used as a thermoplastic mixture or copolymer [38]
Lignosulfonate (LS)	(i) High ash content of 4–8% and a high molecular weight than KL [40] (ii) The high molecular weight of 1,000 to 50,000 Da [40] than KL with a polydispersity index (PI) of 4.2–8 [33, 41] (iii) Many impurities (approximately 30% by weight), such as ash or carbohydrates [41] (iv) Required to modify for high compatibility with other polymeric materials [42] (v) Dissolve in a variety of aqueous solutions but not in organic solvents [40] (vi) Good water solubility [40] (vii) Having a variety of functional groups (carboxylate, phenolic, as well as sulfur-containing groups) provide exceptional colloidal properties, dispersants, additives (plasticizers), surfactants, and flocculants [43]
Soda lignin (SL)	(i) Sulfur-free and has a higher purity than KL and LS [44] (ii) Suitable for a range of high products (resins, chemical reactants, composites, and antioxidant agents) and natural feed supplements for monogastric animals [45–47] (iii) Commonly derived from soda or soda-anthraquinone pulping of agricultural wastes or perennial crops [44–46] (iv) Soda lignin contains lower phenolic and aliphatic hydroxyl groups [38]
Organosolv lignin (OL)	(i) OL has high reactive groups, no sulfur, high purity, and the highest reactivity and is nontoxic [36, 48] (ii) High-quality lignin for higher value-added applications such as chemicals [49] (iii) High solubility in an organic solvent but insoluble in water [50] (iv) The possible application in the adhesive in wood panel production, a monomer in a biopolymer, antioxidant, and pesticide [51] (v) Low content of residual carbohydrates [52]

temperatures, many active lignin groups are destroyed, significantly reducing their activity. Sulfonate groups in aliphatic side chains in LS contain a lot of sulfur. However, the cations used in the production of pulp and recovery usually contaminate them. LS reactivity is influenced by the cations to some extent. The lowest and highest reactivity was found in calcium and ammonium-based LS, respectively, while sodium and magnesium-based LS were moderately reactive [57].

Because of the addition of a sulfonate group, these LS typically have extremely high average molecular weights, even higher than KL. The catalytic transformation of LS is more difficult as a result of incorporating these groups [58]. Ethylene bonds are broken; methoxy groups are destructed; and new C–C bonds are formed during the sulfite pulping process.

2.3. Sulfur-Free Process

2.3.1. Organosolv Process. Organosolv lignin (OL) has an advantage over KL in that it lacks sulfur, which reduces the formation of compounds. It also deactivates the catalyst used in the subsequent process by emitting a foul odor. Some solvents used in solvent pulping at temperatures between 150 and 200°C include acid, formic acid, ethanol, methanol, ethylene glycol, and water [49, 54]. To isolate the biomass components, this method employs a combination of aqueous organic solvents including acetone, ethanol, methanol, or organic acids. Distillation allows the cooking liquor to be reused. Because no toxic sulfur chemicals are used in this process, it is eco-friendly. It is also ideal for direct use due to its high purity.

OL are highly hydrophobic in organic solvents [50]. Precipitation is used to remove them from the solvent, which usually involves adjusting various parameters including pH, temperature, and concentration. The most widely used organosolv processes are ethanol/water pulping (e.g., Alcell) and acetic acid pulping (which contains trace amounts of mineral acids) [59]. A new extraction method based on a mixture of acetic acid, water, and formic acid was also established by the CIMV Company (France). According to several published works, the lignin produced is known as bio-lignin, and it is linear and has a low molecular weight [60]. The organosolv process uses acid-catalyzed solvolysis of β-ether bonds in lignin to dissolve them. Acids (for example, H2SO4, oxalic acid, HCl, formic acid, and acetic acid) can be added to the reaction mixture or made in situ by deacetylating hemicellulose to release acetic acid [61, 62]. The external acid promotes the acidolysis of β-ether, giving a cellulose pulp with lower lignin content. In contrast, the lignin produced by in situ acid formation is thought to retain a higher number of β-ether bonds present in the original lignin [63, 64].

2.3.2. Soda Process. Soda lignin (SL) is obtained during soda-anthraquinone and soda pulping. Its availability is limited due to the inefficiency of the soda pulping process [44]. In an aqueous alkaline medium with a temperature of 150–170°C, NaOH is used to produce pulp [54]. By lowering the pH of the alkaline solution after delignification, the dissolved SL can be recovered. According to the "cleaner production" standard, soda-oxygen pulping technology is a cleaner pulping process [65, 66]. Remarkably, two-stage acid precipitation of SL at pH 5.5 yields a higher purity and larger particle size than that obtained at a pH of 3.98. The granite process, which was developed by paper mills for the synthesis of cellulose from perennial crops or agricultural wastes, is a particular approach for lignin deposition in the black liquor of the soda process. Acidification, usually with a mineral acid, lowers the pH of the liquid. Due to the high silica content, which can precipitate lignin, as a result, recovering cooking liquid from nonwood pulp is difficult, and the obtained lignin is of poor quality [46, 67].

To separate lignin from cellulose and hemicellulose, a variety of extraction methods have been reported. The structure and physicochemical characteristics of lignin can be affected by different isolation methods, isolation types, isolation temperature, and raw material sources [68–70]. Physical methods utilizing membranes can be used to catalyze the isolation processes [71]. Membrane technology allows for the separation of black liquor from various lignin fractions of varying molecular weights, which can then be used to make a range of high products (resins, chemical reactants, composites, and antioxidant agents). Lignin can be degraded by enzymatic isolation of lignin, such as using laccase-producing microbes [47]. Chemical isolation is possible using chemicals such as NaOH and KOH. Approximately 75% of the lignin is precipitated as a sodium salt in this step. After that, the lignin is separated using a filtration method. When lignin is hydrolyzed, it leaves behind solid lignin residues as well as a large amount of unhydrolyzed cellulose. Obtaining lignin for use in polyurethane production requires the extraction with NaOH [72–75]. Acid can also be used to isolate lignin [76, 77] such as HCL and H_2SO_4 [6, 7, 78–80] and HPO_3 [81]. Besides that, biological isolation of lignin and combination processes [82], ionic liquids [83, 84], or processed in hydrothermal [85] or supercritical solvents [86] can also be applied. Supercritical pretreatment provides the best environmental performance and economic considering solvents and carbon dioxide recycling [86].

3. Fabrication of Lignin-Based Green Composites

Lignin can be combined with synthetic and natural polymers [16]. In this review, some kinds of polymers that have been incorporated with lignin were discussed. Lignin can be used as filler material, a nucleating agent, compatibilizer, and coupling agent in composites. When used directly, native lignin becomes less reactive and reduces the mechanical strength of copolymer composites [87]; thus, it can be modified before being mixed with polymers to achieve better performance. The different process is possibly affected by the composite properties and the potential applications. The composite properties are strongly influenced by the interfacial interaction between the matrix and reinforcement to maintain stability between phases and the stress transfer process, among others. To improve compatibility, the miscible constituent is not always used, but it can also be modified by surface modification or by adding a compatibilizer agent. Compatibilizer serves to increase the miscibility to form the desired mixture. Lignin shows low miscibility in polymers, causing agglomeration and phase separation. If adding more lignin, proper homogenization techniques and additives are needed. Lignin tends to agglomerate due to its intermolecular hydrogen bonding, π-π stacking, aromatic groups, and van der Waals interaction between polymer chains [88, 89]. The amount of lignin was added to the polymer so that it can act as a reinforcement is on a 25–40% scale [90]. The homogeneity of lignin in the polymer matrix remains a challenge. Thus, lignin modification is added to achieve the dispersion or compatibility of lignin. The commercial polymers including PP, PVA, and epoxy were discussed, while natural polymers such as PLA, chitosan, starch, rubber, and cellulose have been overviewed. Because of the different characteristics between existing polymers and lignin, some methods for composite preparation have been developed continuously. Table 2 presents the common limitation in the fabrication of lignin with polymer matrices related to possible applications of lignin-based composites.

3.1. Composite Lignin-Polypropylene. Polypropylene (PP) is a thermoplastic that is commonly used as a raw material for various types of products, having the disadvantages of not being able to degrade naturally and having low mechanical properties [91]. One way to address this problem is to mix natural polymers such as lignin to initiate the natural degradation process of PP after its use and to increase its tensile strength, bending strength, or modulus. Furthermore, Dias et al. [92] revealed that the cost to produce KL on an industrial scale was \$70–182/ton, while the PP price was \$1,210/ton [93]. Therefore, adding lignin to PP will be economically beneficial and have a constructive impact on the environment. The process of mixing two or more materials in a composite manufacturing formula needs to consider the suitability and dispersion between the phases of the constituent materials. PP is a synthetic resin formed from the polymerization of propylene, which is a hydrocarbon with a molecular structure of CH_2=$CHCH_3$. Through the process of breaking the double bond in the propylene molecule, a polypropylene chain is formed on the backbone of the carbon atom to which the CH_3 group is attached. The arrangement of CH_3 groups on one side of the carbon chain produces isotactic PP, which is the most widely produced type of PP. This molecular structure causes PP to have rigid properties, melting at a fairly high temperature, which is around 170°C, easy to be flexed, water-resistant, inert, and chemically inert (nonpolar) but easily oxidized [94]. The function of lignin as a reinforcement in composites is strongly influenced by the manufacturing process. The use of native and technical lignin in the manufacture of PP-based composites will be described.

Mixing of PP and lignin (pre-hydrolyzed lignin (PL) or OL) with various weight ratios of the materials was carried out using a roll mill for 1 hour, then formed into granules, at a temperature of 200°C. Then a film sheet was obtained by extruding PP/lignin granules with stabilizer 2,6-di-tert-butyl-4-methyl phenol (0.15 mass %) + calcium stearate (0.15 mass %), as a comparison. In general, composite films containing lignin outperformed PP films without lignin in terms of photodegradation stability. The phenolic group in lignin acts as a stabilizer that can break the polymer chain reaction due to oxidation and radical products. However, the higher the lignin content, the more it encourages radical reactions that can cause oxidative degradation of PP with a hydroperoxide radical mechanism. OL is a more effective initiator of oxidation reactions, compared to PL. Thus, it gives a more significant role in assisting the PP degradation process. The addition of lignin not only can trigger the photo oxide reaction to accelerate the degradation of PP but also stimulates the activity of microorganisms to degrade PP/lignin composites [95]. PP/lignin film composite (4% wt) was produced by processing PP isotactic powder and OL powder in a roll mill for 1 hour, then granulated in a homogenizer at 200°C. Then, film sheets were obtained by extruding PP/lignin granules without the addition of a stabilizer.

Toriz et al. [90] developed an isotactic PP/KL composite with the addition of maleic anhydride-modified PP (MAPP) as a compatibilizer. Isotactic PP with a melting point of 152°C, added with 10–60% KL (5–20 microns in powder form) in a Brabender bowl mixer, with process conditions at 170°C, mixer speed of 120 rpm, for 3–4 min. The resulting compound is ground and then molded by injection molding at a pressure of 600–1,000 psi, a temperature of 170°C. Maldhure et al. [96] developed a PP composite with modified KL from sugarcane bagasse (alkylated lignin and arylated lignin) to improve compatibility and produce a reactive side. The alkylation reaction uses dichloromethane, while the arylation reaction uses chlorobenzene. Mixing PP and modified lignin using Brabender electronic plasticorder (HAAKE AEV 153 mixer, Germany) at 60 rpm, at a temperature of 190°C, for 10 minutes. During mixing, dry nitrogen was added to prevent polymer degradation. The antioxidant properties of modified lignin can increase the stability of the composite to heat. The addition of lignin causes an increase in melt flow index (MFI), indicating that lignin can act as a stabilizer in the PP extrusion process. Dias et al. [92] compared the composite properties of PP and KL from sugarcane bagasse and softwood with 3% MAPP as compatibilizer. Before compounding with a twin-screw extruder, lignin and PP were oven-dried at 80°C for 3 hours. Then PP, MAPP, and lignin were mixed manually. The process in the extruder was carried out at 180°C with a speed of 50 rpm for 5 min. After the extrusion process, lignin behaves as a stiffness-inducing filler in the PP matrix, improving the stiffness and thermal properties of the composite in a synergistic manner. In general, there is no significant difference between the use of sugarcane bagasse KL and softwood KL on the characteristics of PP/KL composites.

In another report, Ahmad Saffian et al. [97] developed a composite as heat and thermal insulator material from PP, 30% KL, and 10% kenaf core fiber with 3% polymeric methylene diphenyl diisocyanate (pMDI) as a compatibilizer. Compounding process with mixer was done at a temperature of 175°C, and finally pMDI was added. The resulting compound was then placed in a mold and pressed on a hot press machine to become a composite board. Hot pressing stage, starting with melting compound for 9 min,

Table 2: Description of lignin as filler in lignin-based biocomposites.

Polymer matrices	A common limitation in fabrication	Possible application of lignin-based composites
PLA	The need for compatibilizer	Packaging, food packaging
PVA	The hydrophilic properties of PVA	Packaging, drug delivery system, biomedical, wound healing application, agricultural application
Cellulose	The need for compatibilizer	Seed coating
Starch	The hydrophilic properties of starch	Packaging, food packaging
Epoxy	The need for compatibilizer due to the incompatibility resulted in hydrophilic properties	Coating
Natural rubber	The need for compatibilizer due to the incompatibility resulted in hydrophilic properties	Tire industry, automotive devices
Polypropylene	The need for compatibilizer due to the incompatibility; the reduction in elongation; the susceptibility to microorganism	Automotive devices
Commercial adhesives	Difficulties in particles distribution	Filler in adhesives system

then hot pressing and cold pressing for 3 and 4 min, respectively. The results showed that the PP composite was suitable as a building insulator. However, the composite tensile strength decreased after the addition of lignin or kenaf core fiber into PP. Seydibeyoglu et al. [98] added 10% SL to the PP copolymer, using two types of commercial coupling agents, at a concentration of 0.1%, 0.3%, and 0.5%. The SL was mixed with PP using a twin-screw extruder with a feeding rate of 20%, extruder temperature between 170 and 185°C with a screw speed of 190 rpm. PP/lignin composites as oil absorbers are made using a thermally induced phase separation (TIPS) technique (Figure 2), which is the process of solubilizing a polymer in a high-temperature solvent and then cooling the polymer solution to form a porous structure. PP was dissolved in a decalin and 1-butanol mixture at a temperature of 115°C. Furthermore, lignin was added at 10 and 20% levels. After cooling at 20°C a separate phase (monolith) was formed. After that, it was soaked in acetone and vacuum oven-dried. The absorption of soybean oil and lubricating oil was better in PP monoliths containing 10% lignin than in monoliths containing 20% lignin [99].

Rozman et al. [100] compounded coconut fiber that had been modified with lignin was carried out using a rheomixer at 170°C for 20 min at a speed of 25 rpm. The resulting compound was put into a mold, which was preheated at 170°C for 10 min, then hot-pressed for 10 min, and cold-pressed for 10 min. The amount of lignin used for the modification of coconut fiber varied as much as 0%–20% of the coconut fiber weight. The addition of lignin can increase the flexural strength but decrease the tensile strength of coconut fiber and PP composites, although not as good as using MAPP. This is due to improved compatibility in the interfacial area. Because a large amount of lignin can cause voids and reduce mechanical properties, a small amount of lignin (max 2%) can increase the interfacial adhesion between PP and sawdust [101].

3.2. Composite Lignin-Epoxy Resin. Epoxy is an attractive material because of its wide range of applications, including aircraft components, sports equipment, construction, and many more. Epoxy is a thermoset material that consists of high degree cross-linking and networking molecules [102]. The chain motion in epoxy is greatly restricted and causes excellent chemical resistance, high electrical insulation, and adequate strength [103]. Epoxy, on the other hand, is unsuitable for many applications due to its low thermal resistance, brittleness, and high viscosity [104]. In commercial applications, epoxy resins are frequently mixed with other materials to change their properties. The use of inorganic fillers such as silica [105], zirconium dioxide (ZrO_2) [106], and iron oxide (Fe_3O_4) [107] into epoxy proved to increase hardness, elastic modulus, toughness, and thermal stability. However, to improve the bio-based content in epoxy nanocomposites, lignin is considered an excellent candidate to substitute inorganic fillers and enhance the sustainability and renewability of the material. The presence of various functional groups such as aliphatic, hydroxyl, methoxy, and carbonyl makes lignin has a significant potential to enhance epoxy's toughness and thermal resistance. It also can enhance the mechanical and adhesive properties [108]. The phenolic backbone in lignin makes high thermal stability and is naturally hydrophobic, which differs from other bio-based materials [109].

The bio-based epoxy resin has been prepared by reacting alkali lignin with epichlorohydrin, resulting in a good mechanical property of bio-based epoxy [111–113]. Figure 3 shows the proposed mechanism of an epoxidation reaction involving epichlorohydrin in alkali with lignin to create lignin-based epoxy resins as a promising alternative for bisphenol A in the production of epoxy resins [110]. Lignin reduces the epoxy network density without significantly affecting tensile strengths and thermal properties [114]. Feldmann constantly investigated the influence of lignin incorporation in bisphenol and epoxy resin [115–117]. Feldmann added 5–20 %wt. lignin to bisphenol A diglycidyl ether (DGEBA). The composite with lignin composition between 10 and 20% displayed a noticeable effect on the adhesive tensile strength. Meanwhile, the adhesive shear strength only improved when received heat curing at 75°C for 4 hours. The excellent dielectric properties of lignin/epoxy composites motivated Kosbar et al. to produce printed wiring boards (PWB) [109]. They added 25–67 %wt. lignin to DGEBA with an amine curing agent. In epoxy resin, lignin

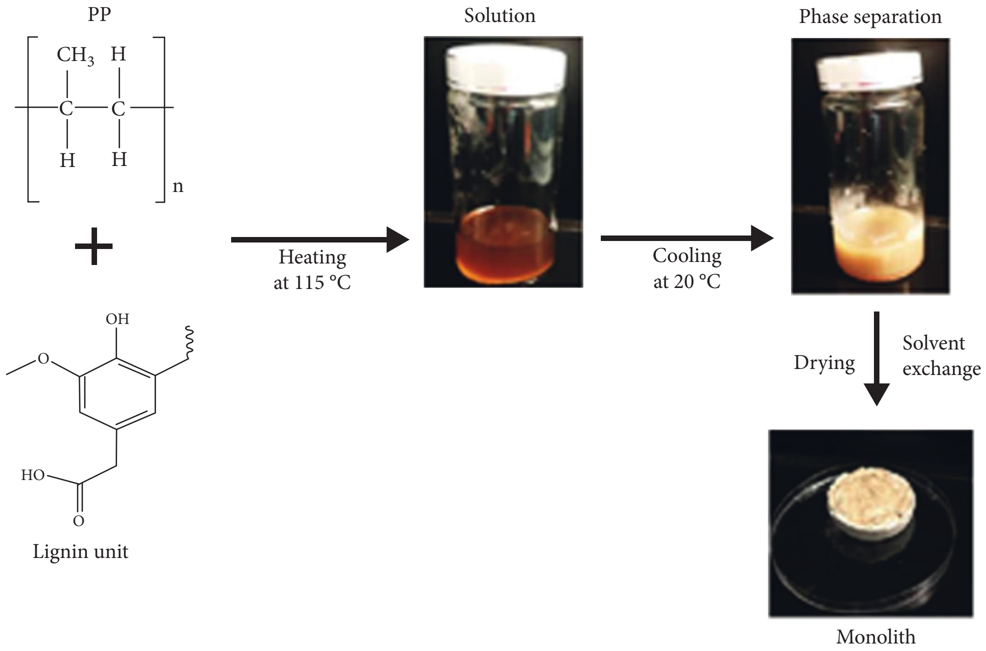


Figure 2: A general protocol for using a thermally induced phase separation technique to prepare PP/lignin blend monoliths [99] (Copyright @ 2020 Elsevier under CC BY-NC-ND 4.0).

can be added to 50% w/w [118]. This addition resulted in properties such as glass transition temperature (T_g), decomposition temperature, and dielectric constant being similar to those of FR4 (glass-reinforced epoxy laminate material), a common resin for FWB. The composite shows an excellent property up until around 60% lignin. Higher lignin content reduces properties but can still be used for lower performance PWB.

Mendis et al. functionalized lignin through hydration of lignin-amine solution and the Mannich reaction [119, 120]. The modification of lignin in this work is not well dispersing the lignin in the epoxy causing a small region of agglomerates. In addition, the water in the hydration sample evaporated after degassing. Incorporating functionalized lignin in epoxy was insignificantly affecting the fracture modulus, but a significant reduction was shown in fracture stress. On the other hand, the strong plasticization effect of water lowered the thermomechanical properties of the Mannich high level and Mannich hydration sample. Liu et al. modified alkali lignin with carboxylic acid via hydroxyl group esterification along with anhydride [89]. The functionalized lignin was mixed with epoxy, which had been cured with anhydride. Introducing 1 wt%. modified lignin into the epoxy resin improved the critical stress intensity factor (KIC) and required strain energy release rate (GIC). The decreased cross-linking density in epoxy was considered proof of the toughening effect of lignin in epoxy resin.

3.3. Composite Lignin Polylactic Acid.

Polylactic acid (PLA) is a bio-based and biodegradable polymer with eco-friendly properties for long-term development, including biodegradability, biocompatibility, and renewable resources [121–123] with solid mechanical properties, and is easy to prepare [124]. PLA can be synthesized from crops such as wheat, sugar beets, and corn [125, 126]. However, PLA has also limitations related to its high cost. Much research was done on PLA with lignin that can act as filler or reinforcement material with emphasis on the investigation of the lignin composition of PLA, either native or modified lignin. The preparation of lignin/PLA composites can be classified into two methods, that is, solvent casting and melt extrusion [127].

The addition of lignin to PLA in the melt extrusion method increased the value of each parameter, whereas the solvent casting method had the opposite effect. This is because of the involvement of lignin particles, which may impair the structure's flexibility due to the internal plasticizers formed by the linking of benzene and ether rings [128, 129]. Because of the hydrogen bonding between the functional groups in lignin and the carbonyl groups in PLA, the melting temperature of the lignin/PLA composites increased. The lignin/PLA composites made from hardwood lignin had the best mechanical properties before and after weathering [12]. Gordobil et al. used acetylated KL (AKL) in a lignin/PLA composite by extrusion at 200°C at a rate of $75\,\text{min}^{-1}$ with a recirculation time of 10 min [130]. Furthermore, some of the hydroxyl groups in the lignin that were added to the blend had an impact on the stability [130]. This method has previously been used to fill lignin/PLA composites with acetylated alkaline lignin and OL from almond shells [126]. According to the study, acetylated lignin/PLA blends are more compatible than unacetylated lignin/PLA blends. Additionally, acetylated lignin can prevent PLA from dissolving hydrolytically. The better dispersion of lignin in the PLA as a matrix may have resulted in a urethane linkage, which resulted in improved PLA/lignin blend reinforcement [131]. Besides the solvent casting

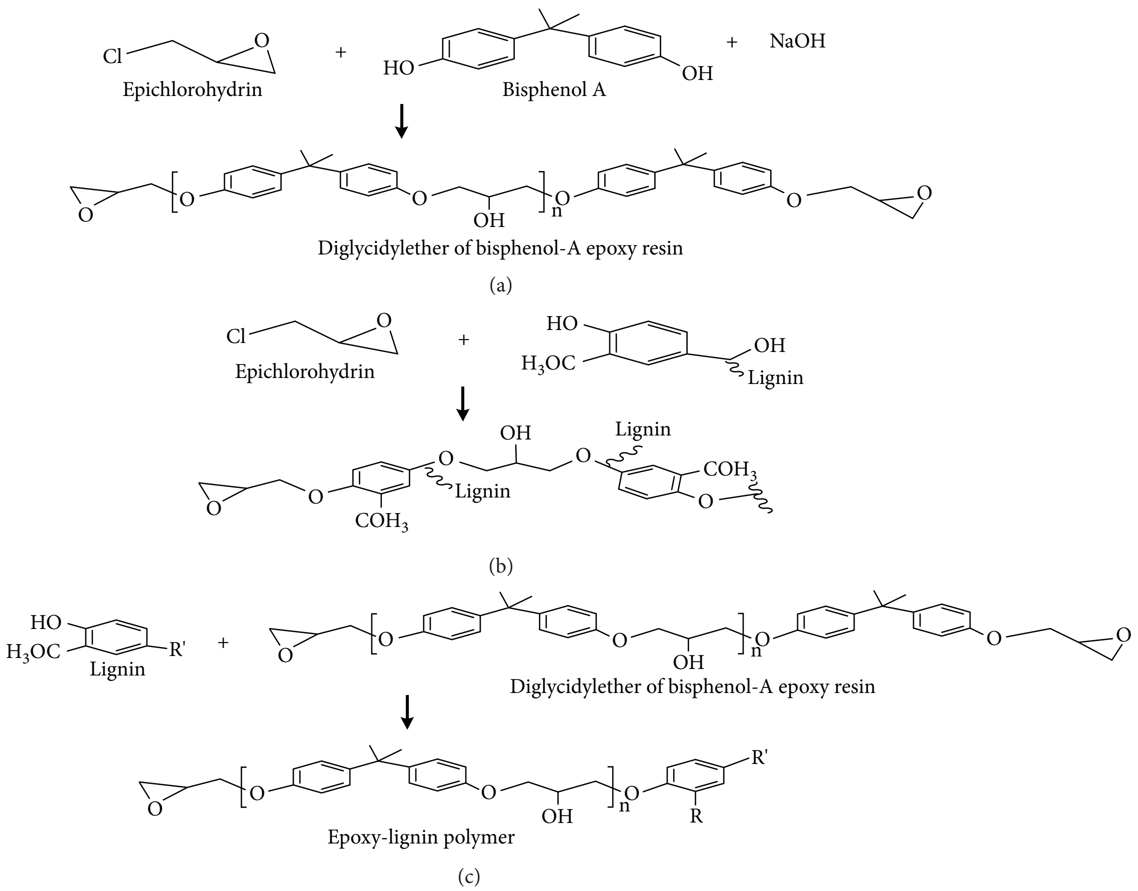


Figure 3: Proposed pathway in synthesis of lignin-bisphenol A epoxy resin: (a) bisphenol A-epoxy resins, (b) cross-linking of lignin with epichlorohydrin, and (c) lignin-bisphenol A epoxy resin [110] (Copyright @ 2017 Springer-Verlag GmbH, Germany).

method, melt extrusion has been employed as a method to incorporate lignin nanoparticles into PLA. The uniform dispersion of lignin nanoparticles was found at 1% but not in 3% wt causes the aggregation of lignin nanoparticles with PLA [127].

The solvent casting and melt extrusion method resulted in a different surface structure [127]. The extruded film sample had a morphology with a smoother surface due to the equal distribution of lignin in the PLA matrix. Solvent casting produces a sample with a lot of microholes and a less uniform surface. When lignin/PLA composites with KL and AKL were compared to PLA/nonmodified lignin, the PLA interaction with AKL showed a significant difference [130]. Zhu et al. investigated unmodified lignin to be use as an adsorbent in lignin/PLA composites prepared by solvent evaporation–cross-linking. The microstructure and compression strength of the absorbent are influenced by the lignin/PLA composites ratio [132]. Using a twin-screw extruder, Park et al. looked into adding KL and its plasticized form to a PLA blend with a pMDI as a coupling agent [131]. Adding lignin to composites as a biodegradation agent has also been studied. The addition of KL and its plasticized form to a PLA blend with a coupling agent pMDI was investigated by Park et al. PLA was melted and mixed with a variety of KL concentrations. A high-speed mixer spinning at 3,000 rpm was used to create the composites. Because of the high friction between the polymer and the rotor, mixing, melting, and homogenization are all possible. According to the findings, lignin may aid in the biodegradation of lignin/PLA composites [133]. It has previously been reported on the effect of using lignin from hardwoods and softwood for lignin/PLA composites to accelerate weathering [12].

As a source of esterified lignin, Vila et al. recovered the liquid phase of organosolv pulping catalyzed with H_2SO_4 from *Pinus pinaster* wood. The extruder was used to blend lignin and esterified OL to PLA in a 30:70 mass ratio [134]. Ye et al. reported the effect of different ratios (5, 10, and 20%) of desulfonation LS or LS particles in lignin/PLA composites (DLS) using a casting method and then drying them at room temperature for 24 hours [135]. Zhang et al. developed PLA composites with lignin-silica hybrid (LSH) as the inflammable flame retardant (IFR). LSH is blended with PLA and ammonium polyphosphate (APP) in a two-roll mixing mill at 180°C for 10 min at a speed of 40 rpm. The samples were then compressed and formed into sheets [136]. Li et al. [137] investigated a Pickering emulsion method for preparing lignin/PLA composites by blending lignin nanoparticle suspension with PLA and dichloromethane solution (DCM). The PLA/lignin films were compression molded at 180°C for 5 min using a 20 MPa pressure. The lignin dispersion was improved by over 5.0 %wt. using this Pickering emulsion method. The performance of PLA composites filled with unpurified, purified, and acetylated OL from corn stover was compared by Gao et al. [138]. When compared to treated lignin, untreated lignin/PLA composites can be used as a filler in PLA composites with comparable or better performance. The use of unpurified lignin as a filler in PLA can lower the cost of purification.

3.4. Lignin-Cellulose-Based Composite. Lignin is an amorphous thermoplastic polymeric substance with a brittle property at high temperatures and a proclivity for not forming a film in solution. Lignin has a high carbon content, has high thermal stability, is biodegradable, has good antioxidant action, and is powerful [42]. Lignin is made up of hydrophilic groups that are surrounded by hydrophobic chains for the most part. The hydrophobic carbohydrate components of cellulose are often incompatible with hydrophobic chains. The most prevalent polymer, cellulose, has an extended linear chain structure made up of (1,4) connected glucopyranosyl-D units that form a single microfibril and polyhydroxy compound that may be joined with other materials via hydroxy hydrogen bonds [42, 139]. The effect of lignin in the drying and dehydration of nanocellulose was investigated by Kim et al. incorporating lignin reduced aggregation, redispersion kinetics, and stability, as well as allowing for reversible restitution during drying and polymer processing [140]. It is important to add ionic liquid using the blending solution technique or by synthesizing the composite membrane with other solutions to mix cellulose and lignin as a composite material.

Some methods have been reported to prepare cellulose-lignin-based composite, such as the solution casting film technique [141], vapor-induced phase separation method [142], regeneration in acetone [143], aqueous suspensions alkaline [144], blending solution [145], bio-membrane amino-modified [146], and nanocomposite coatings [147]. Cellulose, starch, and lignin are combined in an 85:10:5 ratio in a solution of 1-allyl-3-methylimidazolium chloride, and the fibers are constantly agitated at 80°C. The solution is put onto a glass plate, which is then placed in a vacuum oven with evacuated air and a transparent membrane that has been cleaned with flowing water [141]. Separately dissolved cellulose triacetate and lignin esterified nanoparticles were prepared. The two solutions are then mixed and placed onto a glass plate after being swirled for 2 hours at 25°C. The solution was applied for 2 hours at a regulated temperature and humidity, then removed off the film using a water bath, and dried at room temperature [142].

Sadeghifar et al. [143] developed and characterized semitransparent flexible cellulose-lignin UV-light-blocking films using a simple technique. The regeneration of cellulose-lignin films in acetone was used. Unlike simple mixtures of cellulose and lignin, these covalently bound cellulose-lignin films were homogenous. The UV protection of the prepared films was excellent. UV-B protection was 100%, and UV-A protection was above 90% with a cellulose film containing 2% lignin. Cellulose nanofiber (CNC) film was created by suspending CNC in a NaOH solution, then adding alkali lignin and KL from softwood at a concentration of (1–10 % wt.). A combination of alkaline aqueous suspensions having appropriate NaOH concentrations is used to make CNC/alkali lignin and CNC/softwood KL films [144]. Bionanocomposite was prepared by melt mixing compounding process of unbleached nanocrystalline cellulose-containing 90% lignin with PP and MAPP as matrix and coupling agent and kenaf core as reinforcement. The presence of residual lignin increases the thermal stability and flammability of nanocomposite [148]. Previously, it was confirmed that higher lignin in CNC contributes to the better thermal stability of CNC which give advantage for promoting the development of nanocomposites [149].

As an additional cross-linker, tannic acid and epoxidized KL was developed to introduce epoxy groups into the lignin to yield the bio-renewable waste cellulose paper (Cell). Cell-epoxy lignin membrane was molded at 80°C for 4 hours with a load of 20–30 kN. At 140°C for 1 hour, a similar load was used to mold the Cell-epoxy lignin-tannic acid membrane [146]. New nanocomposite coatings based on CNC and synthetic/fractionated spruce and corn stalk lignin were created without any chemical modification or functionalization (through covalent bonding) of one of the two CNCS and lignin. Nanoparticles increase the weight ratio of the medium and the CNC-specific surface area of the solid film, allowing more lignin molecules to disperse throughout the solid film. The π-π aromatic aggregates dislocate as a result of the increased molecular expansion of these aromatic polymers on the surface of the cellulose nanoparticles, boosting the extinction coefficient and decreasing UV transmittance. At visible wavelengths, the nanocomposite coverings were optically transparent [147].

3.5. Lignin-Polyvinyl Alcohol Composites. PVA is a biodegradable polymer material that has high strength, chemical stability, and abrasion resistance, as well as biocompatibility [150, 151]. PVA is a polar polymer composed of mostly hydroxyl groups in its polymer chain bonds and has solid hydrophilic properties. On the other side, lignin has polar functional groups and has strong hydrogen bonds between molecules. Based on its properties, theoretically, the combination of PVA and lignin has good compatibility in conversion as a composite membrane.

Melt blending was reported for preparing lignin/PVA flame retardant composites. Adding lignin to PVA has proven to be an efficient way to create ecologically friendly and flame-retardant polymer materials, and the use of PVA and lignin will continue to expand [151]. PVA/poly (glycerol sebacate)/lignin composite was reported suitable for tissue engineering [152], while PVA/lignin nanofibers introduced by silver nanoparticles have been used for wound-healing application [153]. A lignin/PVA nanofiber membrane has been created as a separator for lithium-ion batteries using a water-based approach. To create a nonwoven membrane, PVA and lignin were dissolved in water, and the fibers were removed. As a result, the lignin/PVA membrane can be used as a separator in a variety of anodes for LIBs that are safer and faster [154].

The PVA/salt of the lignin sulfonic acid hybrid membrane was used for preparing the cation exchange membrane for the diffusion dialysis process to recover alkali [150]. Before introducing lignin, sulfonic acid, and HCl with tetraethoxy silane (TEOS) as a cross-linker, PVA was diluted in dimethyl sulfoxide (DMSO) to obtain a homogenous solution. To create the membrane, a homogenous slurry was thrown over just a glass plate at 60°C for approximately 1 day. The sustainable copolymer lignin-rubber-PDLA was produced in two steps employing ring-opening polymerization. To create the composite in chloroform, PVA was mixed with alkaline lignin-g-rubber-g-poly (D-lactide; PDLA). Alkaline lignin dissolves in aqueous alkali at room temperature [155].

The film casting method was investigated by Xu et al. [156] for preparing membrane by mixing alkaline lignin and PVA. Introducing lignin into PVA improves the thermal stability, oxygen, and carbon dioxide barrier, and visible light at 600 nm blockage in which the optimum content of alkali lignin was 15% for blending with PVA. This composition is suitable for green packaging materials. A similar technique was also used by Korbag and Mohamed Saleh [157] in which dissolved lignin was dropped into acetic acid and heated for 2 hours at 80°C. After that, PVA was mixed and stirred for 6 hours at 60–70°C before the drop in the Petri dish. The investigation of the chemical structure of PVA/lignin film presented strong intermolecular hydrogen bonds between PVA and lignin. In such studies, the introduction of a coupling agent is required to increase the tensile strength while maintaining the composite's elastic modulus. PVA biocomposite was synthesized by diluting PVA in Millipore water and agitating it with CNC and lignin nanoparticles before adding glutaraldehyde as a cross-linker and sonification. Figure 4 depicts the intermolecular reaction that occurs between CNC, PVA, and lignin nanoparticles.

3.6. Lignin-Starch Composites. Starch is a low-cost, available, biodegradable, renewable, and flexible thermoplastic that makes it considered a composite in packaging, engineering applications, and biomedical materials [14]. However, hydrophilic properties with poor mechanical and permeability characteristics need to improve by adding filler and enhancing agents such as lignin [159, 160]. Lignin has both hydrophilic and hydrophobic groups, making it suitable for organic and inorganic polymers. Starch composite can be prepared by the extrusion process under high temperature and pressure being added plasticizer such as water or glycerol [161]. However, the heterogeneous and nonuniform structure of lignin limited its application in starch film [162]. The lignin of about 3–10% has been utilized as reinforcement in the starch film by mixing the starch solution with lignin solution by casting technique [163–166]. SL has been utilized to improve the characteristics of lignin-corn starch biofilm blends. By solvent casting, the starch was combined with lignin and was heated to 100°C and then was added glycerol, continued by heating and drying [159]. This method was also used to blend corn and cassava starch with SL as a filler material to prepare biofilm with the addition of glycerol [167].

For the development of starch-lignin composites, fractionalized SL was used. The ultrafine friction grinding process was used for reducing SL size reduction before use as a filler in starch-based biocomposites [161]. The starch/KL foam was prepared by compression molding with no deleterious effect on the density or morphology of the film when 20% lignin was incorporated into the starch. The blending solution approach has been used to create starch-based films and membranes [168]. By introducing corncob, SL (20–40 %wt.), thermal compounding was used to fabricate a thermoplastic starch (TPS)/poly (butylene adipate-co-terephthalate; PBAT) composite. Lignin acts as a reinforcement, improving the composites' compatibility with the TPS and PBAT phases, as well as their hydrophobicity and water repellency [169]. The TPS-lignin film was prepared by extrusion method by a single-screw extruder continuing irradiation with an electron beam accelerator. Introducing lignin (LS/alkali lignin) in the film improves the hydrophobic surface properties without loss in biodegradability. It is affected by the role of lignin for radical cross-coupling reactions to form hydrophobic condensed networks [164].

3.7. Lignin-Chitosan Composites. Chitosan contains high amino and hydroxyl groups, while lignin has high hydroxyl and carboxyl groups. Chitosan-based composite has been considered in food packaging and biomedicals such as tissue engineering, drug delivery, and biosensors [170]. Chitosan is a biodegradable and biocompatible material extracted from crab shells. It is also appealing because of its antibacterial and antioxidant qualities, but there are several drawbacks to composite utilization, such as high cost, low water resistance, poor surface area, very acidic solutions, antioxidation, poor thermal stability, and low mechanical capabilities [170, 171]. Some materials such as lignin and PVA can be added to the system. The mechanism to combine two polymers is van der Waals' force or hydrogen bonding [172].

The solution casting method was commonly used in lignin-chitosan functional composite preparation [170, 172]. By this method, a chitosan film with the inclusion of micro ramie fiber and lignin has been created [170]. Mixing ternary and a binary film containing PVA-chitosan and lignin nanoparticles have been prepared via solvent casting for packaging application [171]. Modification of solvent casting method in water/ethanol solvent system was also performed by Crouvisier-Urion et al. [173]. Using the solution casting approach, a membrane comprising chitosan and lignin was successfully used to remove hazardous dyes from wastewater effluents, including methylene blue dye, by up to 95%. Membranes can be used up to five times before they need to be replaced [174]. The blending technique of lignin and chitosan has been carried out in the study of Li et al. [42].

Cross-linking between hydroxymethylated lignin and chitosan was reported in the preparation of composites by Shiyan et al. [175]. The other method involved mechanically fibrillating unbleached tree bark after alkaline extraction to produce lignin-containing cellulose nanofibrils (LCNF),

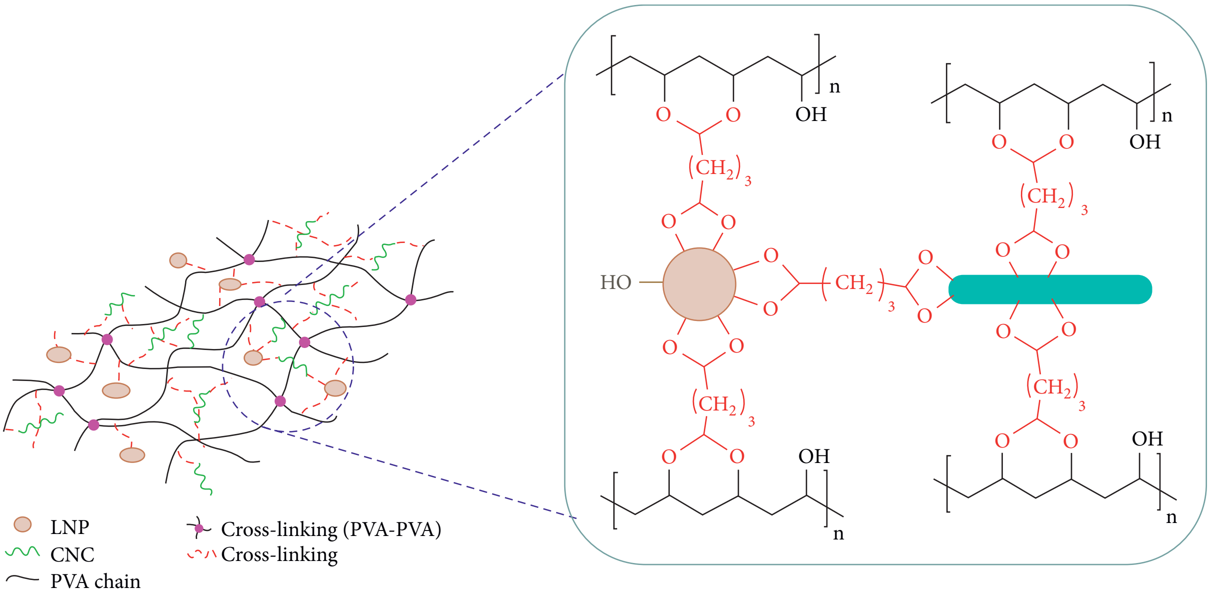


Figure 4: The proposed cross-linking reaction of polyvinyl alcohol, cellulose nano cellulose, and lignin nanoparticles uses glutaraldehyde as a cross-linker [158] (Copyright @ 2020 MDPI under CC by 4.0).

which were used as a reinforcement in TPS to create innovative biodegradable composite films. Lou et al. [176] looked into it using a microwave-assisted synthesis of chitosan-acrylamide-lignin terpolymer. Mechanical fibrillation of unbleached tree bark followed by alkaline extraction resulted in the formation of lignin-containing LCNF, which was used as a reinforcement in TPS to create newly biodegradable composite films. Lignin contributes to increase water barrier properties and thermal stability of TPS-LCNF composites [177]. The composite hydrogel containing chitosan and lignin has been prepared for wound dressing in which its mechanical improvement when lignin was introduced [178].

3.8. Lignin-Based Rubber Composite. Silica and carbon black are typical reinforcing materials in rubbers [179, 180] to improve the mechanical and thermal properties of the compounds while lowering the cost and, in some cases, the weight. However, due to pollution concerns and the dark color of carbon black, lignin, which has high availability, annual renewability, and low cost, is being considered as a possible replacement for the carbon black [181, 182]. In rubber composites, natural rubber (NR) is a common matrix [183, 184]. The combination between NR and lignin was a good combination to produce a green composite, especially in the tire industry. The addition of filler to NR alters its character due to a change in the system's polarity level. Furthermore, bio-filler and rubber are incompatible due to the lack of polarity in rubber [185]. Fillers (lignin and silica) can work together to prevent the formation of a filler network. In exchange, silica facilitates the dispersion of lignin in the rubber matrix. Various types of lignin were found in this study, including KL, SL, calcium lignosulfonate, and sulfate lignin [186–188]. Coupling agents such as maleic anhydride (MA) were grafted onto the polymer backbone to enhance interfacial bonding and mechanical properties.

The tensile strength of natural rubber composites improved by about 10% when maleated natural rubber was produced in internal mixing [189–191]. The disadvantages of current techniques are that they take time and are not eco-friendly because they use solvents. Due to the high surface energy of rubber matrices, obtaining a homogenous distribution of lignin particulates is difficult. As a result, by covalently bonding incompatible polar fillers and nonpolar rubber in rubber composites, MA has the potential to improve interfacial adhesion [192–196]. The interfacial adhesion of the wool fabric and NR latex was improved using sodium lignosulfonate (SLS). SLS plays a role as cementing materials in wool fabric and rubber latex and then was applied in varying amounts to the coarse wool is a woven fabric. SLS treatment improves the density and UV resistance of rubber-based composites. Composites have a broad range of applications, including acoustic panels, mats, and bags [197]. To increase the hydrophobicity of lignin and thus its dispersion in the rubber system, lignin has been modified with silylation. After incorporating 5 %wt. silylated KL into the natural rubber matrix, it can result in a 44.4% improvement in tensile strength. The greater the amount of modified lignin in the filler, the higher the elastic moduli and Payne effect intensity [198]. By mixing KL, OL, and SL into natural rubber composites in a lab-scale two roll mill, better properties were obtained than with soda lignin. The Payne effect was reduced after lignin was added, and their processability, aging resistance, and thermal stability all improved. The filler loading ranged from 5 to 20 phr, with a total hybrid filler content of 50 phr [188].

4. Lignin as Filler in Wood-Based Composite

The main concern is the long harmful effect of formaldehyde emission in the use of traditional formaldehyde resin such as urea-formaldehyde (UF), melamine-formaldehyde (MF), or phenol-formaldehyde (PF) have motivated the researchers to find renewable alternative sources to substitute the adhesive in the wood composite system. Lignin with phenolic structure has been reported prospective as a substitution in wood composite adhesive for creating eco-friendly composites. Despite its complex structure, low reactivity, low solubility, high polydispersity, and strong hydrophobicity, lignin's chemical reactivity is limited, necessitating greater catalyst concentrations and longer heating durations in the manufacture of wood-based composites [33, 132, 199–204]. Chemical modification of lignin, mixing with additional bio-based materials such as starch, tannin, polyethyleneimine, or glyoxal, is a way to improve the performance of lignin-based adhesive [199]. Some different lignin types have been used such as LS, KL, OL, SL, or hydrolysis lignin [18, 201, 205–207] in adhesive applications of plywood, oriented strand board (OSB), fiberboard, particleboard, or structural application.

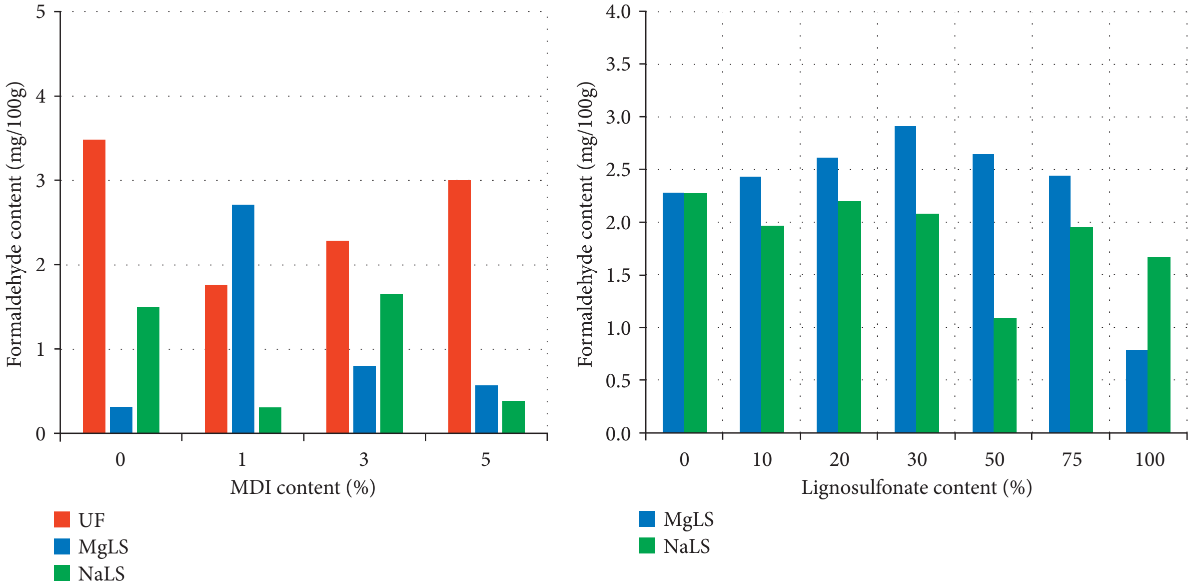
OL was able to substitute phenol in PF adhesives for particleboard by up to 30% [208]. Magnesium and sodium lignosulfonates (LS) of 30% were introduced in UF adhesive formulation as particleboard that has comparable properties to UF-bonded particleboards [205]. Savov and Antov [209] investigated the use of LS as a medium-density fiberboard adhesive with the optimum content without deterioration the board strength was 35%. Ammonium LS was also used as a bio-based additive in high-density fiberboard in which it was mixed with UF resin at varying levels from 4 to 8% on dry wood fibers mass. The application of up to 6% ammonium LS decreases the mechanical properties of wood fibers impacted by the short press factor while also increasing moisture content [18]. Those studies revealed that LS acts as a formaldehyde scavenger [18, 205] as presented in Figure 5.

In another study, magnesium lignosulfonate was used as fiberboard binder for limited structural application with 15% gluing content based on the dry weight of the fibers [207]. Because of the lower resin reactivity and longer reaction time, lignin can be used to substitute phenol below 50% [201]. Lignin reacted with glyoxal resin and furfuryl alcohol and then was added by variation epoxy resin for particleboard adhesives as reported by Zhang et al. [210]. KL was employed up to 35% in the stein hall process to create starch-based adhesives for use in paperboard. Water resistance and strength of paperboard products are both improved by this addition [211]. KL and SL were also used to substitute lignin-phenol-glyoxal with various weight percentages of 10–50 % wt. with which the optimum substitution at 30% [212]. For altering soy flour adhesives, a lignin-based adhesive was made by batch copolymerizing lignin, formaldehyde, and phenol. It was then employed to improve the wet and dry bond strength of plywood adhesives [213]. In the preparation of dry-process fiberboard with modification pressing technique, 10% dilute sulfuric acid hydrolysis lignin (based on dry fiber) was combined with 2% PF resin. The board possesses physical and mechanical qualities that are comparable to board bonding with 10% PF during a standard hot-pressing cycle [214]. Enzymatic hydrolysis lignin was incorporated with NR latex to prepare bio-adhesive by filler and blending base component method as medium density fiberboard adhesive. Mixing 5 and 10 g lignin into 10 g natural rubber latex provides the highest modulus of elasticity and modulus of rupture value of medium density fiberboard, respectively [215].

5. Characterization of Lignin-Based Greener Composites

5.1. Morphological Analysis of Lignin-Based Composites. A morphological study of lignin-based composites was carried out by analyzing SEM micrographs. Sahoo et al. [216] reported that the SEM micrographs of lignin obtained from Arboform F45 showed the presence of lignin fragments (5–10 μm) and dispersed fibers (20–150 μm). Fibers are connected with varied proportions with flat and uneven textures, suggesting the existence of diverse varieties of fibers in the lignin group. While on the other hand, SEM photographs of the lignin-based poly(butylene succinate) (PBS) composite revealed that the surface morphology of the broken surface of the lignin-PBS composite showed relatively smoother and uniform phases. This could indicate that lignin is more compatible with PBS; this affinity could be explained by the alignment of solubility characteristics and the possibility of polar-polar interaction; the lignin-PBS compliance supports the increased Tg of lignin composites. In the micrograph of the broken layer, deformations and cavities with irregular forms were also visible, indicating a stronger polymer-filler interaction in the polymer matrix. Filament pull-outs having attached polymer matrices are marked by these cavities. In the SEM image of lignin-PBS composites, plucked-out fibers can also be seen precisely. Crushed fibers (short strands) in the lignin encouraged easy fracture deformations via a short fiber pull-out process, which greatly contributed to the composites' poor mechanical characteristics. The size of pulled-out fibers from lignin-PBS composites influences this effect.

Graupner [217] investigates the effects of lignin on the mechanical properties and sustainability of natural fiber-reinforced polymer composites. Lignin plays a critical role as a naturally occurring adhesion stimulant. Furthermore, the hypothesis suggested that lignin promotes matrix-reinforcement adherence in cotton fiber-based biocomposites. Cotton was an excellent choice for this project since it contains no lignin. According to the results of the SEM study, the presence of lignin increased adherence between fiber and resin; lignin also strengthens the interaction between constituent layers of multilayered networks. The unmodified cotton/PLA materials had obvious deformations between constituent layers of multilayered networks; the lignin-treated cotton fiber composites showed few imperfections. Wang et al. [218] studied the morphology of PLA/lignin-containing cellulose nanofibrils (L-CNFs) composites; they analyzed the SEM photographs (Figure 5) and



(a)

(b)

Figure 5: (a) Formaldehyde content of board by using magnesium lignosulfonate and sodium lignosulfonate adhesives with a variation of MDI cross-linker and (b) formaldehyde content of board at the variation of lignosulfonate content [205] (Copyright @ 2021 MDPI under CC BY 4.0).

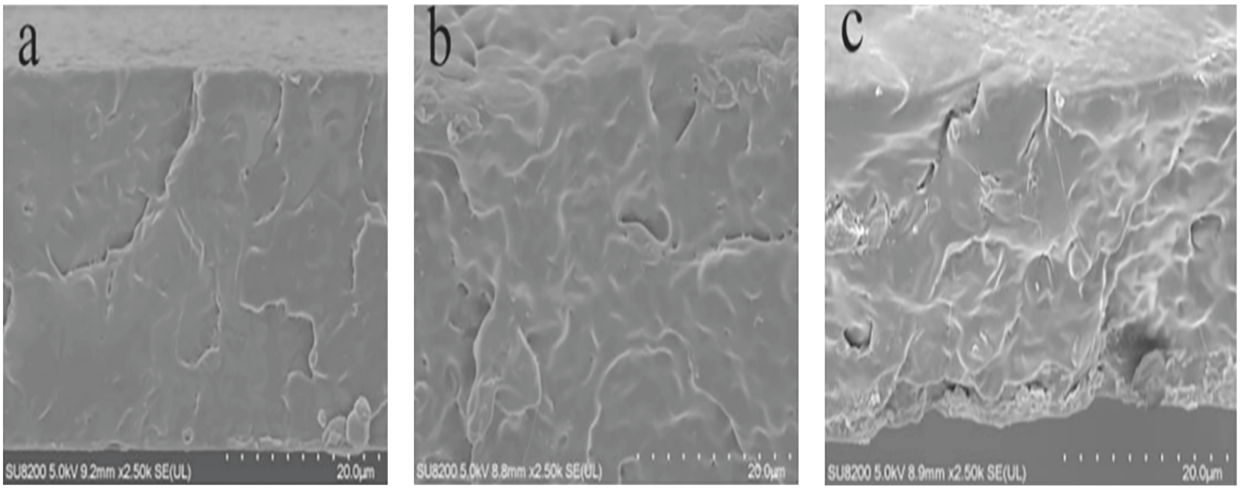
concluded that L-CNFs improve the strength and embrittlement of nano-composites. Furthermore, L-CNFs significantly impact the composite stiffness compared to CNFs. Another study on composite SEM reveals that lignin, organosolv, and LigninBoost lignin can be well distributed in PLA matrix during melt blending in which lignin and PLA have good adhesion [12]. The addition of lignin to PLA seems to have a lousy affinity because it tends to agglomerate with some cracks (Figure 6) displaying poor interfacial compatibility.

Guo et al. [219] analyzed morphology changes that occur due to chemical modification of lignin by reacting lignin with different compounds; they studied how chemically modified lignin causes changes in the morphology of biocomposites. Lignin was modified with acid (Ad), acetoacetate (Act), and butyric (Bt), respectively, and their morphology and compatibility with PLA matrix were examined from SEM micrographs (Figure 7). The particle sizes of modified lignin ranged from 0.5 to 2 μm, although the particle size of Act-lignin was substantially lower than that of other types of modified lignin. The morphology of lignin fibers influenced the degree of anisotropy. All lignin fragments seemed to be fairly spherical, showing that variable levels of lignin aggregation resulted in the formation of diverse microspheres. Simultaneously, the lignin size influenced the surface area per unit volume, leading to strain transmission and composite filler wetness changes, thus altering and controlling the lignin filler's compliance and homogeneity with the polymer composite. Modification of lignin with alkylation has been reported by Maldhure et al. [96] in which the addition of more than 15% alkylated lignin into PP composite causes agglomeration (based on SEM analysis) so that the crystallinity decreases.

Figure 7 depicts the morphological characteristics of PLA/chemically modified lignin biomaterials. The introduction of lignin into the pure PLA increased the irregularity of the composites, leading to a variety of fractions and, as a result, to more regions of vulnerability throughout the composite materials, potentially lowering their tensile strength. Furthermore, chemically modified lignin is evenly diffused and fully integrated into the composite material; the resilience of slightly larger diverse particle sizes is influenced by the degree of permeability and interfacial bonding between PLA and chemically modified lignin [220]. The interface of PLA/lignin biomaterials was noticeably harder and stronger than those of composites without lignin [221]. When Act-lignin was added, a significant characteristic that can be noticed was the presence of distinct phases with recognizable lignin aggregates, whereas in the case of Ad-lignin or Bt-lignin, a considerable number of lignin fragments pierced from the surface, showing poor connectivity among PLA and Ad-lignin or Bt-lignin. Incorporating Act-lignin into the PLA matrix enhances dispersal [218], indicating the greater contact and adherence between PLA and Act-lignin, which establishes a stronger interface region [222]. Due to the increased interfacial contact of matrix filler, PLA/Act-lignin biocomposites demonstrated superior characteristics to other composites. However, a different SEM result was reported Mendis et al. [119] in which the lignin modification cannot disperse well, as shown by the agglomeration of lignin in epoxy (Figure 8).

5.2. *Effect of Lignin Concentration on the Morphology of Biocomposites.*

Hong et al. [223] analyzed the surface morphology of lignin-based biocomposites containing lignin in



PLA (a)

PLA/CNFs (b)

PLA/L-CNFs (c)

Figure 6: SEM photographs of (a) polymer, (b) polymer composite without lignin, and (c) lignin-containing biocomposite [218] (Copyright @ 2018 MDPI under CC BY 4.0).

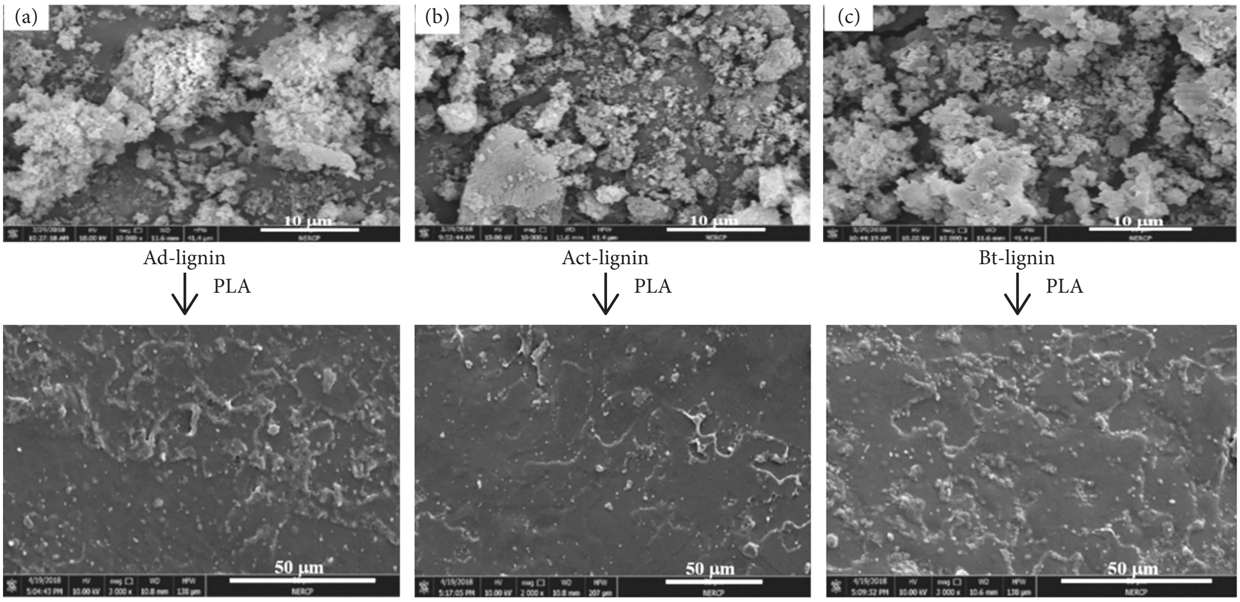


Figure 7: SEM images of biocomposites of chemically modified lignin: (a) PLA/Ad-lignin, (b) PLA/act-lignin, and (c) PLA/Bt-lignin [219]. (Copyright @ 2019 MDPI under CC BY 4.0).

different concentrations. They observed that with increased lignin concentration, the surface of the filaments on the 3D printed items grew noticeably rougher and deeper in color. This is owing to the decreased melt strength of the biocomposites. The interface texture of the biocomposite fibers having more than 15% lignin was too rough to bond layer-by-layer, resulting in a large gap. It is due to the diminished melt flow and the lack of adherence between layers. Filler dispersibility and interfacial adherence to the matrix material are important elements in defining the filler-reinforced composite's ultimate physical quality. SEM results demonstrate the influence of lignin on the matrix interface. Furthermore, the findings show that size abnormalities and fiber dispersion may have a greater impact on the quality and compatibility of biocomposites. In lignin-based composites, water absorption and thickness swelling were reduced [100]. The more lignin added, the rougher the cross-sectional morphological surface became.

The presence of hydroxyl groups in lignin facilitates it to link to PLA, resulting in strong interfacial interaction and enhanced mechanical characteristics [224]. Dias et al. [92] demonstrated that the matrix could survive lignin loading at a low lignin level according to the SEM pictures. The mechanical characteristics of uncompatibilized and compatibilized composites are strikingly comparable; Figure 9 shows the SEM image of softwood KL (SKL) and

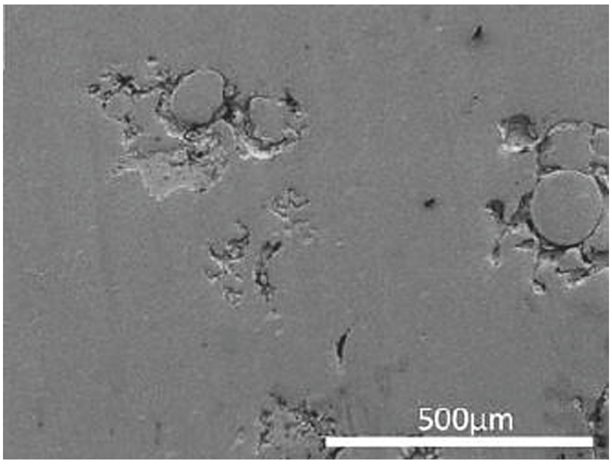


Figure 8: SEM figure of the composite sample that was mixed with a hardener in only 5 minutes (IMM) [119] (copy with permission from Wiley).

polypropylene grafted with maleic anhydride (MAPP) composite of SKL.

The SEM micrograph demonstrated that the integration of lignin caused an irregular and hard interface. Lignin aggregation was most likely driven by the increased density of hydroxyl groups with increased lignin content. The image of the lignin-loaded composites with MAPP, on the other hand, shows a more homogenous dispersion of lignin particles. Increased lignin concentration raises the plasticizing effect; thus, it causes chemical alteration, suggesting that interchain interactions alter the physical characteristics of the composite. The rough surface was observed in all lignin-containing biocomposites with varied lignin concentrations. Microspheres and many aggregates scattered on the interface of lignin blends indicate the presence of lignin. Lignin blends have a finer interface because the dispersion size of the lignin is decreased. While the morphological analysis of polystyrene/lignin composites with compatibilizer addition revealed that adding lignin made the cracked surface of the composites seem rougher [225]. Another study reported that incorporating lignin into chitosan until 20% resulted in substantial changes in the chitosan composite morphology. The more porous and textured composite surface was presented, and an increase of pores occurred with an increase in lignin content resulting in a more pronounced [9]. Previously, Chen et al. [172] reported the good dispersion of lignin (until 20% wt.) into chitosan confirmed by SEM analysis contributed by the strong interfacial interaction. When compared to composites containing citric acid (caLNP) and acetylated lignin (aLNP), the PLA composite with 1 %wt. unmodified lignin showed higher particle aggregation. Furthermore, no evident effect of lignin chemical alteration was seen in composites with a filler content of 3% wt. [226].

5.3. *Effect of Temperature on the Morphology of Biocomposites.*

The influence of temperature on the morphology of lignin-based biocomposites was investigated by Tanase-Opedal et al. [227]. Experiments and surface morphology (Figure 10) proved that 215°C temperature was the best for the applications of lignin-based biocomposites in the printing area because this temperature may boost interlayer adhesion and mechanical strength. SEM images of the lignin-based biocomposites at 205°C showed distinctly marked imprinted fibers, suggesting poor interlayer adherence and limited mechanical characteristics. On the other hand, raising the temperature to 230°C resulted in a drop in mechanical performance. This might be due to the degradation of carbohydrates into gaseous products present in the lignin fraction, thus generating microstructures inside the biocomposites.

Rahman et al. [228] reported that the lignin-based biocomposite's surface has fracture toughness characteristics. All lignin compositions in the polymer matrix had a decent dispersal of lignin fibers. This is most likely due to strong lignin particle adherence to the polymeric matrix. Furthermore, adherence improved for materials produced at higher temperatures. Figure 10 showed that the synthesis of the lignin-based biocomposite, poly-L-lactic acid (PLLA) at higher temperatures increased the tensile strength and roughness of the surface.

5.4. *Effect of Catalyst on the Morphology of Biocomposites.*

The catalyst has no discernible influence on the morphology of the composites. Moreover, water absorption tests on biocomposites in the presence of an aqueous medium might provide useful information regarding the influence of the catalyst on structural strength. In the preparation of PP/lignin composite using a melt mixer, MA-modified lignin was well dispersed in PP up to 25%, while dichloromethane-modified lignin was only up to 15% based on morphological analysis [96]. Yu et al. [229] investigated the catalytic effects of nickel (cobalt or zinc) acetate on the properties of PP-based lignin composites (PP/N-lignin); ($Ni(Ac)_2$) catalytically destroyed PP/N-lignin while significantly increasing char by-products. Ni (0) and NiO also increase carbonization via the catalytic activity of the composite material and seem to be principally accountable for the PP/N-lignin system's improved physical and chemical properties. SEM photos of PP/N-lignin showed several large cracks, while PP/N lignin-containing nickel has no cracks with a relatively smoother surface (Figure 11).

5.5. *The Effect of Particle Size on the Compatibility of Lignin with the Matrix.*

The reduction of particle size of lignin in the nanoscale with an ultragrinding process contributes to improving the compatibility between lignin and starch in this composite preparation based on microscopic analysis and membrane filtration [161]. Before extrusion with PLA, caLNP and aLNP lignins were improved dispersion, minimized aggregate size, and improved performance quality. The size of lignin aggregates in the PLA matrix is determined by the compatibility of lignin and PLA, and it decreases as phase compatibility improves [226].

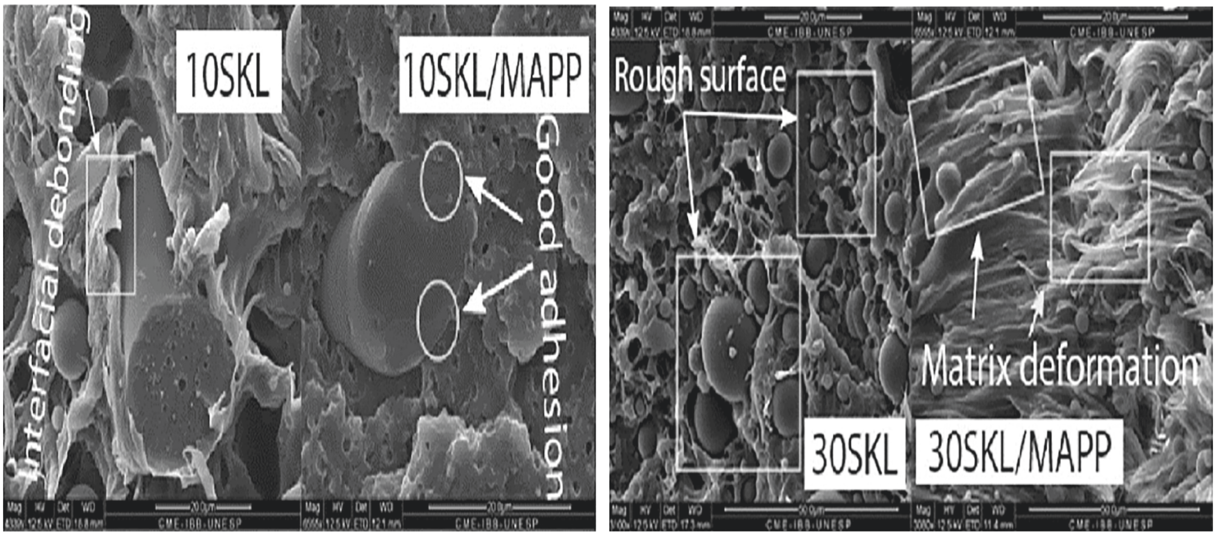


Figure 9: SEM photographs of lignin and lignin-based biocomposites containing different concentrations of lignin [92] (Copyright @ 2018 John Wiley & Sons).

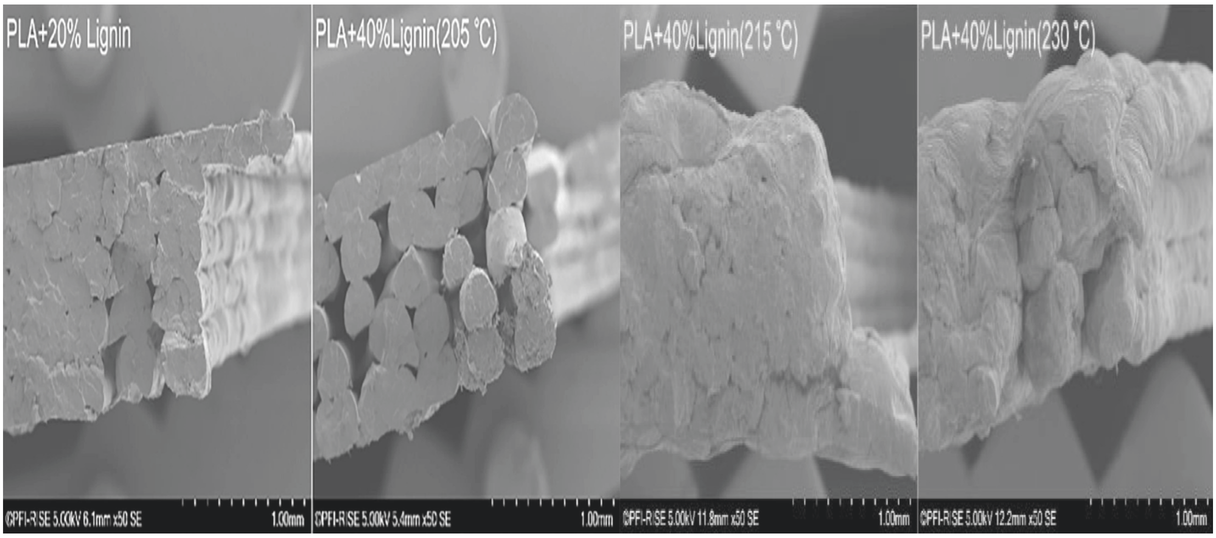


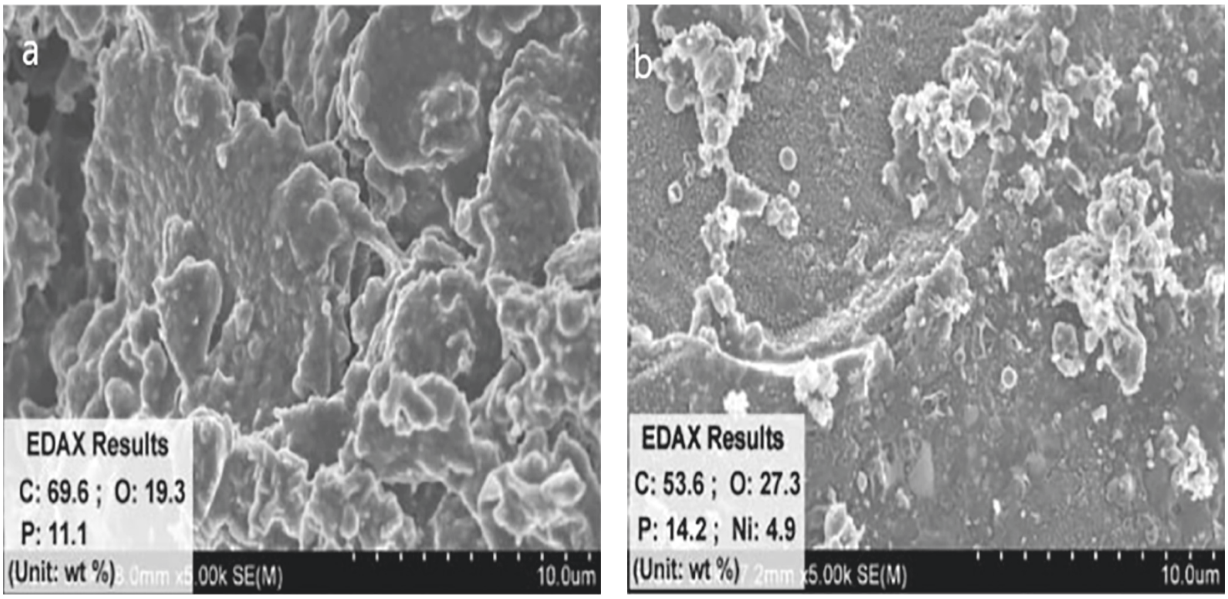
Figure 10: SEM images of lignin-based biocomposites at different temperatures [227] (Copyright @ 2019 MDPI under CC BY 4.0).

6. X-Ray Diffraction (XRD) Analysis of Lignin-Based Composites

The crystallinity of lignin-based composites was measured by using the XRD technique. The crystallinity of the composite was determined by dividing the total amount of sample material by the amount of crystalline cellulose. With the addition of lignin to cellulose, XRD peak intensities were dropped, which presents that the addition of lignin should decrease the crystallinity of cellulose, the cellulose crystallinity of lignin was still over 54% [230]. The addition of alkylated lignin (up to 15%) into PP increases the crystallinity compared to PP [96]. However, the addition of more than 15% alkylated lignin causes agglomeration (based on SEM analysis), so the crystallinity decreases [96]. Meanwhile, the addition of arylated lignin into PP decreased the crystallinity, indicating that arylated lignin was not compatible with PP.

Jiang et al. [187] explored the influence of lignin addition on cellulose crystallinity to fabricate lignin-cellulose biocomposites. After introducing lignin and hot-pressing, the crystallinity of cellulose was preserved. Figure 12(a) shows XRD patterns, which reveal no differences between cellulose sheets and lignin-cellulose composites. The moderate alkali swelling and hot-pressing procedures did not damage the crystalline structure of cellulose, as shown by the diffraction pattern corresponding to the (110), (020), and (040) planes of the cellulose.

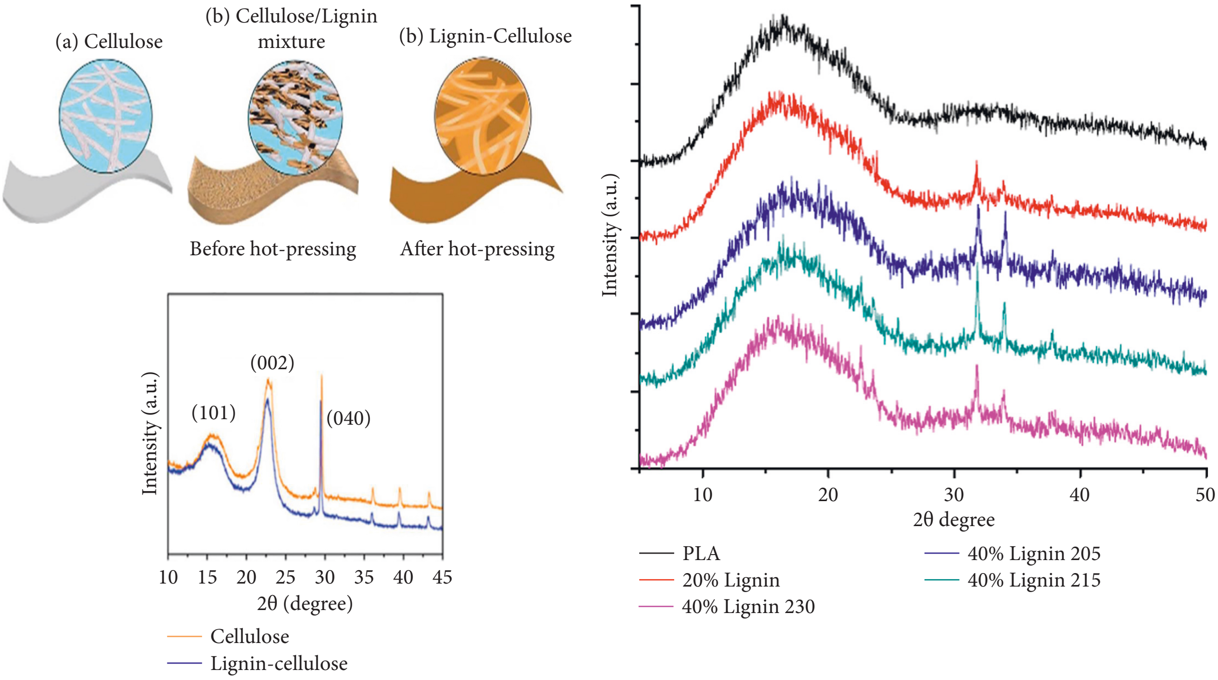
Iglesias Montes et al. [231] reported reported that XRD spectrum of lignin nanomaterials (LNMs) consists of wide peak which revealed that LNMs formed a three-dimensional nanocomposite lacking an organized and consistent



(a)

(b)

Figure 11: SEM images of char residue: (a) polypropylene-based 20% PN lignin composites and (b) polypropylene-based 20 PN-lignin-Ni composites (reprinted with permission from reference [229]; Copyright @ 2012 American Chemical Society).



(a)

(b)

Figure 12: (a) Diagrammatic illustration of the crystal phase of lignin-cellulose composite [187] (Copyright @ 2015 Wiley periodicals) and (b) XRD pattern for lignin-based biocomposite [227] (Copyright @ 2019 MDPI under CC BY 4.0).

supramolecular structures, suggesting that the LNMs were only partly crystalline. Crystallization was not produced by the addition of lignin in the case of PLA monolayers; as no extra peaks formed in the XRD spectrum, also there was no noticeable alteration in the crystalline phase as a result of the addition of lignin and cellulose as binding agents in the XRD patterns of PLA bilayers. Tanase-Opedal et al. [227] reported that when the lignin was added to the matrix, X-ray examination indicated a change in the crystalline phase (Figure 12(b)). PLA has a broad peak due to its microcrystalline structure. XRD Peaks of lignin biomaterials suggested that PLA crystallization had increased due to lignin's role as the nucleation site.

XRD studies of rubber composites revealed the strain-induced crystallization of epoxidized natural rubber (EDR), demonstrating the basic lignin reinforcing effect on EDR (Figure 13). EDR particles might act as additional connectivity cross-links, strain enhancers, and filler materials to a certain level. As a result, when compared with other synthetic rubbers, the clean EDR outperformed them. The diffraction patterns of EDR crystallites steadily decreased with rising lignin concentrations, showing that lignin prevented EDR crystallization caused by strain. There was no strain-induced crystallization in the rubber composites containing a higher lignin concentration. We could conclude from the XRD results that the rubber composites' reinforcing came mostly from lignin instead of particles of EDR [187].

Rosova et al. [9] studied the crystalline structure of biologically active chitosan-based composites with varying quantities of lignin as reinforcement material. Figure 13(b) depicts the wide-angle X-ray diffraction patterns of chitosan and composite materials with various levels of lignin. The existence of peak positions at $2\Theta = 10.5^\circ$ and 15.3° supports the presence of anhydrous type and hydrated chitosan in the sheets [232]. The addition of lignin to chitosan caused the sheets to be amorphized. Furthermore, when comparing composites to the pure chitosan, the peaks at $2\Theta = 10.5^\circ$ were more prominent, whereas the peaks at $2\Theta = 15.3^\circ$ were relatively weak. With the introduction of lignin, the content of the hydrated type increases while the amount of the anhydrous type decreases.

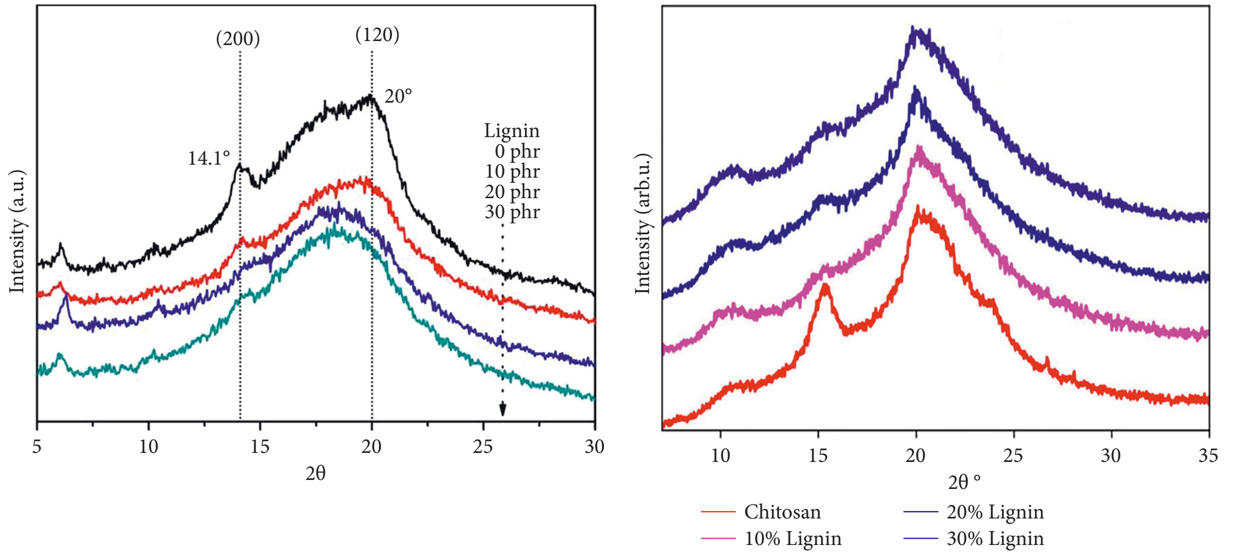
7. Thermal and Mechanical Properties

The phenolic backbone and aromatic structure in the lignin chemical structure have been well known to contribute to its thermal stability [233, 234]. The phenolic backbone itself degrades at around 200 and 450°C; meanwhile, the aromatic structure decomposes between 400 and 600°C [235]. The addition of modified KL into PVA resulted in a slight thermal degradation improvement of the resulting composite film [236]. The increment of the thermal degradation temperature is in line with the lignin content. For example, 5 %wt. lignin contents increase the decomposition temperature ($T_{\text{decomposition}}$) by 2°C, while the addition of 25 %wt. increases the $T_{\text{decomposition}}$ by 15°C. In addition, the presence of lignin in PVA broadens the thermal degradation peak of the composites. In another, Canneti and Bertini introduced lignin to polyethylene terephthalate (PET), and it was observed that lignin strongly influences the composites' thermal stability. However, high lignin loading resulted in a decrease in $T_{\text{decomposition}}$ [237].

The thermal properties of lignin/PLA composites were influenced by the preparation method [238]. According to Gordobil et al. [130], adding KL and acetylated KL (AKL) to PLA greatly boosted its thermal stability but did not support PLA crystallization (Table 3). The incorporation of lignin 0.5 and 5% were increased during the initial degradation (T_{onset}) and $T_{\text{decomposition}}$ up to 70°C. The composite with KL has higher thermal stability than the composite with the acetylated sample. After lignin was added, the amount of char residue was reduced. On the other hand, Sahoo et al. discovered that when lignin was added to the composites, the char residue increased [239].

The incorporation of lignin into polymers appears to have an insignificant effect on the thermal transition temperature, T_g, and the melting temperature (T_m) [240]. The addition of lignin to PLA has displayed no significant change in T_g [130]. A similar study on introducing lignin into PLA reported that the thermal degradation property of composites has not changed significantly with this treatment [241]. Meanwhile, T_g was slightly increased after adding polybutylene succinate (PBS) [216]. Lignin addition has contributed substantially to the T_g and degradation temperature of chitosan [172]. When compared to LS in PLA composites, the addition of DLS improved compatibility and thermal stability (T5% and $T_{\max}$) [135]. In another study, Li and his groups [242] reported that the introduction of lignin increased the decomposition temperature but decreased the mechanical properties of lignin/PLA composites. Thermal properties analysis of alkylated lignin produced a composite that was more stable to heat than arylated lignin because it was more miscible in the PP matrix [96]. The melting point of the PP + alkylated lignin compound was lower than the PP + arylated lignin compound, indicating that alkylated lignin is more compatible with PP, compared to arylated lignin. After the addition of kenaf core fibers and lignin, the maximum degradation temperature of PP composites increased. According to the thermal conductivity analysis, the addition of kenaf core fiber and lignin did not affect the conductivity of the PP composite. In comparison to other PP composites, PP/L/KCF composites had a higher thermal diffusivity and a lower volumetric specific heat [97]. TGA analysis of PP/lignin blend monoliths showed that the addition of lignin caused T_{onset} to shift to a lower temperature, increasing the degradation temperature and the number of char residues, indicating that PP/lignin composites had better heat stability than PP [99].

The modification of lignin and the interaction between lignin and the functional group in the polymer matrix were suggested to be responsible for these variance results [240]. In some reports, the lignin composition in composites influences the viscoelastic properties of the polymer. The cross-linking ability of lignin is accountable for the rise in the stiffness of the composite. In a study, lignin increased the storage modulus of PP/lignin composites with MAPP as the compatibilizer between the two [92]. The loss modulus and storage modulus were increased in line with the lignin content, indicating the successful cross-linking of lignin by polymer molecules. Besides that, the addition of lignin as much as 30% of the total weight decreased the tensile strength and (unnotched Izod) impact strength of PP/lignin composites but increased the flexural strength, Young's modulus, and flexural modulus. The addition of MAPP, as much as 6.67% by weight of lignin, increased the flexural strength and tensile strength [90]. Meanwhile, adding lignin to polystyrene increases its storage



(a)

(b)

FIGURE 13: (a) XRD analysis of lignin-based rubber composite [187] (Copyright @ 2015 Wiley periodicals) and (b) wide-angle X-ray diffraction patterns of chitosan and composite materials with varying levels of lignin [9] (Copyright @ 2021 MDPI under CC BY 4.0).

modulus from 1.24 to 2.25 GPa [225]. Stewart [118] found that lignin provides high impact toughness and stability in epoxy resins when compared to lignin-free phenol epoxy resins. The addition of 10% KL to PP decreased the impact strength (−11.11%), flexural strength (−15.50%), tensile strength (−27.42%), and tensile modulus (−33.63%) of PP/lignin composites compared to PP. The addition of MAPP as much as 3% can improve the impact strength, flexural strength, and tensile modulus of the PP/KL composite. Even the addition of 30% KL along with 3% MAPP was able to increase the tensile strength of PP/Lignin composites (40.37 MPa) compared to PP (36.19 MPa) [96]. After incorporating SL or kenaf core fiber into PP, the composite's tensile strength decreased. Tensile strength of PP composites decreased when lignin or kenaf core fibers were added. Mechanical properties of pure PP and PP/L/KCF composites, on the other hand, were not significantly different [97].

Despite being known for its antioxidant properties and thermal stability; lignin is also famous for its negative effect on the mechanical properties of the corresponding polymer. For instance, lignin and carbon black filler mixture in styrene-butadiene rubber (SBR) matrix has a lower tensile strength than the composite with only carbon black filler [243]. The lignin increases tensile modulus and thermal stability while lowering the tensile strength and strain. Because a high ash content in lignin as a filler can reduce the mechanical properties and thermal stability of lignin/PLA composites, inorganic ash in lignin cannot be overlooked [15]. PLA composites using a combination of LSH and APP have significantly improved thermal stability [136]. The introduction of lignin into cellulose and chitosan polymers upgraded the mechanical characteristics, for example, tensile strength, storage modulus, and water vapor permeability due to some functionalities [244]. It can be affected by the strong interaction and good dispersion of lignin and chitosan [172].

TABLE 3: Thermal degradation properties of PLA and lignin [130].

Samples	Consent (°C)	$T_{\text{decomposition}}$ (°C)	Char residue (%)
PLA	269	314	6.5
PLA + 0.5% KL	340	365	0.5
PLA + 5% KL	337	367	2.1
PLA + 0.5% AKL	328	364	0.2
PLA + 5% AKL	323	366	1.4

The strong molecular interaction, large particle size, and polarity of lignin are considered responsible for its limited use as a filler [4, 245]. Thus, extensive research is being made to improve the compatibility of lignin with the polymer matrix to improve the mechanical properties of the polymer/composite. Some of the efforts are adding a compatibilizer [246, 247], reducing the particle size [248–253], and changing the polarity. Compatibilizer was added to increase the interfacial adhesion between lignin and polymer. In a report, a linear triblock copolymer based on ethylene, styrene, and butylene (SEBS) was introduced to the polystyrene/lignin composite to boost the interaction between polystyrene and lignin [246]. The flexural modulus and tensile modulus were enhanced significantly as evidence of the improvement of the lignin/PS interfacial adhesion. In another study, maleic anhydride graft-PBAT was added to enhance the tensile properties of polybutylene-adipate-co-terephthalate/lignin composite (P/L) [247]. The improvement in tensile strength and elongation at break were noticed in the sample with compatibilizer compared to the P/L composite. However, the tensile properties are still considered lower than the neat polymer.

Sahoo et al. prepared the polybutylene succinate (PBS) and lignin composite using melt mixing [239]. The addition of lignin reduced the elongation at break, increased Young's modulus, and removed the stress softening in PBS. The 30 and 50 %wt. loading of lignin decreased the tensile strength of the composite; meanwhile, the loading of 65 %wt. increased the tensile strength. pMDI was added as a compatibilizer in a composite with 50 wt%. lignin. Only the sample with 1 %wt. of pMDI showed an increase in tensile strength; meanwhile, the addition of 2 %wt. resulted in the opposite. The addition of 10% lignin decreased elongation (43–17%) and increased Young's modulus (583 to 744 MPa) but did not significantly affect tensile strength. The addition of a 0.3% coupling agent in a mixture of PP and lignin improved elongation and had no significant effect on the values of Young's modulus and tensile strength [98]. Toriz et al. [90] added 10–60% lignin in PP and compared it with mica and talc (mineral filler) as fillers in PP. The addition of lignin causes a decrease in tensile strength, flexural strength, and impact strength. However, the addition of mica in the mixture of PP and lignin was able to increase the tensile strength because the distribution and interaction of lignin and PP became better in the presence of mica. The addition of 30% eucalyptus lignin increased the tensile strength and flexural strength because there was an increase in the interaction between PP and eucalyptus fibers [254]. The addition of lignin to PP increased the torsion modulus and flexural while decreasing tensile properties, according to Thakur et al. [4]. Dias et al. [55] stated that the addition of 30% lignin with 3% MAPP increased the flexural strength of PP composites.

The large particle size of lignin is one of the significant challenges to developing lignin as a filler. Various attempts have been made to reduce lignin particle sizes, such as ball milling [248–250], acidification [251], and hybrid filler [243]. Ball milling is considered an environmentally friendly, facile, and low-cost technique compared to other techniques [252]. The ball milling incites the impact energy of the rotating grinding medium to break down solid particles. However, the chances of contamination and lignin with irregular shape and size have resulted in the poor compatibility between lignin and polymer matrix [255]. The reduction of lignin particle size through ball milling was noticed to increase only Young's modulus and reduce other properties such as elongation at break and tensile strength in PP/lignin composite [249]. While ball milling is aimed to break down the particle size physically, acidification is aimed to reduce it chemically. Acidification is conducted during the lignification of black liquor and followed by washing with sulfuric acid and water [249]. Some reports mixed lignin with other types of filler such as carbon black [243], silica [256], ZnO [257], and magnesium oxide (MgO) [258] to enhance the properties of each precursor. Bula et al. investigated the effect of silica/lignin hybrid material as polypropylene filler [256]. Only low concentration (<5 %wt.) of the hybrid filler displayed improvements in elongation at break and notched impact strength. On the other hand, the high loading of the hybrid filler resulted in insignificant improvement in those properties.

Modification of lignin with various methods, including surface grafting [259], hydroxymethylation [260], epoxidation [113], alkylation [96, 261], esterification [134, 262–264], carboxymethylation [265], arylation [96], esterification [134], and sulfonation [43], were used to enhance the compatibility between lignin and polymer matrix. Besides that, the reduction of lignin size in nanoparticles increases 40% of the tensile strength of starch-lignin composites than that of starch composites with higher thermal stability [161]. The lignin modification successfully prevented the reduction of cross-linking rate and thus reinforced the impact and adhesion strength. Meanwhile, when esterified lignin was incorporated in low-density polyethylene (LDPE), the mechanical properties of the composite were similar to the neat polymer [262]. Lee et al. [266] used PP-esterified lignin as a compatibilizer in PLA/coffee grounds composites. The lignin increases stability to heat, while PP increases flexural. The lignin increases the miscibility between PP and PLA, while PP reduces the phase separation between PLA and lignin. In general, the addition of alkylated lignin has a better effect on the mechanical properties of the composite than the addition of arylated lignin. This is indicated by an increase in Young's modulus and elongation, with less decrease in impact strength and tensile strength [96]. Modification of lignin with esterification agents such as butyric anhydride and then blending with PLA resulted in higher Young's modulus and elongation at break than PLA itself [134]. The mechanical properties of lignin/PLA composites have been improved by some esterified lignins [88, 267]. Even though the mechanical properties of neat LDPE did not improve, the lignin's negative effect could be reduced. Despite all efforts to improve lignin's ability to reinforce the mechanical properties of polymers, it appears that more research is needed to improve the lignin compatibility and polymer matrix, such as by adding a compatibilizer.

8. Other Properties

Lignin can function as a free radical scavenger that can prevent oxidation reactions. Stabilizers are compounds that play a role in preventing polymer decomposition due to oxidation reactions with an increase in temperature or light radiation, by suppressing the action of free radicals or preventing the formation of free radicals. Lignin acts as a UV barrier because it blocks UV transmission. The addition of lignin in PP is to accelerate the photooxidation reaction so that PP is more easily decomposed after use. Lignin can act as a stabilizer or initiator of PP degradation, depending on the type and amount of lignin added to PP [268]. Peng et al. [269] investigated the addition of lignin in a mixture of cellulose and PP to produce a composite with a smoother surface, suppressing the decrease in flexural strength and resistance to heat, after weathering treatment. This proves that lignin can act as an antioxidant and stabilizer. Gadioli et al. [254] added eucalyptus lignin in the PP extrusion process, resulting in a composite with mechanical properties and resistance to UV for 200 hours (preventing elongation loss). The addition of 30% eucalyptus lignin increased the tensile strength and flexural strength because there was an increase in the interaction between PP and eucalyptus fibers

Figure 14: Potential application of lignin-based composites.

[254]. The UV transmission of the starch-fractionated SL composite shows promise as an ultraviolet barrier for packaging applications [161]. A similar investigation on the improvement of UV protection of starch-lignin films has been reported previously [162, 165]. Lignin is primarily used as an additive at very low concentrations below 5% because lignin-based materials exhibit high antioxidant and anti-UV properties. These properties were contributed by belonging to the amount of oxygen and the phenolic group as functional groups in lignin [270, 271]. These properties have prospected lignin in wound dressings or anti-inflammatory biomaterial [272].

9. Potential Application Lignin-Based Composites

Lignin has a high potential available biopolymer that can be converted into various bioproducts. The main utilization of lignin is for low-grade fuel by burning that can cause wastage of resources and environmental pollution [273]. Only a limited quantity of lignin has been valorized into biomaterials and renewable chemicals [274, 275]. To promote lignin valorization, some researchers develop advanced technology in this area. Lignin-based composites are biomaterials that utilize lignin for incorporation with typical polymers. As presented in an earlier discussion about the method for lignin-based composite preparation, thus, this section presents some possible applications of lignin-based composites in diverse applications. The properties of the lignin-based composite were influenced by the method for preparation besides polymer constituents. Some functional groups of lignin including methoxyl, hydroxyl, carbonyl, and carboxyl groups [274] become active points in the fabrication process of lignin-based composites. Lignin offers excellent properties such as high carbon content, high thermal stability, biodegradability, antioxidant activity, and rigidity [276] that can be utilized in composites. The potential representative application of lignin in a composite system is depicted in Figure 14, while some effect of introducing lignin in the biocomposites has been summarized in Table 4.

10. Challenges and Future Perspective

Lignin has proven interesting for conversion into renewable biomaterials and chemicals as the second most abundant biopolymer after cellulose. The technique of extracting lignin from biomass has changed the characteristics of lignin. In comparison to OL and SL, KL and LS have a higher ash concentration and lesser purity. The properties of lignin should be examined for acceptable uses, with KL being appropriate for low value-added applications and OL being appropriate for higher value-added applications. In light of environmental concerns, particularly biodegradability issues, lignin becomes more appealing to combine with other polymers in biocomposite systems, including synthetic and natural polymers. The inclusion of lignin into synthetic polymers such as epoxy and PP was covered in this paper, as well as natural polymers such as PVA, PLA, cellulose, starch, NR, and chitosan. Solvent casting and extrusion processes are the two methods utilized to combine lignin in both polymers, with compatibility being the most important factor in the final composite. For better lignin dispersion into polymers, it has been recognized that the differences

Table 4: The lignin introduction and its utilization.

Utilization	Remarks
Particleboard (advanced composite)	The addition of lignin into the particleboard can increase hydrophobicity, reduce thickness swelling and water absorption, and increase the density of particleboard [277].
Rubber (reinforcing agents)	The lignin can improve mechanical properties, tensile strength, and thermal stability in rubber [245, 278].
Filler in adhesives	Lignin is the initiator of cross-linking and condensation reactions, increasing the adhesive's polarity and increasing the penetration of the wood veneer [279].
Anticorrosion	Lignin can form self-cross-linking in the coating process, thermal stability, and hydrophobic properties increase after using silanized fraction lignin [280].
Antimicrobial agents in packaging	Adding lignin to food packaging materials can increase antimicrobial against Gram-positive and Gram-negative bacteria [281].
Carbon fiber or carbon black	Adding lignin to the carbon fiber can increase the spinning performance. High and stable mechanical properties of carbon fiber at the lowest lignin composition [282].
UV blocker	Lignin nanoparticles have higher antioxidants and higher efficiency in protection against *Escherichia coli* and as a good UV protector [283].
Flame retardants	The addition of lignin to the refractory material can improve good thermal stability, increase the residual char, and has excellent coating properties when applied to films [284].
Hydrogel	The addition of lignin into the hydrogel can increase the swelling of the hydrogel. Also, it can be applied as a colorant pollutant remover of soil and seed cultivation [285].
Phenolic resin	Resin from depolymerization alkali lignin phenol-formaldehyde (DAPLF) has higher bonding properties, lower emission, and a faster curing rate [276].
Polyurethane	The addition of lignin into polyurethane as a coating causes cross-linking reactions to increase thermal stability, moderate hydrophobicity in polyurethane coating, and mechanical properties [286].
Lignin-based foam	Incorporating lignin in wood-polyurethane composite foams can increase the density and improve mechanical properties. In addition, it can improve thermal properties as indicated by increasing the mass of charcoal residue, as well as decreasing foam reactivity [287].
Biomedical materials	The application of lignin in wound dressing hydrogels can accelerate wound healing and increase elasticity strength, protein absorption capacity, and the ability to regulate the wound environment in hydrogels [178].

in basic properties between them must be addressed. Lignin can acts as a filler, reinforcing, and nucleating ingredient in biocomposite systems. To improve the compatibility of lignin with polymers, the coupling agent and compatibilizer can be added to lignin-based composites. The low reactivity of lignin, which can be improved by modifying it before adding it to polymers, has limited its usage in combining it with other polymers. The process conditions, amount of lignin, and other chemicals used in biocomposite fabrication remain a problem when it comes to achieving the best composite properties. To broaden the applicability of lignin-based composites in the market, a new optimization strategy is required.

11. Concluding Remarks

To improve biodegradability, lignin can be added to polymeric matrices to form lignin-based composites. However, the compatibility of the biocomposite system is influenced by the differences in characteristics between lignin and conventional polymers. In the preparation of lignin-based composites, solvent casting and extrusion are two popular processes. However, variable levels of lignin, lignin-to-polymer ratios, and operational conditions are influenced by the type of polymer that interacts with lignin, as well as other considerations. To improve biocompatibility between two polymers, additional chemicals such as coupling agents and compatibilizers can be added to the process. The morphological qualities, mechanical-thermal properties, and other aspects of lignin-based biocomposites have all been explored. The influence of lignin on the characteristics of composites varies according to the studies studied. The introduction of lignin contributes to increasing the thermal stability of composites, but it is influenced by the preparation method. The 3D printing, advanced composites, reinforcing filler in natural rubber, filler in adhesives, anticorrosion, antimicrobial agent in packaging and textile, flame retardant, carbon fiber, UV blocker, biomedical materials, polyurethane, lignin-based foam, and phenolic resin are just a few of the applications for lignin-based composites. As a result, the preparation technique, which includes the amount of lignin or other additives in the lignin-based system, is still being developed to improve the performance of lignin-based biocomposites for a variety of applications.

Data Availability

The data presented in this study are available on request from the corresponding authors.

Consent

Not applicable.

Disclosure

This study is part of Muhammad Rasyidur Ridho's master's thesis at IPB University.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

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Research Article

Investigation on Forestry Wood Wastes: Pyrolysis and Thermal Characteristics of Ficus religiosa for Energy Recovery System

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Pyrolysis is the most important thermochemical process that can be used for the production of biofuel, from wood and wood-based lignocellulosic materials. In this study, bio-oil is produced from the bio-weed named *Ficus religiosa* by the thermal pyrolysis process by utilizing laboratory-scale fluidized bed reactor. This study deals with the production of maximum bio-oil by optimizing process parameters such as process temperature, particle size, and sweep gas flow rate. Further different analytical techniques were used to describe the properties of bio-oil for different applications. Wood and wood barks of *Ficus religiosa* were chosen as the raw material due to their higher volatile content (72.4%). The maximum yield of 47.5 wt% bio-oil was collected at the optimized operating conditions of 450°C temperature, 1.0 mm particle size, and 2.0 m^3/h sweep gas flow rate. Compared with other operating parameters, temperature is observed as the most significant one to determine the product yield. Through chromatographic analysis, it was identified that the bio-oil is found with the variety of chemical compounds including alcohols, alkenes, phenols, saturated fatty acids, and esters.

1. Introduction

The promotion of sustainable energy development has been aided by increased global energy demand with increased greenhouse gas emissions [1]. Wood and biomass materials are taken as promising alternative material for producing fuels and chemicals [2]. These sources are renewable, available abundant, carbon-neutral, and does not compete with food [3]. Since the mid-1970s, when the oil crisis began, numerous efforts were made to convert wastes into fuels and chemicals. To meet country's energy needs, United States, China, India, and other countries are significantly dependent on foreign countries, creating tremendous incentives for the development of renewable energy sources. In 2020, India contributed 4.6% to the global fossil fuel use, placing it third in the world after United States and China. In accordance with the findings of Huber, biomass-derived biofuels are the viable resource for liquid hydrocarbon production for transportation [4], but the economically viable method of producing them has not yet been identified.

Wood and wood-based lignocellulosic materials are the most important sources of renewable energy. It is the feedstock obtained from dedicated crops, forest wastes, and a collection of municipal solid wastes. The energy is recovered directly from wood in the form of either electrical or thermal through combustion and other thermal processes [5]. The management of wastes obtained from wood is critical to raise the sustainability requirements. The wood-based biomass generally consists of wood waste, tree bark, tree trunk, and leaves, and it contains energy obtained by photosynthesis. These wastes can be burned to generate energy or can be processed to produce fuel [6]. The energy from the direct burning of wood and wood products is mostly used for many heating applications, but pyrolysis and gasification allow them to produce more valuable liquid and gaseous combustibles that can be used for other processes. In particular, the pyrolysis has emerged as an effective conversion process of wood-based biomass materials into liquid fuels. When compared to other conversion techniques, pyrolysis yields more liquid products with increased energy contents. It is a thermal breakdown process that produces char, oil, and gaseous compounds due to the breakdown of polymeric chain components without air/oxygen [7]. Pyrolysis oil is generally having higher energy density and is easier to store and transport than conventional diesel oil. Fast or flash pyrolysis is the advanced technique that enhances more liquid oil production than slow pyrolysis [8]. Several articles have been published previously utilizing many biomass materials for the pyrolysis process. Makkawi et al. [9] performed pyrolysis of palm wastes at 525°C and obtained bio-oil production of 38.8 wt%. The bio-oil has calorific value of 20.88 MJ/kg, which is significantly lower than the heating value of softwood bark pyrolysis oil. In another study, Pinto et al. [10] utilized pinewood bark for the pyrolysis process and produced 37 wt% of bio-oil at the optimum temperature of 550°C. Recently, Sakthivel et al. [11] utilized wood wastes of *Calophyllum inophyllum* for pyrolysis. The study utilized various chromatographic techniques to analyse the obtained oil products. The FTIR study on bio-oil showed the presence of C=C, –CH, C=O, and –OH functional groups. Dhanalakshmi and Madhu [12] carried out the research on flash pyrolysis of wood bark of *Azadirachta indica*. The study yielded a maximum of 49.5 wt % pyrolysis oil at 450°C under 1 mm particle size and at 2 m^3/hr N_2 flow rate. Fast pyrolysis of pine and oak wood barks was performed by Ingram et al. [13] at 450°C. The results of this study suggested that the portable auger reactors can be utilized for biofuel production in various sites in forests. This method also allows more efficient transportation of less bulky bio-oil compared with raw wood. In addition to the above studies, some of the studies utilized wood bark for co-pyrolysis experiments. Co-pyrolysis experiments on mixed food wastes with wood bark were conducted by Park et al. [14] in a continuous pyrolysis reactor. The study characterized three pyrolysis products of oil, char, and gas. When food waste and wood bark were combined at a mass ratio of 1 : 1 at 700°C, the maximum yield of H_2 gas and phenolic and polycyclic aromatic hydrocarbon compounds were obtained.

In the above background, the wood and wood bark of *Ficus religiosa* are utilized for the effective conversion of bio-oil products. *Ficus religiosa* is a type of fig tree origin in the Indian subcontinent. It is a huge, dry-season deciduous tree that can grow up to 30 metres in height having a trunk diameter of up to 3 metres. It is indigenous to most of the Indian subcontinent, including Bangladesh, Nepal, Pakistan, and Assam. In India, this tree is marked with religious and medicinal symbols. It is possible to utilize different parts of trees to treat different ailments such as diarrhoea, jaundice, heart-related diseases, and skin allergies. The diverse parts such as stem, bark, fruits, and resins are used in many different ways. The presence of numerous phytoconstituents such as phenolics, sterols, and flavonoids also has been examined for the past ten years [15]. The wood of the tree is used for carpentry work, and their barks are used for cooking purpose. To the best of the author's knowledge, the publications available related to the utilization of wood and wood barks for biofuel production are very minimum and there was no study that has been discussed the use of wood and wood barks of *Ficus religiosa* for energy recovery through pyrolysis process.

Motivated by waste to energy concept and conversion of wood-based materials for value-added fuels and chemicals, this study focused on pyrolysis of mixed wood and wood barks of *Ficus religiosa*. The ultimate goal of this study was to investigate the pyrolysis behaviour of the feedstock under various operating conditions. To obtain the maximum bio-oil products, flash pyrolysis studies are carried out in a laboratory-sized fluidized bed reactor at various temperatures, particle sizes, and sweep gas flow rates. In the end, the complete characterization of the bio-oil products has been carried out using FTIR and GC-MS.

2. Materials and Methods

2.1. Material Collection and Preparation. The materials used for this study are the collection of wood and wood barks collected from newly cut trunks. The trunks were cut from the tree available in the residential area near Vadavalli, Coimbatore, India (11°02′10.5″N 76°54′50.4″E). The materials were cut into small pieces and dried in an open sunlight for 15 days. After drying, the wood pieces are ball-milled and converted into powder form using a ball mill (Emkad–India made). The samples are then sieved to get desired sizes of 0.5, 0.75, 1.0, and 1.25 mm using Test Sieves made by Jayant Scientific Industries, Mumbai, India. To prevent biological degradation, the powdered materials were oven-dried at 60°C for 1 hr using BSSCO BSEX-1401, B.S. Exports, India. Figure 1 shows *Ficus religiosa* tree.

2.2. Characterization Study

2.2.1. Proximate and Ultimate Analysis. The proximate analysis of the feed materials was carried out by the following ASTM protocols. This analysis gives the information about the amount of total volatiles present in the feedstock

Figure 1: *Ficus religiosa* tree.

materials. The elemental analysis of the feed materials and bio-oil is found by utilizing Perkin-Elmer II 2400 CHNS elemental analyzer by the following ASTM D5373 procedure.

2.2.2. Thermogravimetric Analysis. The thermogravimetric study is used to identify the fundamental behaviour of the feed material during pyrolysis. The analysis was carried out using thermogravimetric analyzer (Q500, TA Instrument). About 5 mg of biomass materials was kept in the furnace. The nitrogen gas and air required for the pyrolysis and combustion experiments are provided during the analysis. For analysis, the feedstock was heated to 700°C at the heating rate of 20 °C/min.

2.2.3. FTIR Analysis. A Bruker Optik GmbH Tensor 27 spectrophotometer was employed to get FTIR spectrum of the oil sample. The spectrum was collected between 400 and 4000 cm^{-1} with 4 cm^{-1} resolution. The spectrum for bio-oil was obtained in the reflectance mode after the sample was dispersed on zinc selenide crystal.

2.2.4. GC-MS Analysis. The bio-oil produced under optimized conditions was examined qualitatively using Thermo GC-Trace Ultra Version: 5.0, Thermo MS DSQ II, GC-MS spectroscopy. The obtained results in this study provided a clear image of the chemical compounds present in the sample.

2.2.5. Physical Characterization. To find the suitability for various applications, the physical qualities of the bio-oil were conducted. To determine the suitability as fuel for IC engine applications, the physical qualities of the oil must be considered. Density, kinematic viscosity, flash point, heating value, and pH value are all analysed in accordance with ASTM standards. Prior to the analysis of the oil, the samples are centrifuged at 3000 rpm to separate the organic phase from the aqueous phase.

2.3. Reactor Setup. The flash pyrolysis experiments are conducted using laboratory-scale fluidized bed reactor of height of 1.0 metre and diameter of 50 mm. The outer surface of the reactor is heated electrically, and the heat input is regulated by the PID controller. Five K-type thermocouples are fixed at different locations to measure the temperature. Nitrogen gas is supplied to the reactor at different flow rates in order to get fluidization along with the sand particles. The temperature of the reactor is raised at 10 °C/min till it reaches the desired value. The reactor can be allowed to heat the samples up to 1100°C. A cyclone separator is fitted at the outlet of the reactor to separate expelled char components from the gas products. A condenser is used to condense volatile fractions released during pyrolysis, and it is collected in a separate glass beaker. The emitted non-condensable gases are allowed to escape into the environment. After cooling, the char elements were removed from the reactor and from the cyclone separator. The reactor facilities available in fuel laboratory are utilized for this study. The reactor was the existing one, and it was not specifically designed for this work. The reactor was supplied by Emkad India Ltd. A high accuracy weighing machine is employed to compute the mass of pyrolysis products. The mass of the uncondensable gas products is identified using the remaining material balance. The overall setup used for this study is shown in Figure 2.

3. Results and Discussions

3.1. Feedstock Properties. Table 1 summarises the important elements of the wood and wood bark of *Ficus religiosa.* The studied material is having higher carbon contents (C), moderate oxygen (O) contents, and higher hydrogen (H) contents. The materials are having low nitrogen (N) and sulphur (S) contents. The values of C, H, N, S, and O of the material are 48.1, 6.3, 2.0, 0.4, and 43.2 wt%, respectively. In addition to that, ash in the feed material is quite minimum (5.2 wt%). The presence of oxygen in the material leads to the production of oxygenated volatiles and acidity. The amount of moisture fractions in the feed materials can reduce the conversion efficiency during pyrolysis and also lowers the heating value of the yield products with the aqueous phase. The higher concentration of volatile matter results in increased volatility and reactivity, which is ideal for the synthesis of large volumes of bio-oil [16].

3.2. Thermal Degradation Analysis. Figures 3 and 4 depict the TGA and DTG plot of the *Ficus religiosa* obtained by heating the samples to 700°C at 20°C/min. As identified from TG curves, the thermal breakdown of the biomass samples appears in gradual manner with clear path. At the temperature of around 260°C, the first peak appeared corresponding to the decomposition of hemicellulose. In this stage, the hemicellulose decomposes into volatiles and an intermediate solid further in the secondary reaction, and the intermediate solid particles are decomposed into condensable volatiles and char products [17]. The second peak started to appear at 350°C, which represents the breakdown of cellulose. Because of the overlap between the other two peaks, the peak associated with the decomposition of lignin

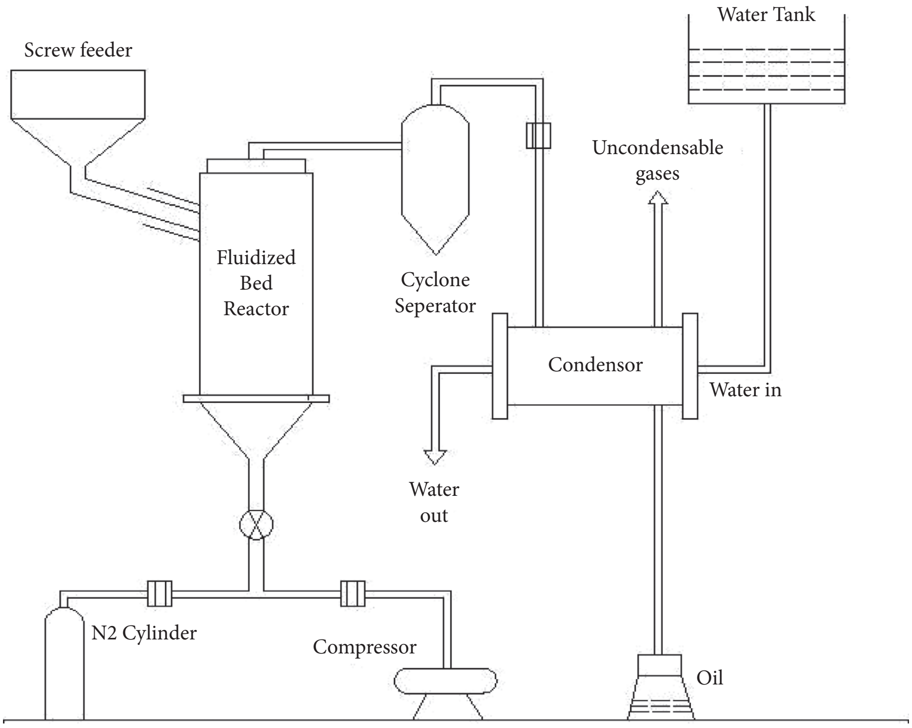

Figure 2: Reactor setup.

Table 1: Properties of feedstock in wt%.

Content	Values
Volatile matter	72.4
Fixed carbon	16.3
Moisture content	6.1
Ash content	5.2
C	48.1
H	6.3
N	2.0
S	0.4
O	43.2
Empirical formula	$CH_{1.56}N_{0.035}O_{0.674}$
Heating value in MJ/kg	17.04

% of O = 100 − (C + H + N + S)%.

is entirely obscured. The decomposition of lignin in the biomass into condensable volatiles and char elements is accomplished by an Arrhenius-type process [18]. It is around 420°C when the thermal breakdown of the entire biomass is practically completed. At elevated temperature, the breakdown of lignin components shows minor weight loss. The utmost weight loss appeared between 370°C and 470°C. The TGA curve shows the char yield of approximately 20 wt%. These results are similar to the previously published literature for rice hulls, olive stones, and sugarcane bagasse [19–21].

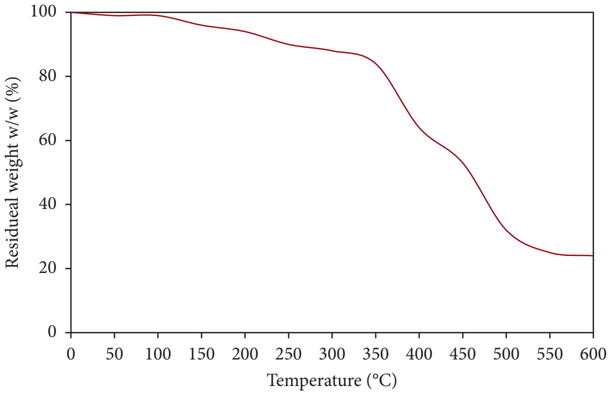

Figure 3: TG analysis of *Ficus religiosa*.

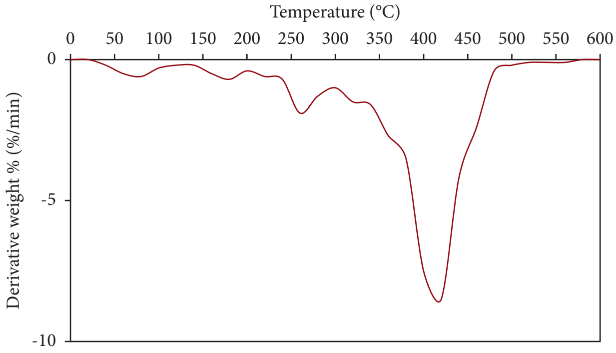

Figure 4: DTG analysis of *Ficus religiosa*.

3.3. Pyrolysis Characteristics

3.3.1. Effect of Temperature. A series of chemical reactions will take place in biomass during pyrolysis. To get primary and intermediate products, it is necessary to decompose the lignocellulosic contents. The heat is supplied to the reactor primarily to break down these constituents. The yields of pyrolysis product distributions at different final temperatures

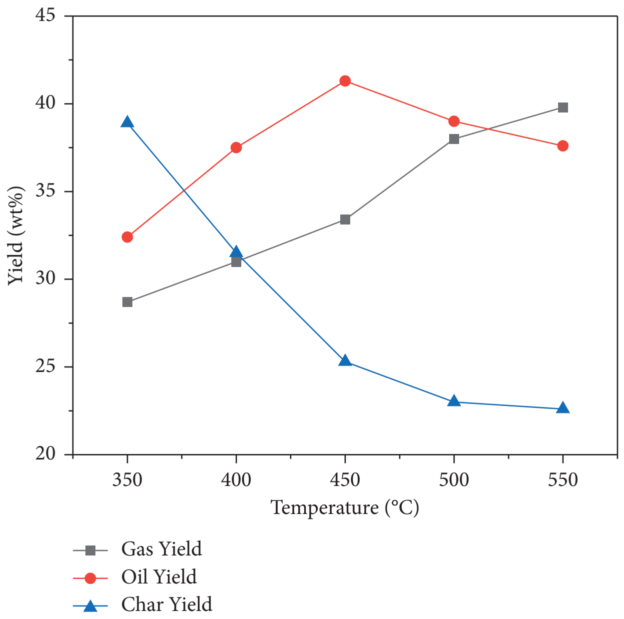

Figure 5: Pyrolysis yield under different temperatures.

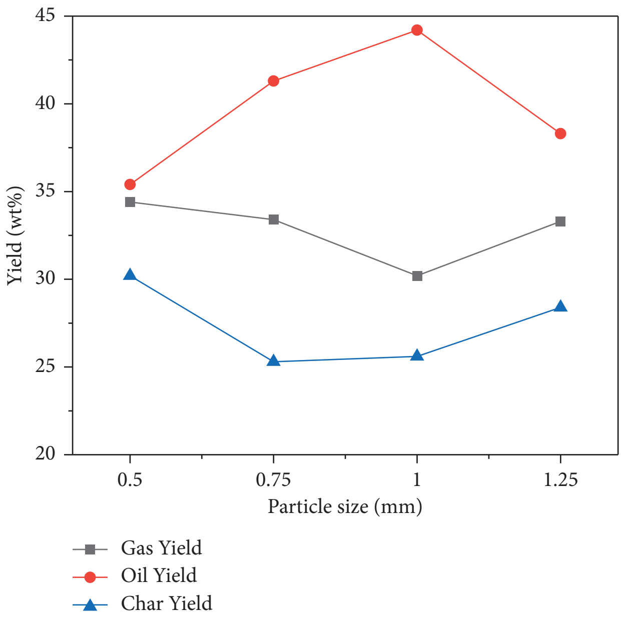

Figure 6: Pyrolysis yield under different particle sizes.

are shown in Figure 5. The experiment in this phase was conducted by changing reactor temperature from 350°C to 550°C under 0.75 mm particle size and 1.75 m^3/hr N_2 flow rate. Increasing heat input facilitates the breaking of heavy hydrocarbon materials resulting in higher liquid and gaseous products, which negatively affect char production. The yield of char products decreased with increased temperature, because of the primary or secondary breakdown of char particles in the reactor [22]. The char yield decreased from 38.9 to 22.6 wt% with increasing temperature from 350°C to 550°C. According to Pütün et al. [23], increasing the reactor temperature from 400 to 700°C reduced char generation by 10% and 17% for hazelnut shells and sesame stalks, respectively. Choi et al. have also showed a drop in the char products with increased temperature [24]. From the figure, it can be understood that higher char products are achieved at low temperatures. When the amount of heat input provided to the feedstock is increased, it breaks the bonds and enhances the release of condensable volatiles. It is also identified that the yield of pyrolysis oil is gradually enhanced from 32.4 wt% to 37.6 wt% and attained maximum at 450°C (41.6 wt%). When the temperatures go beyond 450°C, the production of pyrolysis oil is decreased in the range of 41.3 wt% to 37.6 wt%, which is consistent [25, 26]. The higher carbon conversion into gas at elevated temperature increases the yield of gas continuously from 350°C to 550°C. The yield of gas is 28.7 wt% at 350°C, and it reached 39.8 wt% at 550°C.

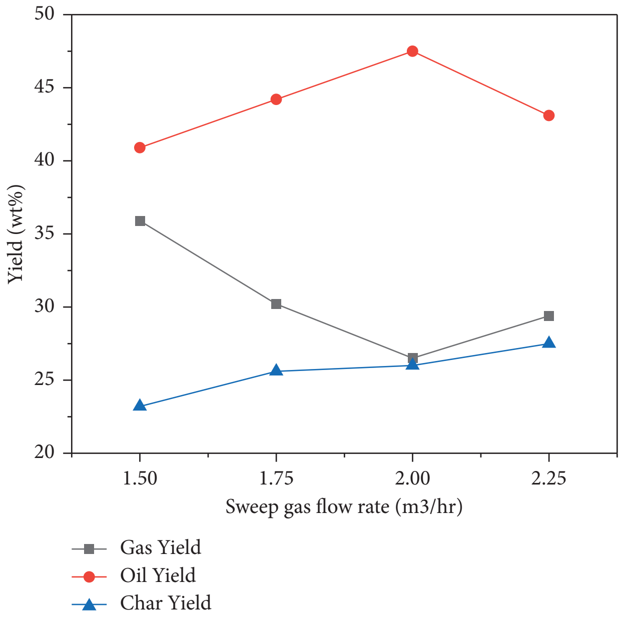

Figure 7: Pyrolysis yield under different N_2 flow rates.

3.3.2. Effect of Particle Size. The size of the particle is an important one that should be considered carefully since it might influence the rate of heat transfer. It significantly creates an impact on chemical reaction during the process. The particle size not only affects the pyrolysis reaction, but also affects the pressure drop in the reactor. When the diameter of the particles increases, the distance between the surfaces to its core rises, resulting in poor heat transfer [27]. The increased diameter is generally favoured for the yield of char products. In many previous studies, the yield of products was significantly affected by the size of the particles [28, 29]. The biomass particle size is a very critical one that affects material flow properties. Sowmya Dhanalakshmi and Madhu [8] reported that the production of bio-oil was influenced by the particle size while using neem wood bark. Figure 6 depicts the yield products with respect to particle sizes. From the figure, it is observed from the results that the quantity of oil collection reached a maximum of 44.2 wt% at

Table 2: Properties of the bio-oil.

Properties	Standard	Feedstock materials				
		Ficus religiosa	Pine chips [35]	Lemongrass [36]	Hardwood [37]	Diesel [38]
Density (kg/m^3)	ASTM D445	1005	1200	1010	1220	850
Viscosity (cSt)	ASTM D4052	7.2	28	7.2	13	3.9
pH	ASTM D7946	3.6	2.4	4.1	#	#
Flash point (°C)	ASTM D92	130	95	150	66	57
Elemental compositions (wt%)						
C	ASTM D5373	44.23	45.7	52.14	47.54	86.5
H		8.4	7.0	7.2	7.8	13.2
N		0.81	0.1	0.61	0.5	0.02
S		0.22	0.02	0.1	0.1	0.24
O	By difference	46.34	47.00	39.95	44.06	—
Heating value (MJ/kg)	ASTM D445	18.3	17.2	19.40	18.63	43.6

#—not reported.

1.0 mm particle size. On the other side, the oil yields were reduced to 38.3 wt% at larger particle size. This could be explained by an incomplete process due to the lack of heat transfer [26]. The larger particles do not get properly heated up and end with lower oil yield. According to Figure 6, the char yield is very minimum at 0.75 mm (25.3 wt%). The yield of gas is also a function of particle size. Here, the higher and lower gas products were recorded at the particle size of 0.5 mm (34.4 wt%) and 1.0 mm (30.2 wt%). When the particle size is increased from 1.0 mm to 1.25 mm, the yield of gas is enhanced from 30.2 wt% to 33.3 wt%. The increased gas yield is due to increased particle size in the reactor, which prolongs the residence time of volatile matter leading to secondary cracking reactions.

3.3.3. Effect of Sweeping Gas Flow Rate. Another parameter that affects the product yields is the flow of the sweeping gas. In the fluidized bed pyrolysis of *Ficus religiosa* , a higher amount of vapours is produced, and it should be expelled properly from the reactor or else they may be involved in further reactions, which change the nature of the products. In accordance with the literature, the allowed sweep gas inside the reactor separated the products from the reactor to avoid the secondary reactions and aiding maximize the oil products. Nitrogen is the commonly used carrier gas for pyrolysis experiments due to its better fluidization, inert property, cheap, and availability. It also helps to drain out the reactor towards oxygen free reaction [30]. In this phase, the experiments were conducted at constant temperature of 450°C and at the fixed particle size of 1.0 mm. Figure 7 shows the experimental yields at different N_2 flow rates. At 2.0 m^3/hr, oil product yields reached their maximum value (47.5 wt %). From the results, it can be noted that the N_2 flow significantly creates the impact on residence time of the vapours created during the reaction and prevents secondary reactions [31]. At a higher N_2 flow rate of 2.25 m^3/hr, the gas yield is increased, due to the removal of uncondensed volatiles. In pyrolysis of *Ficus religiosa*, the sweeping gas has not affected much the production of char. The yield of char is 23.2 wt% at 1.5 m^3/hr and 27.5 wt% at 2.25 m^3/hr. Previously, pyrolysis of corncob showed a minor change in char yield from 24.4 to 22.6 wt% when N_2 flow rate was increased from 1.2 to 4.5 L/min [32]. During the Laurel pyrolysis, increasing the N_2 flow rate from 50 to 400 mL/min resulted in a drop in char production from 28.48 wt% to 27.21 wt% [33].

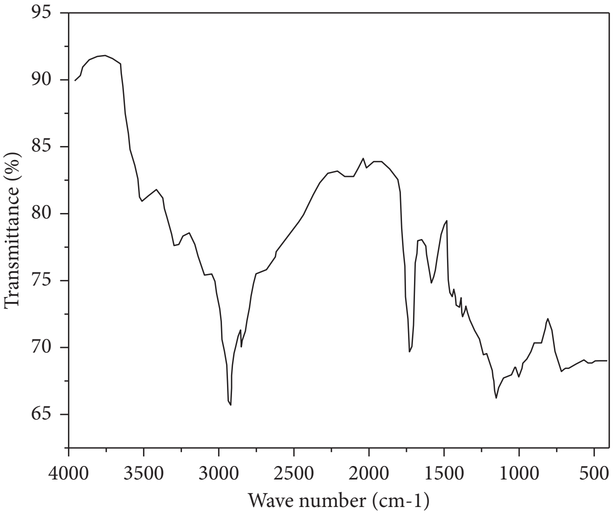

Figure 8: FTIR analysis.

3.4. Characterization Study

3.4.1. Physical Analysis of the Bio-Oil. The basic physical analysis is a very important one since it can be used for any applications based on its physical and chemical properties. The analyses in this study are made on the bio-oil produced at optimum conditions of pyrolysis temperature of 450°C, particle size of 1.0 mm, and sweep gas flow rate of 2.0 m^3/hr. Viscosity, heating value, density, oxygen content, pH value, and density are some of the physical qualities measured in this study and reported in Table 2. The table also compares other pyrolysis oil obtained from different sources. Viscosity is a crucial property of any liquid fuel that determines fluidity. It plays a major role in the design of engines that utilize biofuels [34]. The viscosity of pyrolysis oil varies due to the fact that it is made from a variety of sources.

Table 3: GC-MS analysis of oil obtained at optimized conditions.

RT/Min	Compound name	Molecular name	% area
3.34	Piperidine-2,5-dione	$C_5H_7NO_2$	4.41
4.26	4-Ethyl-2 methoxyphenol	$C_9H_{12}O_2$	1.54
5.42	5-(Benzyloxymethyl)uracil	$C_{12}H_{12}N_2O_3$	0.94
6.31	2-Furanmethanol	$C_5H_6O_2$	0.34
6.96	Phenol, 3,4-dimethyl-	$C_8H_{10}O$	6.58
9.01	Phenol, 2-methoxy-	$C_7H_8O_2$	8.62
12.22	Phenol	C_6H_6O	9.54
13.04	2-Methoxyphenol	$C_7H_8O_2$	3.41
13.56	5-Hydroxymethylfurfural	$C_6H_6O_3$	0.28
14.02	Hexadecanenitrile	$C_{16}H_{31}N$	5.45
14.54	Oleic acid	$C_{18}H_{34}O_2$	0.94
14.69	Pyrrolidine, 1-(1-cyclopenten-1-yl)-	$C_9H_{15}N$	4.44
15.73	2H-Pyran, 2-(2 heptadecynyloxy)tetrahydro-	$C_{22}H_{40}O_2$	3.72
17.43	4,6-Dihydrofuro[3,4-b]furan	$C_6H_6O_2$	1.66
19.77	2-Cyclopenten-1-one	$C_7H_{10}O$	2.60
20.08	1,4-Dimethoxy benzene	$C_8H_{10}O_2$	0.88
21.41	Benzenemethanol, 4-hydroxy	$C_7H_8O_2$	2.04
22.57	Kaempferol	$C_8H_8O_4$	3.44
22.98	9-Octadecenamide	$C_{18}H_{35}NO$	5.32
23.58	4-Methyl-5H-furan-2-one	$C_5H_6O_2$	1.42
23.74	2-Isopropyl-2,5-dihydrofuran	$C_7H_{12}O$	5.20
23.99	Vanillin	$C_8H_8O_3$	3.71
24.09	1H-Imidazole, 1-methyl-4-nitro	$C_4H_5N_3O_2$	0.30
25.16	9-Octadecenoic acid (Z)-,methyl ester	$C_{19}H_{36}O_2$	8.03
27.64	4-Ethyl-2-methoxy phenol	$C_9H_{12}O_2$	1.86
28.43	2,3-Butadiene	$C_4H_6O_2$	0.45
29.10	1,2-Benzendiol	C6H6O	2.55
29.57	Cyclopentanol	$C_5H_{10}O$	3.04
30.42	1-Propyne, 3-iodo-	C_3H_3I	3.23
32.81	Benzhydryl vinyl ether	$C_{15}H_{14}O$	1.59
36.42	1-Methyl-1,3,3-triphenylindan-2-one	$C_{28}H_{22}O$	1.04
37.42	Stigmasterol	$C_{29}H_{48}O$	0.76

Obviously, the viscosity is identified as 7.2 cSt. The higher flash point of 130°C shows that the oil can be stored safely at room temperature. Bio-oil has a density of 1005 kg/m^3, which is denser than mineral diesel (78 kg/m^3). The pH value is shown as low as 3.6 due to acidic chemicals in the sample. When compared to mineral diesel, the heating value is low as 18.3 MJ/kg, which is more enough for the bio-oil obtained from lignocellulosic wastes and it can be improved by further processing.

3.4.2. FTIR Analysis. Figure 8 shows the FTIR image of bio-oil derived from wood and wood bark of *Ficus religiosa* obtained at optimized maximum yield conditions. The existence of phenolic group is identified by a large stretch at 3500–3200 cm^{-1}. The O-H stretch at 3400–3100 cm^{-1} shows the occurrence of polymeric hydroxyl compound. The C-H stretch obtained at 3000–2850 cm^{-1} represents the alkane compounds in the oil. The C=O stretch found at 1750–1700 cm^{-1} demonstrates the presence of a stable ester group. The N-H bend at 1550–1500 cm^{-1} confirmed amides in the sample. The alcohols and aromatic compounds in bio-oil are represented by stretching vibrations at 1152 cm^{-1} and C-H bending vibration at 710–690 cm^{-1}. The result obtained in this study is consistent with the previous literature [8, 11]. By comparing with previous studies, the oil contains abundant O-containing structures. The functional group of the oil is basically dependent on the elemental compositions of the feedstock materials. The figure also indicates that the oil is complex and has some aromatic compounds, for example, phenols. However, the exact assignment of each band appears to be tough and gives the provision for further study [39].

3.4.3. GC-MS Analysis. This analysis was done to identify the presence of numerous chemical elements in the oil sample. The sample for this analysis was obtained at optimized maximum yield conditions. The compounds that appeared in this analysis are listed in Table 3. The compounds were significantly identified related to peak area. The analysis revealed the chemical components including alcohols, alkenes, phenols, saturated fatty acids, and esters. In chromatogram, phenolic compounds are observed more than 30%. The degradation of lignin in the wood may be responsible for the increased phenolic content. At the same time, 9-octadecenoic acid (Z)-, methyl ester, hexadecanenitrile, and 9-octadecenamide showed an area percentage of 8.03, 5.45, and 5.32%, respectively. Piperidine-2,5-dione, kaempferol, and stigmasterol identified in the samples are used for medicinal applications, whereas phenol and its derivatives, hexadecanenitrile, oleic acid, vanillin, cyclopentanol, and 2-furanmethanol are used for various chemical processing industries. Apart from that many other chemicals identified in the bio-oil are used as oxidizing agents, reducing agents, food additives, precursors, plasticizer, and perfume additives. Furthermore, several studies also reported the presence of these chemical compounds in various pyrolysis oil [40–42].

4. Conclusion

This study confirms that combination of wood and wood barks of *Ficus religiosa* is valuable raw materials for pyrolysis-based bio-oil production. The most favourable operating conditions for getting higher yield of bio-oil are 450°C temperature, 1.0 mm particle size, and 2.0 m^3/hr sweeping gas flow rate. The yields of liquid oil were in the ranges from 32.4 wt% to 47.5 wt% under these favourable conditions. This study offers a successful method for extracting energy-rich biofuels from *Ficus religiosa* wood. The bio-oil was examined and compared with other pyrolysis oil produced from different wood-based lignocellulosic wastes. The comparison results confirmed that the oil products can be utilized as fuel and have the potential to be employed as a renewable fuel. According to the chemical characterization study, the obtained bio-oils are complex in nature containing a wide range of organic components indicating that they can be used for chemical industries. Further study is also possible to increase the production of char and gas elements by optimizing process parameters with deep product characterization.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

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Review Article

Engineering Wood Products from *Eucalyptus* spp.

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Forest covers 4.06 billion hectares (ha) or 31% of the total land area worldwide, where 93% (3.75 billion ha) are natural regenerating forests and the remaining 7% (294 million ha) are planted forests. *Eucalyptus* spp., being one of the most important plantation species, has been planted in 95 countries around the world, and the area of plantation has exceeded 22.57 million ha. In the southern hemisphere, it is a significant industrial fast-growing tree species. These plantations serve as a valuable resource for the timber and fibre-based industries. *Eucalyptus* is the main fibre resource for the pulp and paper industries in developed countries. Timber extracted from the planted eucalyptus trees has long been used for solid wood and its fibres were used for manufacturing medium-density fibreboard. In comparison to most softwood species, *Eucalyptus* timber is reported to have a higher rigidity, making it ideal for manufacturing structural products. Therefore, this paper presents a review and analysis of the recent state of research on the utilisation of planted eucalyptus for engineered wood products (EWPs) manufacturing. This study investigated *Eucalyptus*-based EWPs such as particleboard, fibreboard, oriented strand board, laminated veneer lumber, plywood, glue laminated lumber, and cross-laminated lumber. The feasibility of using planted *Eucalyptus* in the production of EWPs, as well as the challenges encountered, was also discussed.

1. Introduction

According to the Global Forest Resources Assessment (FRA) report published by the Food and Agriculture Organization of the United Nations (FAO) in 2020, the total forests area worldwide is amounted to 4.06 billion hectares (ha), which covers 31% of the total land area [1]. Two broad categories of forests have been identified by FRA, namely, naturally regenerating forests and planted forests. Natural regenerating forests cover around 3.75 billion ha or 93% of the total forest area. Meanwhile, the total area of planted forests globally is estimated to be 294 million ha or 7% of the world forest area. Asia has the largest area of planted forests which amounted to 135.23 million ha, or 46% of the total planted forests area globally, followed by Europe, North and Central America, South America, Africa, and Oceania. Figure 1 displays the increment of the planted forest area in all regions between 1990 and 2020. As of 2020, the total planted forest area was significantly increased by 72% compared to 1990.

Planted forest typically refers to the forest that is primarily made up of trees that have been planted and/or intentionally seeding. Planted forests provide many benefits including traditional timber and fibre production, economic development, and employment in rural areas [2] and have been identified as a key means to fight climate change in the short to medium term, restore degraded land, and maintain sustainable ecosystem functions and services [3–6]. In the context of a broader geographic and economic context, well-managed planted forests can contribute to sustainable development [7]. Planted forests are now also being proposed as a way to reduce harvesting pressure on natural forests [8].

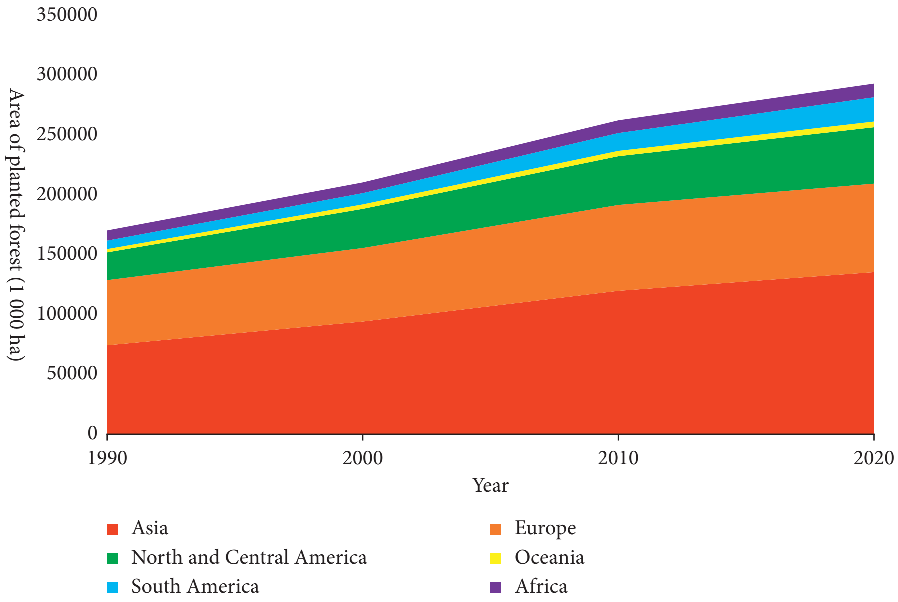

Figure 1: Planted forest area by region for the period 1990–2020 [1].

FRA divided the planted forests into 2 categories: plantation forests and other planted forests. Based on the definitions, plantation forests are intensively managed for productive purposes, with one or two species, even age class, and consistent spacing. Plantation forests are grown for the purpose of producing timber, fibre, energy, and nonwood forest products. A subtype of plantation forests is those primarily made up of introduced species. On the other hand, other planted forests consist of one or more tree species and are less intensively managed, typically for multiple purposes and do not meet the criteria of plantation forests and may even resemble natural forests at stand maturity. The areas of plantation forests and other planted forests by region and subregion are shown in Figure 2. Plantation forests cover 131 million ha worldwide, accounting for 45% of all planted forest land. The remaining 55% is classified as other planted forests, which cover 163 million ha. East Asia has the highest share of plantation forests while other planted forests predominate in Europe [1].

Of all tree species being planted worldwide, *Pinus* species (native and nonnative) are dominant in most regions in the world, while nonnative *Eucalyptus* species are the most common in the tropics and subtropics [9]. *Eucalyptus* is typically managed on short rotation to enhance economy with the production of timber, pulpwood, charcoal, and firewood [10]. *Eucalyptus* is very adaptable, tolerating low soil fertility, acidic soils, and soils rich in aluminium, often periodic moisture stress, diverse climates and soil types, and even fire and insect damages and low water availability [11–15]. Other favourable characteristics of *Eucalyptus* include its good efficiency at capturing CO_2 and producing oxygen, better efficiency in water consumption compared to other species, increasing soil fertility, and restoring land degradation or unproductive land [16, 17].According to Myburg [18] and Iglesias Trabado and Wilstermann [19], currently there are more than 100 countries across six continents around the world planting *Eucalyptus* and covering over 20 million ha, making it the most widely planted broad-leaved tree species worldwide. From 1990 to 2015, the global *Eucalyptus* plantation area increased by 16.57 million ha, with an average annual increase of 1.1 million ha. The ratio of the area of global *Eucalyptus* plantations to planted forests area has also increased from 3.41% to 7.80% within the same period [20]. According to Wen et al., the *Eucalyptus* plantation area in the top 15 countries accounts for nearly 90% of the world's total eucalyptus plantation area. Brazil (22%) has the largest proportion of *Eucalyptus* plantation area in the world, followed by China (20%), India (17%), Australia (4%), Uruguay (3%), Chile (3%), Portugal (3%), Spain (3%), Vietnam (3%), South Africa (3%), Sudan (2%), Thailand (2%), Peru (2%), Argentina (1%), and Pakistan (1%). Although the genus *Eucalyptus* includes more than 700 species and their varieties, those planted for industrial purposes do not surpass a dozen. The "big nine" species (*Eucalyptus camaldulensis*, *E. grandis*, *E. tereticornis*, *E. globulus*, *E. nitens*, *E. urophylla*, *E. saligna*, *E. dunnii*, and *E. pellita*) and their hybrid are dominating 90% of the current *Eucalyptus* plantation [21]. These plantations have the potential to be easily certified with environmental certification schemes such as the Forest Stewardship Council (FSC) if the good forestry practices are followed along the productive chain.

2. Utilization of *Eucalyptus* spp. Fabricating EWPs

Eucalyptus plantations have the potential to be a valuable resource for the timber and fibre industries. However, most countries rely on *Eucalyptus* plantations primarily for low-value applications such as pulp, energy products, or board [22]. Most *Eucalyptus* species are rarely processed into sawn lumber due to the problems related to poor dimensional

Figure 2: Area of plantation forest and other planted forests, by region and subregion, 2020 [1].

stability, regular knots, cell collapse, excessive radial and tangential shrinkage rate, splitting, warp, and brittle heart during processing [23–25]. Splitting, cracking, and warping is most pronounced during the drying process and can be reduced by joining the wood into engineering products or composite components before the drying process [26, 27]. Splitting at the time of logging and warping during milling can be caused also by growth stresses [28, 29]. Growth stresses are often responsible also for brittle heart, especially in large older trees [30]. Most of these problems can be mitigated by applying the heat treatment to *Eucalyptus* logs [31], harvesting young eucalyptus trees, logs sawmill processing, and joining timber into EWPs before drying the wood [24, 32].

2.1. Types of EWPs.

EWPs are a type of manufactured composite material made from hardwoods and softwoods. These products are frequently processed to improve their quality and capacity. EWPs comprise a wide range of product types with a variety of manufacturing processes and applications. Particleboard, plywood, fibreboard, oriented strand board (OSB), laminated veneer lumber (LVL), glue laminated timber (GLT), and cross-laminated timber (CLT) are examples of engineered wood products [33] (Figure 3).

Eucalyptus wood could be potentially converted into a wide variety of EWPs. For instance, in the production of hardboard, *Eucalyptus* fibres are preferred. *Eucalyptus* fibres are short, according to Hillis and Brown [34], and thus do not easily form little clumps or masses like lengthy fibres do.

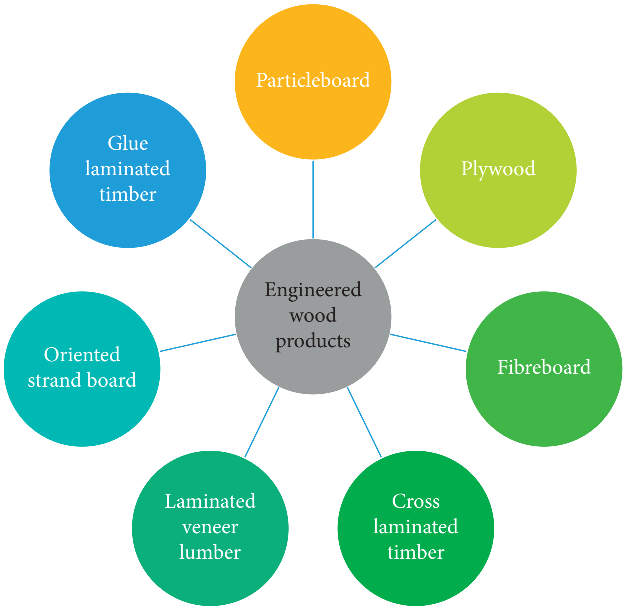

Figure 3: Types of engineered wood products.

As a result, the panels produced are acknowledged to have good surface attributes. Furthermore, when compared to other softwood species, *Eucalyptus* fibres have substantially higher strength qualities. As a result, the boards do not require any additional adhesive to provide the requisite strength. Therefore, this review would mainly focus on the studies reported *Eucalyptus*-based engineered wood products including particleboard, plywood, fibreboards, OSB, LVL, GLT, and CLT.

2.2. Publications on Eucalyptus-Based EWPs.

Scopus database was used to search for the publications on EWPs fabricated from *Eucalyptus* spp. wood. The results are displayed in Figure 4.

The most extensively reported EWPs made from eucalyptus are particleboard, which has 127 publications in the Scopus database dating back to 1990. Researchers from Brazil were responsible for over three quarters of the articles. With a total of 100 articles, plywood is the second most widely reported product, with researchers from China and Brazil dominating the field. The production of fibreboard panels derived from eucalyptus wood was covered in 34 publications, whereas the OSB was covered in 20. Between 2012 and 2021, 25 publications on *Eucalyptus*-based CLT were published. Researchers from Brazil, Australia, and China dominated the published data. Between 2003 and 2021, 15 articles on glue laminated wood were found, with Brazilian researchers dominating the published data once again. The only exception is LVL, for which Australian researchers have made the biggest contributions. Since 2013, there have been a total of 18 publications on LVL made from *Eucalyptus*. The increasing numbers of publications over the years reveal that the application of *Eucalyptus* wood in the manufacturing of EWPs has become more and more important. In recent years, CLT is the most widely researched type of EWPs worldwide.

2.3. Particleboard.

Several studies have demonstrated the practicality of using *Eucalyptus* species in particleboard manufacture. Da Rosa et al. [35] made particleboards from five eucalyptus species, i.e., *Eucalyptus benthamii*, *E. dunni*, *E. grandis*, *E. saligna*, and *E. urograndis*. As a control, particleboard produced from *Pinus taeda* was used. When compared to particleboard made from *P. taeda*, the results showed that particleboard made from *Eucalyptus* species had a higher modulus of rupture (MOR) and modulus of elasticity (MOE). The particleboard manufactured from *E. grandis* had the greatest MOR and MOE values. Particleboard manufactured from *E. grandis* also had the highest internal bonding (IB) strength. Overall, all of the *Eucalyptus* species met the minimum European EN standard requirements to the MOR and MOE values [36]. Only particleboards manufactured from *E. grandis* and *E. saligna* exceeded the minimum requirement of 18 MPa for MOR when compared to Standard NBR14810-2 (2006) [35]. On the other hand, Rangel et al. [37] employed *E. urophylla* to make particleboard and found that the mechanical qualities of the boards met the German Standards Institute (DIN) and the Venezuelan Industrial Standards Commission's basic standards (COVENIN). In terms of water absorption (WA) and thickness swelling (TS), all *Eucalyptus*-based particleboard had greater dimensional stability than the control, as evidenced by lower WA and TS values.

Figures 5 and 6 display the MOE-density chart and MOR-density chart for particleboard made with *Eucalyptus* spp. and other wood species. From the figures, one can see that the bending strength of the particleboard made of different raw materials does not necessarily follow the trend of strength improved along with increasing density. The findings have been supported by Klimek and Wimmer [47]. However, it does prove that the particleboard manufactured from *Eucalyptus* species has comparable or even better bending strength compared to that of other wood species. Even at lower board density, particleboard made from *Eucalyptus* species displayed better MOE and MOR than that of particleboard made from pine, poplar, and rubber wood.

Niekerk and Pizzi (1994) reported data from studies conducted at a South African particleboard factory which utilised *E. grandis* as raw material and a tannin-based adhesive to produce a moisture-resistant product [48]. The authors outlined two important problems that had to be overcome, i.e., the low pH of the eucalypt furnish, particularly in the high steam environment in the mattress during hot pressing, which inhibited the tannin adhesive curing, and the resistance of the *Eucalyptus* wood particles to crushing during the hot pressing process, which resulted in poor dimensional stability of the fabricated particleboards. Cabral et al. (2007) investigated the properties of particleboards made with particles generated from planer shavings of *E. grandis*, *E. urophylla*, and *E. cloeziana*, bonded with urea-formaldehyde (UF) adhesive [49]. Slash pine *(Pinus elliottii)* particles were mixed with *Eucalyptus* particles to achieve a target panel density of 700 kg.m^{-3}. Overall, particleboards fabricated with the highest proportions of *Eucalyptus* particles demonstrated the highest WA and TS values. Pan et al. (2007) studied the properties of thin particleboard panels fabricated from *E. cinerea*, bonded with polymeric 4,4-diphenylmethane diisocyanate (pMDI) and UF resin [39]. The properties of the panels were compared with those made from *E. camaldulensis*. The authors investigated a wide range of production parameters, i.e., particle size, resin type and addition level, bark content, and hot-water pretreatment. In general, particleboards produced from *E. cinerea* wood particles exhibited significantly better properties than those made from *E. camaldulensis*.

2.4. Fibreboard.

Fibreboard is a term used to describe a flat-pressed EWP manufactured from wood fibres obtained by thermomechanical wood pulping and traditionally bonded with a synthetic adhesive. In addition, hardboards represent a flat-pressed EWP composed of randomly oriented wood fibres obtained by thermomechanical wood pulping and bonded without an adhesive by hot pressing by the very high density (900–1100 kg.m^{-3}) and the high-temperature-induced flow of the lignin component of the fibres [50]. *Eucalyptus* has been shown in several researches to be a potential material to produce fibreboard panels. Several *Eucalyptus* species have been reported to be used as feedstocks for medium-density fibreboard (MDF), for example, *E. obliqua*, *E. sieberi*, *E. globoidea*, *E. loxophleba*, and *E. rudis* [51]. Krzysik et al. [52] used *E. saligna* to make medium-density fibreboards (MDF) in three thicknesses (6 mm,

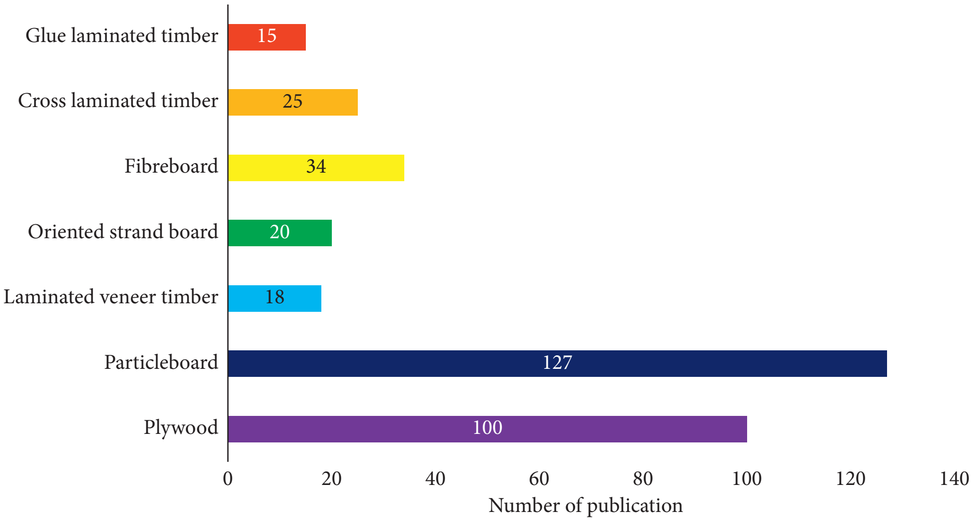

Figure 4: Number of publications on EWPs fabricated from *Eucalyptus* spp.

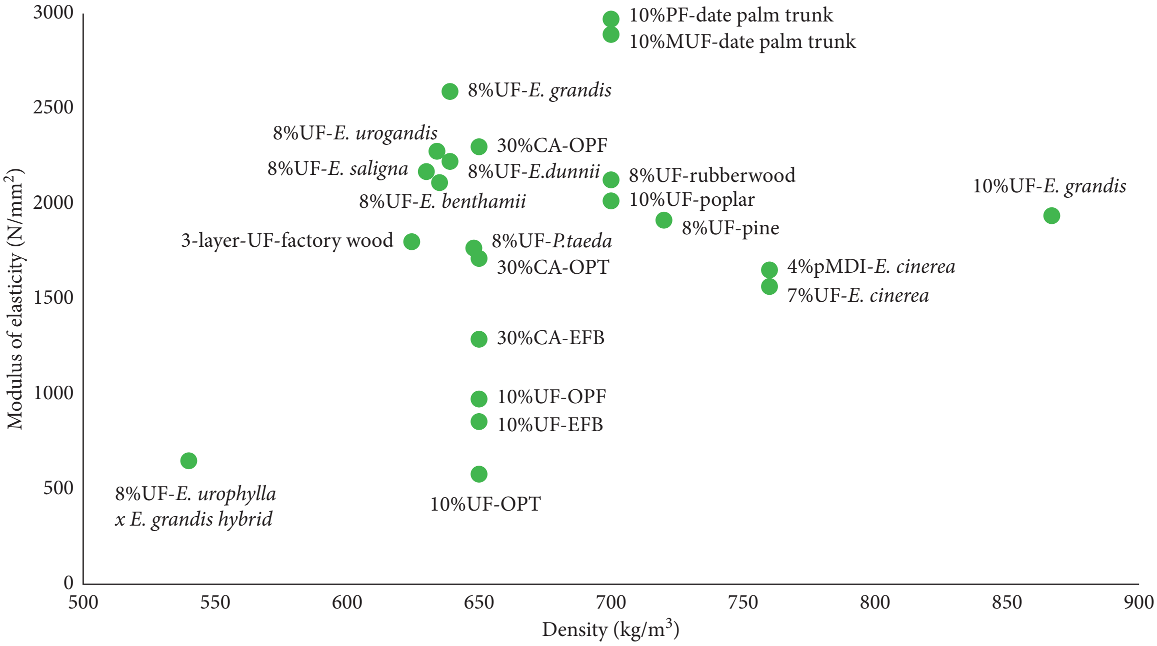

Figure 5: MOE-density chart for particleboard made with *Eucalyptus* spp. and other wood species [35, 38–46]. *Note: UF: urea-formaldehyde; PF: phenol-formaldehyde; MUF: melamine-urea-formaldehyde; pMDI: polymeric 4,4-diphenylmethane diisocyanate; CA: citric acid; OPT: oil palm trunk; OPF: oil palm frond; EFB: empty fruit bunch.

13 mm, and 19 mm), bonded with 10% UF resin. The produced MDF was compared to the specifications of the Interior ANSI A208.2 MDF standard [53]. The developed MDF panels exceeded the minimum MOR criteria established by the ANSI A208.2 standard for all three thicknesses examined. The laboratory-fabricated MDF panels of all three thicknesses exceeded the ANSI minimum standard requirements to MOE and IB values. Pranda [54] reported MDF panels fabricated from *E. globulus* and found that the resulting MDF had higher WA and TS than MDF made from *Pinus pinaster*. Furthermore, to achieve comparable mechanical properties, MDF panels made from *E. globulus* required a higher resin amount than MDF made from *P. pinaster*.

Some authors demonstrated the potential of *Eucalyptus* as a feedstock in manufacturing binderless fibreboards. Most of this research is based on the oxidative modification of lignin [55, 56]. Authors in [57] prepared a binderless board from *Eucalyptus grandis* of hydrothermal pretreated *Eucalyptus* wood fibres and characterized it in terms of chemical analyses, mechanical strength, and self-bonding mechanism. The reduction of lignin content of the *Eucalyptus* wood after hot pressing resulted in a decrease in the glass transition temperature and decrease of the softening point of lignin,

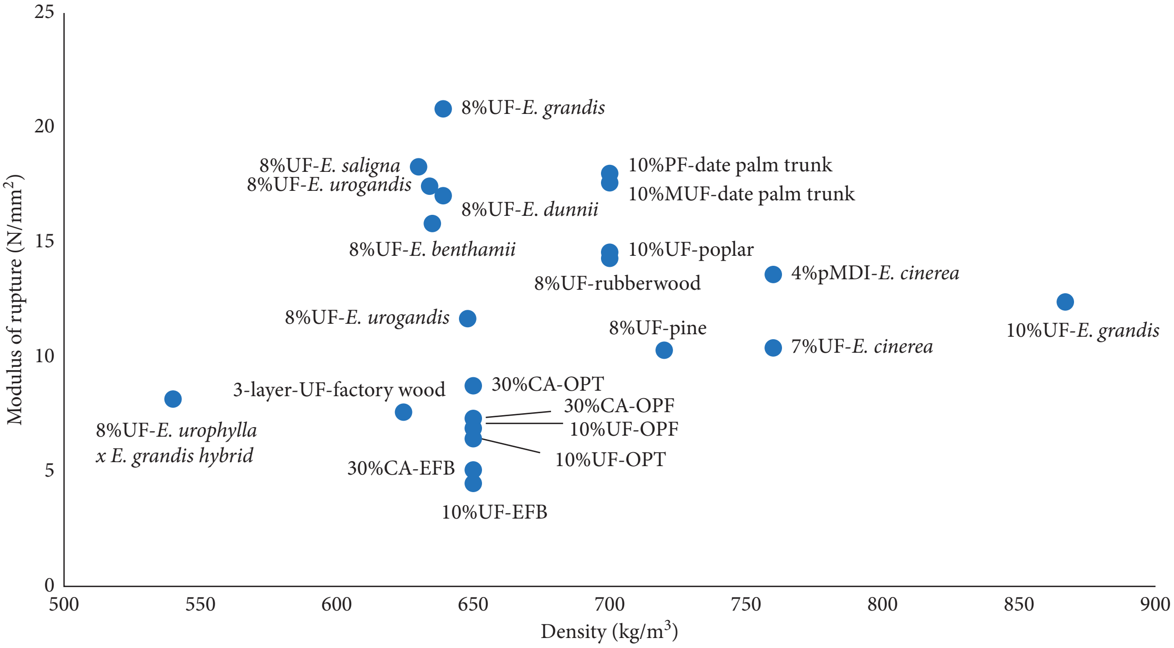

Figure 6: Modulus of rupture- (MOR-)density chart for particleboard made with *Eucalyptus* spp. and other wood species [35, 38–46]. *Note: UF: urea-formaldehyde; PF: phenol-formaldehyde; MUF: melamine-urea-formaldehyde; pMDI: polymeric 4,4-diphenylmethane diisocyanate; CA: citric acid; OPT: oil palm trunk; OPF: oil palm frond; EFB: empty fruit bunch.

which makes lignin more accessible to the fibre surface and plays important role in the self-bonding mechanism [58, 59]. It was explained that high IB value was caused by the intermolecular hydrogen bonding between the cellulose and lignin molecule [60]. In general, it was suggested that the combination of hydrothermal pretreatment and hot pressing process is a good way for conditioning *Eucalyptus* sawdust for the production of binderless boards [61]. Other authors in [62] used Kraft lignin (KL) from *E. globulus* with laccase from *M. thermophila* in a two-component system enabling the manufacturing of MDF panels totally free of synthetic resins or additives, with *E. globulus* as the main raw material. The MDF panels exhibited remarkable high IB and low TS values.

2.5. OSB. OSB is a type of flat-pressed EWP comparable to particleboard that is made by applying thermosetting adhesive resins to layers of wood strands and compressing them. Traditionally, wood from the *Pinus* genus has been used to make OSB [63]. The density of OSB made in the United States is normally between 500 and 800 kg.m^{-3}. The fabrication of panel products, according to Molesmi [64], requires timber with a density of up to 550 kg.m^{-3}. As a result, *Eucalyptus* species could be used to replace *Pinus* spp. wood. As a source of raw materials in the production of OSB, *E. grandis* has been reported as a suitable feedstock for the production of OSB panels [65]. According to the study, using 4.5 and 6% phenol-formaldehyde (MUF) resin is enough to produce OSB panels with mechanical strength complying with the Canadian Standard CSA 0437–0. A study by Domingos and Moura [66] also demonstrated that the *Eucalyptus* bark could be a promising material for the manufacturing of OSB panels.

Following that, several *Eucalyptus* species that are suited for OSB production have been found. Iwakiri et al. [67] used 6% PF resin to make OSB from six different *Eucalyptus* species: *E. grandis*, *E. dunnii*, *E. tereticornis*, *E. saligna*, *E. citriodora*, and *E. maculate*. The OSB panels were produced at a density of 700 kg.m^{-3}. The findings demonstrated that, in terms of physical and mechanical qualities, *E. grandis* and *E. saligna*, particularly *E. grandis*, had a lot of potentials for making OSB. When compared to OSB created from *P. taeda*, OSB made from *E. grandis* had equivalent or superior physical and mechanical properties. The potential of using *E. grandis* and *E. urophylla* as feedstocks in non-oriented and oriented panels was investigated by Gouveia et al. (2000) [68]. The authors concluded that *E. grandis* was the more suitable raw material. In addition, Gouveia et al. (2003) also investigated blends of *E. grandis* and *Pinus elliottii* feedstocks for OSB panels, and optimum results were reported with blends comprised between 50% and 75% *E. grandis* wood [69]. Another study by da Rosa et al. [35] looked at the technical feasibility of using five different *Eucalyptus* species to make OSB. *E. benthamii*, *E. dunni*, *E. grandis*, *E. saligna*, and *E. urograndis* are among the *Eucalyptus* species employed. When compared to OSB panels made from *P. taeda*, OSB fabricated from *Eucalyptus* species demonstrated higher dimensional stability. Furthermore, the MOE value of *Eucalyptus* OSB was higher than that of pine OSB, while there was no significant difference in MOR between the two panels. In comparison to OSB made of pine, the IB strength of OSB manufactured of *Eucalyptus* was shown to be lower. All five species, however, have been identified as suitable for the production of OSB. The practicality of OSB manufactured from *E. grandis* and *E. dunnii* was compared by Iwakiri et al. [70]. OSB was manufactured in two densities: 700 kg.m^{-3} and 1000 kg.m^{-3}. OSB produced from *E. grandis* with a density of 700 kg.m^{-3} met the minimum requirements set out in the Canadian and European standards. OSB panels with a higher density of 1000 kg.m^{-3} exhibited greater mechanical strength, allowing them to be used in more demanding load-bearing applications.

2.6. LVL.

LVL is a type of EWP that is made up of numerous layers of thin wood that are adhered together. Owing to its uniform engineering properties and dimensional flexibility, LVL is a vital EWP, particularly for the construction of buildings. Several studies have proved that the LVL fabricated from *Eucalyptus* spp. has comparable physical and mechanical properties to that of LVL made of poplar, beech, and even Norway spruce [71, 72]. Large faults can be avoided when logs are cut into thin veneers and assembled and glued parallel to the grain, which may explain why reported LVL values are typically higher than those achieved for other traditional hardwood products [73]. Bal and Bektaş [74] stated that LVL is typically made from softwood species and low- and medium-density hardwood species with densities ranging from 290 to 693 kg.m^{-3}. Several studies have found that *Eucalyptus* species are suitable for the generation of LVL. *E. grandis* was used to make LVL in a study by Bal [72], and it was compared to LVL made from poplar. The mechanical properties of *E. grandis* LVL were superior to those of poplar LVL, owing to the higher density of the *E. grandis* veneers. Meanwhile, *E. globoidea* was utilised to make LVL by Guo and Altaner [75]. Unfortunately, despite the high quality and good drying properties of the veneers acquired, the bond performance of the LVL produced is inadequate and does not fulfill The New Zealand standards. Despite some promising results that have been shown, manufacturing of LVL from *Eucalyptus* still encounters some problems such as glue difficulty, end-splits, and breakage of sheets of veneer [76]. Adhesive failure is one of the main issues encountered during the production of *Eucalyptus*-based LVL. However, this issue can be ameliorated by alternating lamination using poplar and *Eucalyptus* veneers as demonstrated by Murata et al. [77]. The authors found the alternating lamination using softer poplar veneers could reduce the quality variations of *Eucalyptus* and hence improved its variation in the modulus of elasticity. Apart from LVL, a new type of structural composite lumber called oriented strand lumber (OSL) has also been developed from *Eucalyptus* spp. Quite similar to that of LVL, OSL is fabricated using strands of wood or strips of veneer with a primary orientation along the length of the member. Chen et al. [78] manufactured oriented oblique strand lumber from *Eucalyptus* wood (*Eucalyptus urophylla* and *E. grandis*) and investigated their mechanical properties. The results revealed that OSL made from *Eucalyptus urophylla* and *E. grandis* had a better compressive and tensile strength in parallel-to-grain direction compared to that of the Sitka spruce, Douglas fir LVL, Spruce-pine-fir (SPF), GLT, and glue laminated bamboo. *Eucalyptus*-based OSL also exhibited higher flexure strength than all of the other wood and bamboo-based products in the study. The study has proven the feasibility of utilizing fast-growing *Eucalyptus* for general use in construction.

2.7. Plywood.

The findings reported in the scientific literature on plywood panels manufactured from *Eucalyptus* are presented in Table 1. Most researchers were interested in the effects of species, type of adhesive, and grain direction on the mechanical properties of the plywood panels produced.

Except for the layer configuration, plywood is very similar to LVL. Plywood is made up of thin layers of wood veneer called “plies” that are bonded together with adjacent layers’ wood grain rotated up to 90° from one another. For the manufacturing of plywood, Iwakiri et al. [79] used the veneers of nine *Eucalyptus* species. *E. viminalis* was shown to produce the greatest overall outcomes. Meanwhile, *E. phaeotricha* and *E. pellita* veneers had lamination yields of less than 50%. The shear strength of *E. robusta*, *E. dunnii*, and *E. deanei* plywood was not equal to or more than 1.0 MPa. As a result, four *Eucalyptus* species, namely, *E. grandis*, *E. saligna*, *E. globulus*, and *E. viminalis* were recognised as having promising potential for making exterior-use plywood.

E. pellita showed great potential in plywood manufacturing although there are some inferior properties in the veneers. The shear strength and MOR surpassed the minimum requirements. This is supported by the study conducted by Muhammad-Fitri et al. [81]. The authors investigated the effects of layers number and species arrangement on plywood made from batai (*Paraserianthes falcataria*), eucalyptus (*E. pellita*), and *kelempayan* (*Neolamarckia cadamba*). Five and seven layered plywood panels were produced with different species arrangements. For the 5-layer plywood, the arrangement was as follows: BBBBB, KBKBK, KEKEK, and KKKKK. The 7-layer plywood was a repetition of the 5-layer plywood where two more veneer layers were added using a similar sequence as of the 5-layer plywood. The results revealed that plywood made from a combination of *kelempayan* and *Eucalyptus* had significantly higher mechanical strength compared to that of plywood made from *kelempayan* solely especially in the perpendicular direction. The authors attributed it to the higher density of *Eucalyptus* veneers which provide higher strength to the plywood. Bal and Bektaþ [80] studied the effects of timber species regarding their density on the mechanical properties of the plywood produced. The mechanical strength values were divided by their corresponding density to minimize the effect of density. They found out that the specific MOE values of plywood fabricated from eucalyptus veneer were the highest among the other studied timber species. However, the specific MOR only showed a slightly higher value when compared to other species. The authors concluded that *Eucalyptus* wood provided sufficient strength to the final product and was able to enhance the performance of plywood when incorporated with other commercial timber

Table 1: Studies on *Eucalyptus* plywood panels.

Eucalyptus species	Variables	Properties tested and findings	Reference
Eucalyptus grandis, Eucalyptus saligna, Eucalyptus globulus, Eucalyptus viminalis, Eucalyptus dunnii, Eucalyptus robusta, Eucalyptus phaeotricha, Eucalyptus deanei, and *Eucalyptus pellita*	Species parallel and perpendicular to the plane	**MOR (N/mm^2)** 72.23–115.68 (parallel) 39.46–53.43 (perpendicular) **MOE (N/mm^2)** 9378–18494 (parallel) 2738–4627 (perpendicular)	[79]
Eucalyptus grandis *Fagus orientalis* Hybrid poplar *(Populus x euramericana)*	Species *Eucalyptus grandis* (A), *Fagus orientalis* (B), and hybrid poplar (C) Direction of load (parallel and perpendicular) Type of adhesive UF, MUF, and PF	**Specific modulus of rupture (SMOR, N/mm^2)** 12–13 (A, parallel), 6.2–6.7 (A, perpendicular) 12.3–13.1 (B, parallel), 5.4–6.0 (B, perpendicular) 12.5–12.9 (C, parallel), 5.9–6.2 (C, perpendicular) **Specific modulus of elasticity (I, N/mm^2)** 1915–1596 (A, parallel), 477–515 (A, perpendicular) 1242–1273 (B, parallel), 394–403 (B, perpendicular) 1459–1548 (C, parallel), 458–478 (C, perpendicular)	[80]
Paraserianthes falcataria *Neolamarckia cadamba* *Eucalyptus pellita*	Species arrangement Batai (B), *kelempayan*-batai (KB), *kelempayan-Eucalyptus* (KE), and *kelempayan* (K) Number of layers 5 layers, 7 layers Bending (parallel and perpendicular)	**MOR (N/mm^2)** 20.38–40.04 (parallel) 32.39–59.82 (perpendicular) **MOE (N/mm^2)** 2453–4781 (parallel) 2879–6027 (perpendicular)	[81]

species. The research performed by Farrell et al. (2011) was focused on assessing the potential of *E. nitens* and *E. globulus* to produce veneer and plywood. The material studied was comprised of two ages *of E. nitens*, i.e., 16- and 26-year-old and 33-year-old *E. globulus*. All plywood made from the *E. globulus* and the 26-year-old *E. nitens* veneer using phenolic adhesive achieved type A bond quality, while the results for plywood manufactured from the 16-year-old *E. nitens* veneer were variable [82].

2.8. GLT. GLT is a structural EWP made up of layers of dimensional lumber bound together with long-lasting, moisture-resistant structural adhesives. In Europe, there is a growing interest in glued laminated structural products made of hardwoods due to a variety of factors, including a lack of softwoods. In addition, hardwoods are abundant owing to the policies of reafforestation using several hardwood species due to better adaptation to soil and climate conditions. Moreover, in most circumstances, GLT made of hardwoods has higher bending strengths than the highest European softwood GLT strength classes, which are often constructed of spruce or pine [83]. Castro and Paganini [84] used a combination of poplar and Uruguayan *E. grandis* to demonstrate the potential of *Eucalyptus* in making structural glue laminated timber. Castro and Paganini [85] conducted a follow-up investigation in which *E. grandis* of four distinct clones was employed in the manufacturing of glue laminated timber. The glue laminated lumber manufactured from *E. grandis* has shown exceptional mechanical strength and structural efficiency. Apart from *E. grandis*, Tasmanian Oak (*E. regnans*/*obliqua*/*delegatensis*) and Blackbutt (*E. pilularis*) have also been glue laminated and their hygroscopic behaviour was examined [86]. Suleimana et al. [87] fabricated glue laminated timber from Portuguese *Eucalyptus* (*E. globulus* Labill.) and concluded that the *E. globulus* is suitable to be used in the production of glue laminated timber. *Eucalyptus* glue laminated timber is suited for structural purposes. In their investigation, Lara-Bocanegra et al. [88] found that glue laminated *E. globulus* timber joined with polyurethane (PUR) adhesive can reach GL45 strength class. If superior solid wood grades were employed, the GL48 strength class may be achieved. It is worth noting that if MUF resin was used to connect the glue laminated timber, strength classifications of GL56 or higher may be achieved. In comparison to the other species, the created *Eucalyptus* glue laminated lumber behaved flawlessly as grid-shells, according to the authors. Petrauski et al. [89] employed glue laminated timber made from *Eucalyptus* sp. to construct porticos. The structures performed admirably and mechanically and demonstrated a high level of technical feasibility in the development of porticos. Carrasco et al. [90] used *E. citriodora* to make glue laminated timber sleepers, and the results were satisfactory, proving the viability of employing *E. citriodora* to make sleepers.

2.9. CLT. Table 2 summarised the findings of *Eucalyptus* CLT panels from various literatures. Most researchers were interested in the effects of species, type of adhesive and primer treatment, strength direction, and timber grade on the mechanical properties of the CLT panels manufactured.

A few *Eucalyptus* species have already been employed in the manufacturing of CLT. According to the literature, the most common resins used to bind CLT are one-component polyurethane adhesive (1C PUR) and MUF resin. Liao et al. (2017) used hybrid *Eucalyptus* wood (*E. urophylla* × *E. grandis*) to make CLT, which they glued with a one-component polyurethane adhesive (1C PUR). They studied the pressing parameters and strength directly on the properties of CLT panels. They found out that the optimal glue spread rate, pressing pressure, and pressing time for the manufacturing of *Eucalyptus* CLT panels were 160 g.m^{-2}, 0.8 N.mm^{-2}, and 200 min, respectively. Mechanical qualities of the resulting CLT panels were comparable to commercially available CLT [96]. Other researchers also concluded that pressing pressure of 0.7 N.mm^{-2} was sufficient to produce CLT panels with sufficient bonding quality without stress grove [94]. Nonetheless, Lu et al. [92] found that using a commercial one-component polyurethane glue led to CLT specimens with lower delamination and shear force resistance compared to commercial softwood CLT. The authors investigated the block shear strength (BSS), wood failure percentage (WFP), and delamination rate (RD) of CLT panels manufactured from hybrid *Eucalyptus* wood (*E. urophylla* × *E. grandis*) using different adhesives and surface primers systems. Four types of adhesives, i.e., epoxy resin (EP), emulsion polymer isocyanate (EPI), phenol resorcinol formaldehyde (PRF), and PUR, were used. Meanwhile, two surface primers, i.e., N, N-dimethylformamide (DMF) and hydroxymethylated resorcinol (HMR), were incorporated. According to the findings, all the adhesives can be utilised to make CLT. Due to its excellent bonding performance and mechanical qualities, CLT bonded with PUR adhesive demonstrated the best properties of all the studied adhesives. The authors also showed that the application of primer can further improve the BSS and WFP of the CLT specimens with HMR primer showing the highest increase in performance. However, RD of CLT showed no significant improvement with the application of surface primer treatment. Therefore, with the suitable adhesive and primer system, the shortcoming of *Eucalyptus* CLT can be overcome. Pangh et al. [93] employed *E. nitens* and *E. globulus* from high-grade and low-grade boards based on their MOE in the production of CLT and discovered that CLT was created from these two eucalyptus species and has better flexural qualities than CLT made from other eucalyptus species. In terms of MOE and MOR, CLT made from *E. globulus* outperformed *E. nitens* between the species evaluated. As expected, the CLT fabricated from high-grade timber board also showed better mechanical performance than their low-grade counterparts. However, the authors found out that the failure mode of the specimens was dependent on the grade of the timber board used. Bending failure on the tensile zone was common among the specimens fabricated from low-grade timber boards while rolling shear failures were observed in the specimens fabricated from high-grade timber boards. Findings from Pereira and Calil [97] also support that *Eucalyptus* wood is an ideal material in the production of CLT as CLT made from *E. urograndis* showed better properties than the CLT made from *Pinus taeda*. Another important aspect of the properties of CLT that needs to be taken into consideration is the rolling shear properties of the transverse layer in the CLT panel. Gui et al. [95] conducted a study to investigate the effect of aspect ratio on the rolling shear properties of CLT made from commercial SPF dimension lumber (spruce-pine-fir) and *E. urograndis*. They concluded that the rolling shear properties of CLT panels fabricated from *Eucalyptus* showed promising results where the rolling shear strength and rolling shear modulus wood were 88% and 260% higher than CLT panels made from SPF lamination.

3. Challenges and Future Perspectives

Even though *Eucalyptus* spp. have shown considerable potential for manufacturing EWPs, various barriers prohibit it from being used more efficiently. The challenges could come from two factors. One factor is the current global trend of the *Eucalyptus* plantation. Another factor is the technical issue of the *Eucalyptus* wood itself. The development of plantation forests is the universal consensus and common action of global climate and ecological governance. Affected by the available forest resources, site conditions, climate change, and public opinion, the development strategies of *Eucalyptus* plantations in countries around the world have undergone major changes. Many countries have altered from encouraging to restricting the development of *Eucalyptus* plantations, making the prospects of *Eucalyptus* plantations uncertain (Wen et al., 2018). According to Wen et al. [20], the future development of *Eucalyptus* plantation is restricted by (i) unsustainable management of eucalyptus plantation under the short-cycle multigeneration continuous planting system, (ii) limited development space of *Eucalyptus* due to shortage of forest land resources, (iii) declining forest stand quality, and (iv) public opinion and the boycott of the development of *Eucalyptus* due to its excessive consumption of soil nutrients and groundwater and negative impact on biodiversity. The high number of growth stresses found in the logs is one of the key challenges. The problem was complicated by the fact that the specific chemical process responsible for the formation of such enormous stresses is still unclear [98]. Processing actions like falling, sawing, and veneer peeling resulted in the production of this growth-stressed substance. As a result, various faults occurred throughout the peeling process, including end-splitting of logs, distortion of sawn boards, and veneer cracking [25]. Worse, these flaws are more visible in plantation species with smaller log diameters. The quality and recovery of *Eucalyptus* veneers were ultimately harmed

Table 2: Studies on CLT made from *Eucalyptus* spp.

Timber species	Variables	Properties tested and findings	Reference
Hybrid *Eucalyptus* wood (*Eucalyptus urophylla* × *E. grandis*	Pressing parameters Glue spread rate (A), pressing pressure (B), and pressing time (C) Strength direction Major strength direction (E0) and minor strength direction (E90)	**Optimal pressing parameter** $A = 160\,\mathrm{g/m^2}$, $B = 0.8\,\mathrm{N/mm^2}$, and $C = 200$ min **MOR ($\mathrm{N/mm^2}$)** 23.8–24.5 (E0) 8.2–9.0 (E90) **MOE ($\mathrm{N/mm^2}$)** 11043–12034 (E0) 661–709 (E90)	[91]
Hybrid *Eucalyptus* wood (*Eucalyptus urophylla* × *E. grandis*	Adhesive Epoxy resin (EP), emulsion polymer isocyanate (EPI), phenol resorcinol formaldehyde (PRF), and polyurethane (PUR) Primer N, N-dimethylformamide (DMF) and hydroxymethylated resorcinol (HMR)	**Delamination rate (RD, %)** 7.6–15.7 CLT bonded with EPI adhesive displayed the highest RD value at 15.7%. Meanwhile, PUR bonded with CLT showed the lowest RD rate at 7.6%. **Block shear strength (BSS, $\mathrm{N/mm^2}$)** 3.01–3.51 (dry state) 1.01–1.62 (wet state) HMR primer increased the BSS values of PRF and PUR bonded with CLT by 31.5% and 4.9%, respectively. **Wood failure percentage (WFP, %)** 73.2–85.6 (dry state) 47.5–58.2 (wet state) HMR primer enhanced the WFP values of *Eucalyptus* CLT at wet state bonded with PRF and PUR adhesives by 27.8% and 12.4%, respectively.	[92]
Eucalyptus nitens and *Eucalyptus globulus*	Species Stress grade of timber	**MOR ($\mathrm{N/mm^2}$)** 41.3–48.6 (*E. nitens*) 56.4–62.7 (*E. globulus*) On average, CLT panels fabricated from *E. globulus* showed 32.5% higher MOR. **MOE ($\mathrm{N/mm^2}$)** 9433–11695 (*E. nitens*) 11250–13610 (*E. globulus*) On average, CLT panels fabricated from *E. globulus* showed 17.7% higher MOE.	[93]
Eucalyptus grandis	Testing method Delamination test EN 16351 (test A), block shear test EN 16361 (test B), block shear test at 45° grain direction (test C), and delamination and shear test at 45° grain direction (test D) Density, grooves, and pressure effect	**Delamination values (%)** 9.7–42.8 (test A) 14.3–58.8 (test D) Higher density and the presence of groove resulted in greater delamination values. In contrast, higher pressure resulted in lower delamination. **Shear strength ($\mathrm{N/mm^2}$)** 3.65–4.96 (test B) 5.08–6.79 (test C) 0.67–2.33 (test D) Higher density and pressure resulted in higher shear strength. In contrast, the presence of grooves resulted in lower shear strength.	[94]
S dimension lumber (spruce-pine-fir) *Eucalyptus urophylla*	Species Aspect ratio	**Rolling shear strength ($\mathrm{N/mm^2}$)** 3.46–3.65 **Rolling shear modulus ($\mathrm{N/mm^2}$)** 375–495	[95]

as a result of these flaws. Only 20% of useable veneers from *E. grandis* were recovered after severe end-splitting, according to Margadant [99]. Unfortunately, no technological solution to this problem has yet been discovered [75]. Apart from faults produced by growth pressures, another issue that needs to be handled is collapse and internal checking during the drying of eucalyptus timber. *Eucalyptus* lumber is difficult to dry due to its limited permeability and the presence of tyloses in the heartwood [100]. Crafford and Wessels [23] found that *E. grandis* has a very high shrinkage and expansion coefficient, with 30% of the *E. grandis* exhibiting warping that did not meet structural lumber specifications. This warping has created challenges in CLT manufacture, where good cross-grain face bonding is essential. According to Ananías et al. [101], drying flaws increased with the increased drying rates and temperatures applied. Even with very slow and cautious drying regimens, the collapse is almost unavoidable.

Despite the aforementioned concerns, various advances have been made to alleviate, if not eliminate, the obstacles associated with effective *Eucalyptus* timber utilisation. Wessels et al. [24] suggested some strategies for addressing the problems, including harvesting *Eucalyptus* trees at a young age, sawmill processing, and green-gluing the lumber into engineered wood products. Green gluing is the method of employing structural adhesive to join green lumber to engineered wood products before the drying process. As the adjacent pieces limit each other, drying-induced splits, cracks, and warping can be reduced. The development of structural adhesive that can be applied to green lumber above the fibre saturation point has made green glued engineered items viable. In the meanwhile, choosing the wood based on its radial placement inside the stem is critical for preventing drying-induced collapse. Wood recovered from the central region of eucalyptus lumber is less prone to collapse, according to Ananías et al. [101] than wood extracted from the transition zone between the centre and the periphery. As a result, while the negative effects of growth stresses and the drying process cannot be eradicated, they can be managed with the right processing approach.

Further research works on the potential of using *Eucalyptus* spp. for manufacturing EWPs should be focused primarily on optimising tree breeding and improving silvicultural practices, e.g., breeding for optimum density and pruning to reduce the wood defects. This would potentially result in enhanced opportunities for wider utilisation of the *Eucalyptus* spp. wood as a promising feedstock for manufacturing EWPs.

Disclosure

A preprint version of this manuscript is available at https://www.researchgate.net/publication/357127148_Engineering_Wood_Products_from_Eucalyptus_spp_A_Review [102].

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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Review Article

A Review on the Effect of Various Chemical Treatments on the Mechanical Properties of Renewable Fiber-Reinforced Composites

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Increased environmental concerns and global warming have diverted effort all over the world to focus on renewable and sustainable resources for the next generation of composite products due to their recyclability, renewability, cost effectiveness, and satisfactory mechanical performance. Bio/natural fibers which are environment friendly materials employed as reinforcement have led to developing a biocomposite for reduction in greenhouse emission and carbon footprints. However, biofibers are also having some limitations that need to be addressed including poor compatibility between the reinforcing fiber matrices, high moisture absorption, swelling, poor chemical and fire resistance, and high dispersion of mechanical properties. A lot of research has been performed on physical and mechanical properties of natural fiber composite. Properties of such novel composite mainly depend on adhesion between fiber and matrices. Consequently, poor adhesion, high moisture absorption, and swelling lead to formation of crack in both the matrix and fiber. Therefore, numerous techniques have been tried till date to modify both fiber surfaces to enhance their adhesion and reduce their water absorption. This review article provides comprehensive information about effect of various surface modification techniques that include alkaline, silane, acetylation, permanganate, peroxide, benzoylation, acrylonitrile grafting, maleic anhydride grafted, acrylation, and isocyanate. In addition, the effects of cellulose, hemicellulose, lignin, and pectin of biofibers are also reported. This review concluded that chemical treatment of biofibers with 5% NaOH concentration improves the physical, mechanical, and thermal properties of the resulting composites compared to untreated fiber composites.

1. Introduction

Presently, the automotive industries do face many challenges, such as the shortage of supply of fossil fuels, new technological innovations, and environmental sustainability to fight global warming. The above listed factors increase the pressure in the current materials design and manufacturing technologies in automotive industries [1, 2]. A lightweight material was a necessity for the structural components for lower energy consumption of vehicles. The research community has shifted the focus towards effective utilization of the biofibers extracted from renewable sources. The natural fibers are being employed for application components such as interior dashboard trims for automobiles and household applications [3, 4]. The utilization of lightweight and low-cost biofibers such as abaca, banana, bamboo, coir, flax, ramie, tea leaf, pineapple, sisal, kenaf, and jute was already tried as reinforcements in the polymer matrix for composite materials used for manufacturing automobile components [5–7]. The biodegradability of biofibers is associated with physical, chemical, mechanical, thermal, and moisture conditions which have increased their scope of use in numerous applications [8, 9]. Based on the usage and utilization, usually biofibers are classified as primary and secondary fibers. Primary fibers include jute, sisal, kenaf, and hemp fibers, where these fibers are grown for their fiber contents. Secondary fibers include agroresidues, coir fibers, and pineapple fibers, which are fibers obtained from plant by-products. These fibers can be further classified into six types: bast fibers (flax, jute, etc.), leaf fibers (sisal, pineapple leaf fiber), seed fibers (cotton, coir), core fibers (hemp, kenaf), grass and reed fibers (wheat, corn, and rice), and all other types (wood and roots). The structure and chemical constituents of biofiber depend on several factors like extraction process, location of plant growth, climate, plant age, and plant nature [10–13]. When compared to synthetic glass fibers, natural fibers have better specific modulus. The cost saving on the material owing to the use of plant based fibers and nonabrasive nature of the materials during mixing and moulding makes the natural fibers a promising reinforcement for polymer composites [14]. These advantages make the biofiber be employed in any application like automobile, domestic utilities, and distinction [15, 16]. The biofiber composites were used in many places because of the following merits: eco-friendly nature, easy availability, low weight, good strength, low cost, and ease of manufacturing process [17–19]. The foremost disadvantages of the biofibers are that they absorb more moisture. The moisture absorption of biofibers has several unfavourable effects on their properties and thus affects the long-term performance of the composites [20]. The mechanical and thermal performances of bio/natural fibers reinforced composites (NFRC) were influenced by weight/volume fraction of fiber, fiber orientation, selection of chemical treatment method, and physical characteristics of the natural fiber [21–23]. However, the water absorption and thermal stability of the composite laminate were noticed to reduce with increase in weight fraction of fiber [24–28]. The natural fibrils are constituted of cellulose, hemicellulose, and lignin in varying percentages. In addition, the natural fibers do have other substances like pectin, wax, and other water soluble compositions. The cellulose is enclosed in soft lignin, while hemicellulose forms the ancillary layer of the fiber material [29]. Ishak et al. [30] studied the tensile strength of bagasse fiber obtained from plants of different heights. The tensile properties were specifically high for the fibers extracted from bottom of tree due to their chemical composition, particularly cellulose, hemicellulose, and lignin. Moisture absorption of the natural composites was increased when fiber content increased because of its higher cellulose content amount. George et al. [31] stated that the biofibers have high amount of hydrophilic property which leads to poor adhesion properties between the hydrophilic fiber and hydrophobic matrix. Hydrophilicity is the stronger affinity of the fibers towards moisture. The major disadvantages of natural fiber are poor adhesion between fiber and matrix, presence of cellulose content, moisture absorption, and voids at interface between fiber and matrix which results in dimensional inaccuracy, thus affecting the mechanical properties [32–35]. In addition to that, presence of high moisture content in fiber leads to swelling of fiber and matrix within composites resulting in dimensional instability. This disadvantage and limitation can be overcome by chemical treatments. The chemical treatments are carried out to reduce the hydrophilic nature of fiber but the surface treatments not only modify the fiber surface but also increase the fiber strength leading to the improvement of adhesion between fiber and matrix [36–38]. The most common chemical treatment and surface treatment methods are silane, alkaline, acetylene, maleated coupling, anhydride, and benzolytation. Optimization of fiber and matrix is aimed to improve the adhesion, surface tension, interfacial strength, and wettability that offer good surface roughness leading to good bond [39]. These could be done by adding suitable compatibilizer/coupling agent and chemically treating the fiber. Recent literature survey showed that several works have been published on different natural fibers extracted from renewable sources which are used as reinforcement with polymer matrix over a wide dimension of applications. This survey depicts various surface treatments done to enhance the properties of the fibers and for enrichment in mechanical properties of the composites in contrast with untreated fiber-reinforced polymer composites. Therefore we summarized the major findings on different types of chemical treatments and surface treatments.

2. Constitution of Biofiber

2.1. Cellulose. In plant based fibers, cellulose is the primary structural constituent and the cellulose portion influences the mechanical properties of the lignocellulose fibers. The cellulose is the fundamental constituent which is liable for the strength of the plant fibers and variance in strength may be due to growth conditions of the plant and soil nature. Cellulose is a lined, semicrystalline polysaccharide build-up of polymer links compromising the recurring modules of anhydroglucose grouped via 1,4-β-D-glucosidase. The recurring modules of the monomers are termed as degree of polymerization. The monomers of glucose in the chain of cellulose result in formation of the hydrogen bonds among the link forming fibers as well as associated chains. The binding of intramolecular and intermolecular hydrogen regions results in development of linear crystalline structure called cellulose.

2.2. Hemicellulose. Hemicellulose is a multidiverged polysaccharide made up of many distinctive glucose monomers, while cellulose is composed of only single 4-βD-glucopyranose repeating units. When compared to cellulose, the constituent of hemicellulose varies from one plant to other. The amorphous nature of the hemicellulose is confirmed by its high degree of chain grouping.

2.3. Lignin. Lignin is an intricate hydrocarbon polymer composed of aliphatic and aromatic elements. In addition to the cellulose, lignin is another important constituent in the lignocellulose fibril. The lignin binds the fibers together to make the fiber surface be stiffer enough by giving compression strength to the plant. The chemical composition of lignin is made of phenylpropane elements obtained from an enzyme-initiated dehydrogenate polymerization of three distinct constituents which are trans-p-coumaryl, trans-coniferyl, and trans-sinapyl.

2.4. Pectin. Pectin is an element of acidic polysaccharides with complex structure. The main constituents of pectin are homopolymeric acid and partial residues of methylated poly-α-(1-4)-D-galacturonic acid. When treated with alkali or ammonium hydroxide, pectin will become a water soluble polymer. The role of pectin is to function as cementing element among the plant fibrils which binds with other constituents to form stacks. Higher amount of pectin is present in the primary cell wall and the middle lamella of lignocellulosic fibril. During the process of retting, most of the pectin contents get removed from the natural fiber. Only after the removal of pectin does the natural fiber get qualified to be employed as reinforcement material with polymer matrices. Pectin is another component which makes the cellulose fibers get attached to all other constituents of the fiber. When compared to cellulose, lignin and pectin are the weaker amorphous polymers [40].

3. Effect of Various Chemical Treatments on the Mechanical Properties of Composites

Kobayashi et al. [41] discussed the mechanical properties of the hemp fiber-reinforced composite fabrication, and the hemp fiber was chemically treated to improve compatibility between fiber and the matrix. The authors found that the physical and mechanical properties of the natural and synthetic fibers were influenced by climate/natural/environmental changes. Hence, the surface of the hemp fiber was treated by chemical treatment process like acetyl, alkali, and silane. Alam et al. [42] explored the tensile strength of a new composites, combined with untreated kenaf, treated kenaf, jute fiber, and jute rope. They observed that the tensile strengths of kenaf and jute fiber are higher than that of jute rope. Similarly the water absorption properties of treated fiber were higher than those of untreated fibers. The stress transfer capacity of the fiber gets improved as a result of micro void exclusion and the fiber surfaces turn more uniform. The diameter of the fiber is also improved owing to the axial splitting of the fibrils [43]. Wang et al. [44] studied the feasibility of using coffee hull as reinforcement member with high density polyethylene matrix. Improvement in mechanical characteristics of the coffee hull polyethylene composites was compared against various chemical modifications done on the coffee hull powder. It was found that coffee hull powder subjected to calcium hydroxide treatment resulted in maximum strength of the composites. The fiber loading was found to increase up to 10 wt% above which the tensile strength decreases, whereas the flexural strength was found to be prominent for coffee hull treated with maleic anhydride grafted polypropylene. The moisture absorption property was found to be significant for the composite subjected to calcium hydroxide treatment. Therefore it was concluded that coffee hull powder could be a possible alternate for synthetic fiber. Rout et al. [45] conducted morphology analysis of palm tree leaf stalk fibers, where the SEM images confirmed the removal of wax, oil, and hemicellulose content from treated fibers. The cleaner surfaces besides pores were noticed in treated fibers compared to untreated fibers. The natural plant fibers have many advantages; there are also a few limitations which have to be studied. The major limitation of natural plant fibers is their hydrophilic nature which restricts the use of the fibers as reinforcement in PMC. The inappropriateness between the hydrophilic fibers and hydrophobic matrix results in swelling due to moisture absorption and it shows the poor interfacial bonding between matrix and the fibers [46–48]. Improvement was found in the interfacial bonding between the fiber and matrix due to the chemical treatments of fibers which also reduce the hydrophilicity, fiber surface cleanness, the moisture absorption, and improvement in the surface roughness [49]. Various natural fiber surface treatments like alkaline, silane, acetylation, and preimpregnation with polyethylene solution resulted in the enhancement of strength due to increase of interfacial bonding between fiber and matrix [50]. Venkatesha Gupta et al. [51] developed a new composite material which had the highest strength to weight ratio in comparison to existing composite materials. Sisal and hemp fibers were reinforced with epoxy matrix prepared using compression moulding method according to ASTM standard. For alkali treatment, NaOH (sodium hydroxide) was used and the amount of reinforcement was changed from 10% to 50% by weight. After the specimen was prepared, various mechanical properties were investigated and it was proved that the prepared specimen was better in terms of mechanical properties. Athipathi and Hegde Sowmitha Vijay [52] discussed the experimental evaluation based mechanical properties of coir and *Roystonea regia*-epoxy laminate with various fiber contents ratios. Orientation of the fiber was maintained as 0°, 45°, and 90°. From the results, three different points were observed. The untreated matrix-material based composite exhibits high tensile strength, high flexural strength, and high impact strength. The fibers were subjected to 30% NaOH solution treatment for 1 h. The mechanical and electrical properties of composites with treated fibers were compared with those of untreated fiber composites. The modification of plant based lignocellulose fibrils by sodium hydroxide is the most widely adopted technique to alter the cellulose molecular portion of the natural fiber. The coir/epoxy composites were used in the seat cushions, mirror casing, storage tank, post boxes, helmet casing, brushes, ropes, bags, brooms, door shutters, and building panels. The alkali treatment results in formation of amorphous region from the densely packed crystalline cellulose structure. The alkali-sensitive hydroxyl units existing in the natural fibers were removed, which further react with the water molecules. Thereby the moisture resistance property of the fiber improved. The alkali treatment also removes some portions of hemicellulose, lignin, pectin, wax, and other surface related impurities present in the natural fiber [53, 54]. Thereby the effective bonding between the fiber and the matrix is also enriched and the mechanical and thermal properties of the composites are improved. When the percentage of alkali treatment is increased, excess delignification of the natural fiber occurs, which leads to damage of fibers, and the mechanical properties of the fibers get reduced [55]. Alkalized lignocellulose fibers have reduced lignin content, partial removal of wax and oil covering substances happens, and disintegration of crystalline cellulose occurs.

More research articles have been published on the influence of mercerization on the mechanical and thermal properties of lignocellulose fiber-reinforced polymer matrix composites [56, 57]. Sathish et al. [53] explored the influence of mercerization of date palm fibers on the mechanical, thermal, and morphological properties. It was reported that 5% of NaOH concentration enhanced mechanical and thermal behaviour of the composites. When the percentage of sodium hydroxide was increased to 10%, deterioration on the properties was observed owing to the damage of fibers at increased concentration. The thermal resistance of the fiber was also improved due to the pulling out of waxy layers and various surface impurities present in the date palm fibers. Chen et al. [58] investigated the wettability and thermal stability of bamboo fibers exposed to alkali treatment. The percentage of alkali treatment was varied; it was found that surface roughness of the bamboo fiber was increased and found to be optimum at 15% of NaOH treatment. The thermal stability and wettability were also found to be promising for the similar alkali concentration. Reddy et al. [59] investigated the tensile and structural properties of borassus fruit fine fibers subjected to 5% of alkali treatment under different treatment time. The crystallinity of the fibers was analysed by X-ray diffraction technique. The removal of amorphous hemicellulose substance was witnessed by FTIR analysis. The concentration time of 8 hours resulted in optimum fiber properties. It was also found that the borassus fibers will be a suitable reinforcement for the manufacturing of green composites. Balaji and Nagarajan [60] investigated the tensile and chemical behaviour of cellulose fibers extracted from saharan *Aloe vera* cactus leaves exposed to mercerization treatment. The fibers exposed to mercerization resulted in removal of hemicellulose, lignin, wax, and other surface related impurities present in the fibers. It was also found that the hydrophobic nature of the lignocellulose fibrils is lowered and interfacial adherence among the fibrils and the matrix enhanced. Increase in thermal stability of the fibrils was witnessed by TG analysis. The SEM analysis also confirmed the removal of hemicellulose, lignin, wax, and other layers present in the fibers. Finally, it was found that natural fiber extracted from saharan *Aloe vera* cactus leaves was found to be a suitable alternate to synthetic fibers for reinforcement with polymer matrix. Dawit et al. [61] explored the property of *Acacia tortilis* fibrils extracted from the barks of *Acacia tortilis* tree. The extraction of fiber was based on natural water based retting. The extracted fibers were subjected to mercerization treatment with 10% and 20% of NaOH solution. The alkalization of the fiber resulted in removal of hemicellulose, lignin, wax, and other surface related impurities present in the fiber. As the percentage of alkali treatment is increased to 20%, decrease in tensile strength of the fibrils was noted. This phenomenon could be explained as follows: when the concentration of alkali solution exceeds the limit, the diameter of the fiber gets reduced even further, which results in reduced tensile strength. It was also concluded that *Acacia tortilis* fiber could be a viable alternate for manmade fiber in polymer composite applications. Narayanasamy et al. [62] explored the possibility of lignocellulose fibril extracted from *Calotropis gigantea* fruit bunch as a possible alternate for artificial fiber-reinforced polymer composites. The fibers were extracted from the fruits of *Calotropis gigantea* fruit bunch through retting process. Then the extracted fibers were mercerized with 5% of NaOH. XRD and FTIR analysis revealed the removal of hemicellulose, lignin, wax, and other surface related impurities existing in the fibers. The thermal stability of the fibrils was also enriched by the influence of alkali treatment, which is inferred by TG analysis. Finally SEM analysis revealed that the surface of fibers was rougher due to alkali treatment. Negawo et al. [63] researched the effect of alkali modification on the *Ensete* stem fibrils obtained from the Ethiopian *Ensete ventricosum* plant. The fibers were subjected to 2.5%, 5%, and 7.5% of mercerization. The mercerization resulted in removal of lignin, wax, and hemicellulose existing in the fibrils. The 5% mercerized fibers exhibited better properties when compared with untreated fibrous composites owing to the enhanced interfacial adherence between the fiber and the matrix. The 5% alkalized fibers exhibited better mechanical properties under static and dynamic conditions. From the experimentation, it was also concluded that *Ensete* stem fiber could be a possible alternate to synthetic fiber-reinforced polymer composites for wide assortment of applications. Reddy et al. [64] studied the influence of NaOH and KOH fiber surface modifications on the mechanical properties of Tapsi fiber-reinforced epoxy composites manufactured by hand layup technique. Initially the fibrils were pretreated with 5% of concentrations for two hours. Composites were manufactured by varying the fiber weight fraction. Tensile and flexural test performed on the composite samples revealed that composites with 15% of fibers exhibited higher properties for NaOH-treated fibers when compared to KOH-treated fibers. FTIR analysis revealed the removal of functional groups present in the fibers and XRD analysis revealed the improvement of crystallinity index and size for NaOH-treated fiber-reinforced composite samples when compared to KOH-treated sample. SEM analysis revealed fiber pull-out as a result of improper fibril wetting, which resulted in poor adherence between the interfaces of fiber and matrix in the composites. Senthilkuamr et al. [65] reviewed the mechanical properties of sisal fiber-reinforced polymer composites. The sisal fibers have been extracted by the process of decortication and the extracted sisal fibers were exposed to chemical treatment like alkalization and coupling agents. The pretreated sisal fiber showed improvement in mechanical, hydrophilic tendency resulting in effective bonding between the interfaces of fiber and polymer matrix. The mechanical properties of the developed composites depend on different characteristics like fiber length, fiber orientation, fiber volume fraction, and several other parameters. Appreciable enhancement in mechanical properties was found owing to the chemical modifications on the surface of sisal fibers. Overall, it was concluded that the enrichment in properties of the sisal fiber-reinforced composites depends on surface treatment concentration type and time; beyond the concentration level, deterioration in properties of the fibers was noticed. It was also concluded that impact strength of the sisal fiber composites decreased as a result of chemical modifications done on the sisal fibers. Balaji et al. [66] explored the influence of fiber content on the mechanical properties of the alkali-treated bagasse fiber reinforced with cashew nut shell liquid. Initially the fibers were chopped to 10 and 20 mm and the composites were prepared by compression moulding technique by varying the fiber volume fraction as 0, 5, 10, 15, and 20 wt%. The tensile and flexural test revealed that maximum strength was attained for 15 wt% of fiber-reinforced composites. FTIR analysis revealed the removal of functional groups present in the fibers owing to the alkali treatment of sugarcane bagasse fiber. The thermal stability of the fibers was also enhanced due to the mercerization and SEM analysis revealed the enhanced interfacial adherence between the fiber and the matrix which resulted in the homogeneous nature of composite. Komal et al. [67] investigated the prominence of alkalization done on the surface of banana fibers. The surface modified banana fibers were reinforced with polypropylene and then the tensile, flexural, and impact strength and degradation behaviour were studied for untreated and surface modified banana fiber-reinforced polypropylene composites. Thermogravimetric analysis revealed removal of hemicellulose, lignin, pectin, wax, and other surface related impurities present in the banana fiber as a result of mercerization. Significant enhancement in tensile and flexural strength was found between untreated and pretreated banana fiber-reinforced polypropylene composites. Improvement of impact strength by 11.5% was observed for untreated and mercerized banana fiber-reinforced polypropylene composites. The tested composite samples were subjected to morphology analysis by scanning electron microscope to study the fracture behaviour of the composite samples. Fiber pull-outs were observed in untreated banana fiber-reinforced polypropylene composite; as a result, tensile and flexural strength of the composites decreased in contrast with alkali-treated and untreated banana fiber-reinforced polypropylene composite. Marginal reduction in weight loss of the samples was also observed. Alkali-treated banana fiber-reinforced polypropylene composites absorbed less water as the hydrophilic tendency of the fiber was altered by alkali treatment, whereas the untreated banana fiber-reinforced polypropylene composites absorbed water due to higher hydrophilic tendency. Ameer et al. [68] explored the mechanical and moisture characteristics of hydrophilic modified jute fiber-reinforced unsaturated polyester composites. The extracted jute fiber was exposed to mercerization treatment to improve the hydrophilic tendency. Better interlocking in the middle of fiber and matrix was attained by the removal of amorphous substances present in the jute fibers. The mercerization treatment resulted in significant reduction in hydrophobic nature of the jute fiber. The mercerized jute composites showed improved mechanical properties in contrast with untreated jute composites. Chin et al. [69] investigated the mechanical and thermal characteristics of bamboo fiber-reinforced composites, where the bamboo fibers were exposed to mercerization with different concentrations over varying time. The effect of alkali treatment was inferred with FTIR and XRD analysis. Enhancement in crystal size and crystallinity index was observed by XRD analysis and removal of lignin, cellulose, and other surface related impurities was observed by FTIR analysis. The thermal stability of the composites was also enhanced, which was evident from the thermogravimetric analysis. The composites with 40 wt% contributed to maximized tensile and flexural properties of the composites. Balaji et al. [66] explored the mechanical properties of sugarcane bagasse fiber-reinforced cardanol composite. Mercerization of fiber resulted in removal of amorphous substances and improved interlocking between the bagasse fiber and cardanol matrix. The composites with 15 wt% of fiber exhibited better properties. Senthamaraikannan et al. [70] explored the possibility of using *Acacia planifrons* fibers as possible reinforcement material with polymer matrices. The fibers were extracted by the process of retting. The extracted fibers were subjected to mercerization with varying percentage of sodium hydroxide. It was found that the mercerization leads to improvement of crystallinity index and thermal stability and removal of amorphous substances present in the fibers. The optimal alkali treatment is found to be 5%. Tables 1–10 present the effects of various chemical treatments of biofibers.

Mouhoubi et al. [174] reported the SEM images of alfa fiber with alkali treatment (5% NaOH) at different time intervals (2 h, 4 h, 6 h, and 24 h). Figures 1(a)–1(e) represent the alfa fiber. Figure 1(b) shows the fiber treated with 5% NaOH at 2 h. During this time period, the waxy substances in the fiber were removed. Figures 1(c)–1(e) show that the fiber resulted in low moisture absorption, removal of extractives, and increase in crystallinity and stiffness.

Table 1: Effect of alkaline treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Kenaf	Alkaline treatment	At 6% concentration of NaOH it has good effect on kenaf fiber resulting in removal of all impurities from the surface.	[71]
2	Bamboo, kenaf, hemp, sisal, jute, and kapok	Alkaline treatment	The treatment removed the noncellulose constituent in fibers such as lignin, wax, and oils, promoted ionization of hydroxyl group of cellulose to alkoxide, and reduced the hydroxyl group content. The treatment improved the surface roughness and hydrophobicity resulting in good adhesion.	[72]
3	Pineapple leaf	Alkaline and acetic	Improvements in tensile strength, impact strength, and flexural strength.	[73]
4	Abaca	Alkaline and silane treatment	The silane-treated fiber has higher thermal transfer coefficient.	[73]
5	Bamboo	Alkaline treatment	An enhancement in tensile strength by adding 30% treated bamboo which is slightly higher than silane-treated composite.	[74]
6	Sisal	Alkaline treatment	Improvement in interfacial shear strength.	[75]
7	Sisal/hemp	Alkaline treatment	At 10% concentration of NaOH, enhanced the flexural strength by adding 40 wt% sisal and hemp.	[75]
8	Curaua	Alkaline treatment	An increase in NaOH concentration and decrease of fiber diameter, fiber density, and fiber weight.	[76]
9	Ramie	Alkaline treatment	Alkali treatment possesses better tensile strength than silane-treated fiber composite.	[76]
10	Hemp	Alkaline treatment	The treated fiber has high crystallinity resulting in improvement in tensile strength and Young's modulus.	[76]
11	Jute	Alkaline treatment	The treatment removed hemicellulose, pectin, and lignin resulting in decreased fiber diameter.	[76]
12	Basalt	Alkaline treatment	The NaOH-treated fiber has superior properties compared to glass fiber.	[76]
13	Banana	Alkaline treatment	5% NaOH-treated fiber has better properties.	[77]
14	Luffa/coir	Alkaline treatment	Improvement in tensile and flexural strength and hardness.	[78]
15	Luffa/groundnut fiber	Alkaline treatment	An increment in mechanical properties by removal of hemicellulose, wax, lignin, and impurities from the fibers, thus increasing the adhesive characteristics of composite.	[79]
16	Abaca	Alkaline treatment	Improvement in moisture resistance.	[80]
17	Alfa	Alkaline treatment	At 10% of NaOH content, increases in flexural strength and flexural modulus by 60% and 62%, respectively, and fiber becomes stiffer and brittle.	[81]
18	Drumstick (*Moringa oleifera*)	Alkaline treatment	The addition of glass fiber increased impact strength and frictional coefficient.	[82]
19	Ladies finger	Alkaline treatment	Double-stage chemical treatment possessed better properties than single-stage treatment, while an increase in span length decreased the tensile strength and increased Young's modulus.	[83]
20	Tamarind	Alkaline treatment	Chemically treated 2 cm fiber length was optimum to achieve better hardness, impact, and frictional coefficient.	[84]
21	Vetiveria zizanioides/jute	Alkaline treatment	The treated fibers improved tensile strength, flexural strength, and impact strength by 26.8%, 30.44%, and 59.1%, respectively.	[85]
22	Borassus	Alkaline treatment	At 5% of NaOH content, significantly increased tensile properties.	[86]
23	Palm wood	Alkaline treatment	The optimum residual mass at 0% to 0.75% NaOH. With further 1% NaOH it decreased.	[87]
24	Palmyra palm leaf stalk fiber (PPLSF)/jute	Alkaline treatment	The alkali-treated PPLSF has maximum tensile and flexural properties by the addition of alkali-treated jute fiber.	[88]
25	*Roystonea regia*	Alkaline treatment	Improvement in tensile and flexural properties.	[88]
26	*Borassus flabellifer* (Asian palmyra)	Alkaline treatment	Improvement in tensile strength.	[88]
27	Buriti and ramie	Alkaline treatment	At 2% NaOH treatment of ramie fiber, increased flexural strength by 70%. However, alkali treatment was only favorable for buriti fibers.	[89]

Table 1: Continued.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
28	Rice husk	Alkaline treatment	An increase in cellulose content, resulting in increased crystallinity index. Therefore diameter decreased from 170 to 7 mm, as well as further diameter value from 10 to 15 nm by performing acid hydrolysis treatment.	[90]
29	Rice husk	Alkaline treatment	Improvement in adhesion characteristics.	[90]
30	Jute	Alkaline treatment	An increase in flexural strength, modulus, and interlaminar shear strength.	[91]
31	Coir	Alkaline treatment	At 5% alkali treatment increases in impact and flexural strength for 72 h by 40%.	[92]
32	Jute	Alkaline treatment	At 5% alkali treatment increases in flexural strength for 4 h by 20%.	[93]
33	Banana	Alkaline treatment	At 1% alkali treatment enhanced flexural strength, flexural modulus, tensile strength, and tensile modulus by 20, 12, 132, and 131%, respectively.	[94]
34	Ramie	Alkaline treatment	At 9% alkali treatment enhanced tensile strength for 1 h by 23%.	[95]
35	Jute	Alkaline treatment	An increase in flexural strength, flexural modulus, and interlaminar shear strength by 35%, 23%, and 19%, respectively.	[93]
36	Abaca/roselle	Alkaline treatment	The treatment increased fiber/matrix adhesion property due to removal of hemicellulose, waxes, lignin, and impurities from the fibers.	[96]
37	Jute	Alkaline treatment	The treatment removed the hemicellulose and promoted the interlocking points in the fiber for better adhesion and stress transfer across the interface resulting in increased tensile strength, flexural strength, flexural modulus, and interlaminar shear strength.	[97]
38	Jute	Alkaline treatment	The treatment increased the cellulose content after removal of pectin, lignin, and other impurities. An increase in cellulose content leads to better interfacial adhesion.	[98]
39	Sisal	Alkaline treatment	The treatment had better mechanical properties due to good adhesion between fiber and matrix.	[99]
40	Oil palm	Alkaline treatment	A bigger increase in flexural strength by performing 24-hour NaOH treatments compared to other chemical treatments.	[100]
41	Jute	Alkaline treatment	At 25% fiber loading and 10% NaOH treatment showed increase in tensile strength due to decrease in fiber diameter and density.	[101]
42	Jute	Alkaline treatment	At 20% fiber loading and 10% NaOH treatment showed increase in tensile strength due to decrease in fiber diameter and density.	[102]
43	Napier grass	Alkaline treatment	The 12 h soaking time of treated fiber had least fiber diameter and mass. The 6 h soaking time exhibited highest tensile strength. An increase in surface roughness with the increase in soaking time beyond 18 h. However, 24 h-treated fiber had damage on its surface.	[103]
44	Henequen	Alkaline treatment	The treated fiber had higher adsorption rate at 100 h to attain adsorption equilibrium.	[50]
45	Sisal	Alkaline treatment	The 45 min of treatment yielded more level of crystallinity with more cell wall structure. Tensile and shear strength were increased by 12.04% and 173%, respectively.	[104]
46	Sisal	Alkaline treatment	An increase in crystallinity decreased the absorption rate. Optimum fiber length 5.8–9 cm displays better performance in tensile strength with increase in fiber loading.	[105]
47	Ladies finger	Alkaline treatment	Removal of hydrophilic hemicellulose led to enhanced surface roughness.	[83]
48	Kenaf	Alkaline treatment	Chemically treated 6% NaOH sample was optimum to achieve better tensile strength and modulus of elasticity.	[106]

Table 1: Continued.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
49	Kenaf	Alkaline treatment	At 9% NaOH alkali treatment displayed cleanest surface although tensile strength decreased. However, 6% NaOH alkali treatment with higher temperature was optimum in cleaning fiber.	[49]
50	Banana	Alkaline treatment	An enhancement in tensile modulus and impact and tensile strength by adding 3 wt% of fiber.	[107]
51	Banana	Alkaline treatment	At 10% of NaOH content, significantly increased thermal conductivity.	[108]
52	Banana	Alkaline treatment	At 4% concentration of NaOH, enhanced the tensile strength, tensile modulus, and flexural strength.	[109]
53	Banana	Alkaline treatment	Alkali treatment possesses better tensile strength and flexural strength when compared with benzoylation and PSMA treatment.	[110]
54	Banana	Alkaline treatment	The treatment decreased modulus of rigidity, tensile strength, and strain due to degradation of cellulose.	[111]
55	Pineapple leaf	Alkaline treatment	An increase in fiber density, cellulose, and crystallinity led to enhanced tensile strength, thermal stability, and water retention with increasing the NaOH up to 7% concentration.	[112]
56	Pineapple leaf	Alkaline treatment	The treated fiber significantly improved the flexural strength, impact strength, storage modulus, and thermal resistance by 79%. Heat deflection temperature (171.3°C) which is close to the melting temperature of neat polymer. Reduction in crystallization by 14°C.	[113]
57	Pineapple leaf	Alkaline treatment	An enhancement in Young's modulus by 30% compared to untreated fiber.	[114]
58	WSF	Alkaline treatment	An enhancement in thermal stability by adding 3% NaOH.	[115]
59	Banana	Alkaline treatment	At 1% NaOH treatment possess better properties.	[94]
60	Hemp	5% NaOH, 0.5% silane	The combined NaOH and silane treatment increased the tensile and flexural strength by 100% and 45%, respectively. But fracture toughness decreased.	[116]
61	Jute	Alkaline treatment	At 4% NaOH treatment increased tensile strength up to 30%.	[117]
62	Agave	Alkaline treatment	Increased the fiber matrix adhesion and fracture strain.	[118]
63	Palm leaf stalk/jute	Alkaline treatment	An increase in storage modulus and loss modulus by addition of jute fiber.	[88]
64	Coir	Alkaline treatment	Enhancement in mechanical properties, moisture resistance, and adhesion properties.	[119]
65	Flax	Benzoylation, peroxide, mercerization, silane treatment.	The treatment exhibited improved mechanical and physical properties.	[120]
66	Hemp/jute	Alkaline treatment	Increase in crystallinity can enhance the fiber strength.	[121–123]
67	Hemp	Alkaline treatment	An increase in crystallinity of PLA matrix due to crystalline cellulose in the alkaline-treated hemp fibers, which acts nucleating sites resulting in increase in fiber strength.	[124]
68	Kenaf/hemp	Alkaline treatment	The treated fiber found to have better mechanical properties, thermal stability, and moisture resistance.	[125, 126]
69	Sisal	Combined NaOH + actylation	Increase in mechanical properties due to better adhesion between fiber and matrix.	[104]
70	*Tridax procumbens*	Alkaline treatment	At 5% concentration of NaOH, enhanced the wettability and crystallinity and reduced amorphous region and fiber diameter.	[127]

Figure 2 represents the SEM images of untreated and treated *Prosopis juliflora* fiber-reinforced epoxy composites at different concentrations (5%, 10%, and 15%). Figure 2(a) shows the untreated fiber surface which consists of impurities and fiber pull-outs on the surface. This was due to the waxy substances present on the surface of the fiber and the existence of hydroxyl groups, which leads to water absorption, weakening interfacial strength with the matrix. Figures 2(b)–2(d) represent the treated *Prosopis juliflora* fiber-reinforced epoxy composites at concentrations of 5%, 10%, and 15%, and increase in alkali treatment beyond 5% damaged the fiber surface and reduced the cellulose content in the fiber, which in turn resulted in lower strength and stiffness [175].

Liu et al. [176] researched the supremacy of silane coupling agent treatment done on the surface of corn stalk

Table 2: Effect of silane treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Kenaf	Silane treatment	The presence of lignin and hemicellulose was removed by performing silane treatment. Removal of lignin and hemicellulose led to enhanced interfacial bonding.	[71]
2	Pineapple leaf	Silane treatment	The treated fiber has fewer voids on the interface which makes strong interfacial bonding and results in better mechanical properties.	[71]
3	Abaca	Mercerization and silane treatment	The silane-treated fiber has higher thermal transfer coefficient.	[73]
4	Bamboo	Silane treatment	An enhancement in tensile strength by incorporation of 30% treated bamboo, while flexural strength is higher than that of NaOH-treated fiber.	[74]
5	Sisal	Silane treatment	The treatment enhances the mechanical properties and moisture resistance.	[75]
6	Hemp/ kenaf	Silane treatment	The treatment possesses higher flexural modulus in comparison with alkali-treated composite and similar to glass fiber composite.	[128]
7	Hemp	Silane treatment	Flexural and tensile strength were increased by 2% and 4%, respectively.	[129]
8	Kenaf	Silane treatment	An enhancement in storage modulus and viscoelasticity by 45% and 25%, respectively.	[130]
9	Oil palm	Silane treatment	Reduced the mechanical properties due to poor adhesion between fiber and matrix.	[131]
10	Henequen	Silane treatment	An enhancement in tensile strength from 21 MPa to 27 MPa by performing combination of silane and NaOH.	[50]
11	Sisal	Silane treatment	The treated fiber had higher impact strength compared to alkali-treated fibers.	[132]
12	Banana	Silane treatment	An increase in flexural strength about 160% and considerable increase in tensile and toughness.	[133]
13	Banana	Silane treatment	An enhancement in impact and tensile strength by 30.84% and 19.43%, respectively, and slight increase in tensile modulus.	[134]
14	Jute	Silane treatment	An increase in strength and modulus about 12% and 7% by alkali treatment followed by silane treatment.	[135]
15	Jute	Silane treatment	At 0.3%, silane-treated composites enhanced the tensile, flexural, and interlaminar shear strength by 40%, 30%, and 55%, respectively.	[113]
16	Pineapple leaf	Silane treatment	Improvement in flexural modulus and storage modulus by 47% as compared to alkali treatment.	[114]
17	Pineapple leaf	Silane treatment	The resulting composite has less Young's modulus than alkali-treated composites.	[136]
18	Pineapple leaf	Silane treatment	Reduction of hydrophilic tendency of the fibers leads to increase in tensile strength and crystallinity size but % crystallinity decreases.	[72]
19	Hemp	Silane treatment	Found maximum mechanical properties compared to other chemical treatments.	[137–139]

Table 3: Effect of acetylation treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Coir/oil palm	Acetylation treatment	A bigger increase in tensile strength, flexural strength, Young's modulus, and impact strength by performing acetylation treatment compared to silane treatment.	[140]
2	Flax	Acetylation treatment	An enhancement in tensile and flexural strength by 35%.	[141]
3	Abaca	Acetylation treatment	The treatment possessed higher tensile strength, Young's modulus, and impact strength by 81, 70, and 8%, respectively.	[133]
4	Oil palm	Acetylation treatment	The treatment has high strain value which resulted in enhanced elastic and impact property.	[100]
5	Green flax	Acetylation treatment	At 65% of relative humidity, decrease in the moisture absorption. An increase in thermal stability with increase in degree of acetylation. About 25% improvement in strength properties was observed compared to untreated composites.	[139]
6	Flax	Acetylation treatment	At 18% of acetylation, concentration of flax fiber exhibited better tensile strength and thermal stability by 25% and 50%, respectively. However, the addition of maleic anhydride resulted in increase in mechanical properties by 20–35%.	[139]

Table 3: Continued.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
7	Sisal	Acetylation treatment	Improvement in tensile strength and shear strength by 14.08% and 435%, respectively. The acetic acid treatment followed by ethyl acetate with H_2SO_4 resulted in high levels of cellulose swelling or loosened cell wall structure.	[104]
8	Sisal	Acetylation treatment	A decrease in dielectric constant with increasing frequency.	[105]
9	Banana	Acetylation treatment	The treated fiber has high fibrillation and is more rougher resulting in better tensile properties than mercerization treatment.	[109]
10	*Grewia serrulata* bast	Acetylation treatment	The treated fiber has better dimensional stability and more moisture resistance. However, high degree of ultraviolent energy can degrade the composite.	[142]
11	Phosphate bonded composite	Acetylation treatment	The treated fiber reduces water absorption and hence improves dimensional stability, tensile strength, and stiffness. However, this treatment reduces the impact strength as compared to other chemical treatments.	[143]

Table 4: Effect of permanganate ($KMnO_4$) treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Sisal	Permanganate treatment	Improvement in tensile strength.	[75]
2	Oil palm	Permanganate treatment	The treated fiber has highly fibrillated structure and hence very good fiber matrix adhesion. As a result, better tensile strength and modulus were observed	[100]
3	Sisal	Permanganate treatment	At 1% concentration of $KMnO_4$ polar groups between fiber and matrix are formed leading to degradation of cellulose. The hydrophilic tendency of fiber decreases as the $KMnO_4$ concentration increases up to an optimum.	[105]
4	Sisal	Permanganate treatment	At 1% concentration, higher degradation of cellulose occurred due to formation of polar group. Optimum properties were found to be better at 0.055% concentration. Tensile properties were observed between alkali and peroxide.	[144]
5	Sisal	Permanganate treatment	Improvement in interlaminar shear strength, tensile strength, and flexural properties compared to silane. But impact properties were lower than those of untreated fiber.	[145]
6	Banana	Permanganate treatment	An increase in thermal diffusivity, tensile strength, and tensile modulus by 16%, 6.4%, and 7.5%, respectively. However, flexural strength and modulus were found to have increases of 6% and 10%, respectively, which were lower compared to alkali and silane treatment.	[108]
7	Flax	Permanganate treatment	Improvement in tensile and moisture resistance as compared to alkali and silane-treated fibers.	[108]
8	Banana	Permanganate treatment	An increase in tensile strength and flexural strength by 5% and 10%, respectively. Increases in polarity and roughness of fiber were also observed.	[146]

Table 5: Effect of peroxide treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Sisal	Peroxide treatment	Enhancement in tensile properties.	[72]
2	Sisal	Peroxide treatment	Crystallinity index and decomposition rate were found to be better at soaking time of 30 min.	[146]
3	Kenaf	Peroxide treatment	At 30% fiber loading exhibited higher tensile and flexural strength, whereas modulus was high at 40% fiber loading.	[146]
4	Oil palm	Peroxide treatment	Improvement in flexural modulus as compared to other chemical treatments.	[147]
5	Sisal	Peroxide treatment	50% higher tensile strength of treated fiber compared to untreated fiber composites.	[105]
6	Jute	Peroxide treatment	The treatment exhibited better tensile and flexural properties than alkali and permanganate treatment, but not as superior as silane treatment, whereas the thermal stability was reduced.	[105]
7	Pineapple leaf	Peroxide treatment	Better in tensile strength, tensile modulus, and abrasion resistance as compared to untreated composites. But there was a reduction in elongation breaks.	[148]
8	Wheat straw	Peroxide treatment	The treated composites were found to have increase in ash content. Removal of lignin was around 50%.	[149]

Table 6: Effect of benzoylation treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Sisal	Benzoylation treatment	Improvement in tensile strength by 91%.	[75]
2	Jute	Benzoylation treatment	Improvement in storage modulus and thermal stability.	[150]
3	Flax	Benzoylation treatment	The treated composites were found to have highest tensile and impact strength for LDPE and highest impact strength for HDPE. Resulted in less water absorption as compared to silane and peroxide. Smooth fiber surface was observed.	[43]
4	Sisal	Benzoylation treatment	At 6% of benzoyl peroxide showed better mechanical properties.	[83]
5	Banana	Benzoylation treatment	The treatment significantly improved thermal conductivity and was found to have increase in tensile strength and modulus by 13% and 5%, respectively, although not as good as alkali and silane-treated fiber.	[109]
6	Sisal	Benzoylation treatment	The treatment increased the activation energy for glass transition temperature (Tg). Maximum activation energy was observed.	[151]

Table 7: Effect of acrylation treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Bagasse	Acrylation treatment	Acrylation treatment offers superior tensile and flexural strength compared to alkali treatment.	[152]
2	Oil palm	Acrylation treatment	The treatment yielded higher extension and impact resistance. At 40% of fiber with 50°C exhibited moderate level of moisture absorption compared to other treatments.	[102, 155]
3	Flax	Acrylation treatment	The treated fiber exhibited higher tensile strength and moisture resistance than those treated with silane, permanganate, and sodium chloride treatment. Higher smooth fiber surface was observed.	[131]
4	Flax	Acrylation treatment	At higher concentration, grafting was increased due to higher availability of monomer molecules in cellulose radicals as well as polymerization medium.	[154]
5	Jute	Acrylation treatment	An increase in tensile strength and flexural strength by 42.2% and 13.9%, respectively.	[155]

Table 8: Effect of acrylonitrile grafting treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Pineapple leaf	Acrylonitrile grafting	Improvement in tensile strength by adding AN (acrylonitrile grafting). It has major effect on tensile strength.	[75, 102]
2	Oil palm	Acrylonitrile grafting	The treatment showed high strain rate and elastic modulus. But slight improvement in stiffness was absorbed. However at 40 wt% of fiber with 50°C exhibited higher moisture resistance compared to other treatments.	[153]
3	Agave Americana fibers	Acrylonitrile grafting	An increase in percentage of graft and decrease in the moisture resistance, in addition to improvement in thermal stability of fiber.	[156]
4	Sisal	Acrylonitrile grafting	The treatment showed enhanced tensile and flexural strength as compared to other treatments. Least degradability of fibers was observed.	[157]
5	Pineapple leaf	Acrylonitrile grafting	The treatment possesses lower grafting yield than unmodified fibers.	[158]
6	Cellulose polymer	Graft copolymerization	Improvement in physical, chemical, and thermal resistance.	[159]

fibers extracted from the waste of corn stalk. Initially the fibers were modified with different percentages of silane concentration such as 1%, 5%, 9%, and 13%. The effect of silane treatment on corn stalk fibers was investigated by FTIR and XRD analysis. Results showed improvement in the crystalline size for 5% silane-treated fiber and also removal of hemicellulose, lignin, and other impurities present in the fibers was found by FTIR analysis. The impact behaviour of the composites was also found to be superior for 5% silane-treated corn stalk fiber-reinforced composites. Finally SEM analysis revealed that the surface of fibers was rough in contrast with untreated fiber owing to the effect of silane treatment. Liu et al. [177] investigated the effect of silane coupling agent on the mechanical, tribological, and morphological characteristics of corn stalk fiber-reinforced polymer composites. The extracted corn stalk fibers were

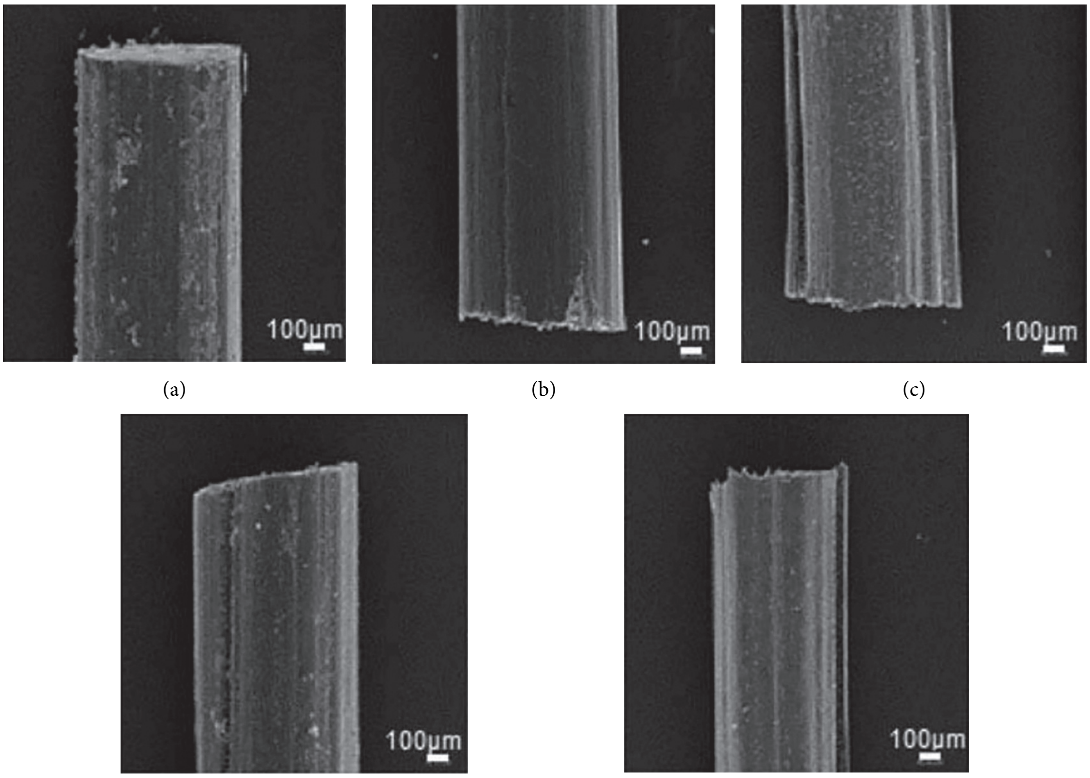
Table 9: Effect of isocyanate treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Oil palm	Isocyanate treatment	At 40% fiber loading exhibited higher moisture absorption (280%) compared to other treatments.	[153]
2	Sisal	Isocyanate treatment	The treatment exhibited superior tensile properties than alkaline and untreated fibers but with decrease in dielectric constant.	[107, 146]
3	Pineapple leaf	Isocyanate treatment	The treatment reduced hydrophilic tendency of the fiber compared to silane-treated composites. Reduction in % of crystallinity leads to increase in tensile strength.	[136]
4	Jute/hemp/flax	Isocyanate treatment	The treatment increased stiffness and reduction in impact by 17%.	[31]
5	Fibrous cellulose	Isocyanate treatment	The treatment shows enhanced tensile, elongation, and interfacial adhesion.	[160]
6	Kenaf	Isocyanate treatment	The presence of isocyanate hydrolysis to urea, reacting with hydroxyl group of fiber, decreases the moisture absorption and increases the mechanical properties.	[72]
7	Sisal	Isocyanate treatment	Improvement in tensile strength	[161]

Table 10: Effect of maleic anhydride grafted treatment on various biofibers.

S. no.	Fibers used	Type of chemical treatment	Effects	Ref.
1	Jute	Maleic anhydride grafted	Coupling agent has greater effect on Young's modulus and dynamic storage modulus.	[162]
2	Flax and hemp	Maleic anhydride grafted	The treatment improved dynamic and mechanical properties.	[163]
3	WSF	Maleic anhydride grafted	The treatment significantly reduced crystallinity and thermal stability is higher than that in acetylation treatment.	[164]
4	Wood flour	Maleic anhydride grafted	An enhancement in tensile strength and modulus properties became twice as compared with untreated fibers.	[165]
5	Sisal	Maleic anhydride grafted	Improvement in tensile, flexural, and impact strength by 50%, 30%, and 58%, respectively. Reduction in water absorption by 61%. Higher level of crystallinity was also observed.	[166]
6	Banana/hemp/sisal	Maleic anhydride grafted	Reduction in moisture resistance compared to untreated fiber. At 50% fiber loading, increases in tensile, flexural, and impact strength. Flexural modulus values were higher than those of untreated fiber.	[167]
7	*Hildegardia*	Maleic anhydride grafted	An increase in tensile properties with addition of compatibilizers.	[168]
8	Pineapple leaf	Maleic anhydride grafted	The treated fiber exhibited increased aspect ratio and matrix resulting in increased tensile strength, impact strength, and flexural strength by 9%, 30%, and 3%, respectively, compared to untreated fibers	[169]
9	Jute/sisal	Maleic anhydride grafted maleated HDPE	At 1% concentration of coupling agent increased the dynamic (storage modulus and loss modulus) and static (tensile, flexural, and impact) mechanical properties.	[170, 171]
10	Wood flour	Maleated polypropylene	An increase in dimensional stability and strengthening by MAPP addition.	[172]
11	Natural fibers	Maleated coupling agents	An increase in interfacial adhesion between fiber and matrix due to removal of hydroxyl group by addition of coupling agent.	[173]

exposed to surface modifications with silane coupling agents. Results showed an enhancement in water absorption and porosity of the silane-treated corn stalk fiber-reinforced composite specimens. Improvement in wear behaviour was noticed between the untreated and silane-treated corn stalk fiber-reinforced polymer composites. SEM analysis revealed the formation of secondary plateaus on the composite specimens which leads to the reduced wear rate on the composite samples. Jappes and Siva [178] researched the influence of silane modification done after the mercerization treatment on coconut sheath fiber to improvise the mechanical properties of the composites. The fabricated coconut sheath polyester composites were taken for testing of tensile, flexural, and impact strength, and the properties were compared with fabricated glass fiber-reinforced polyester composites. The coconut sheath fiber-reinforced composites showed better mechanical properties against glass fiber-reinforced polyester composites. The alkali and silane modification done on the surface of coconut sheath improved the hydrophilic nature of the fiber which ensured enhanced adhesion between the coconut sheath fibril and the matrix. SEM analysis was done to study the result of alkalization on the coconut sheath fiber; dismissal of waxy layers and other surface related impurities to make the fiber



(a) (b) (c)

(d) (e)

Figure 1: 5% NaOH-treated alfa fiber at different time intervals: (a) raw fiber, (b) 2 h, (c) 4 h, (d) 6 h, and (e) 24 h [174].

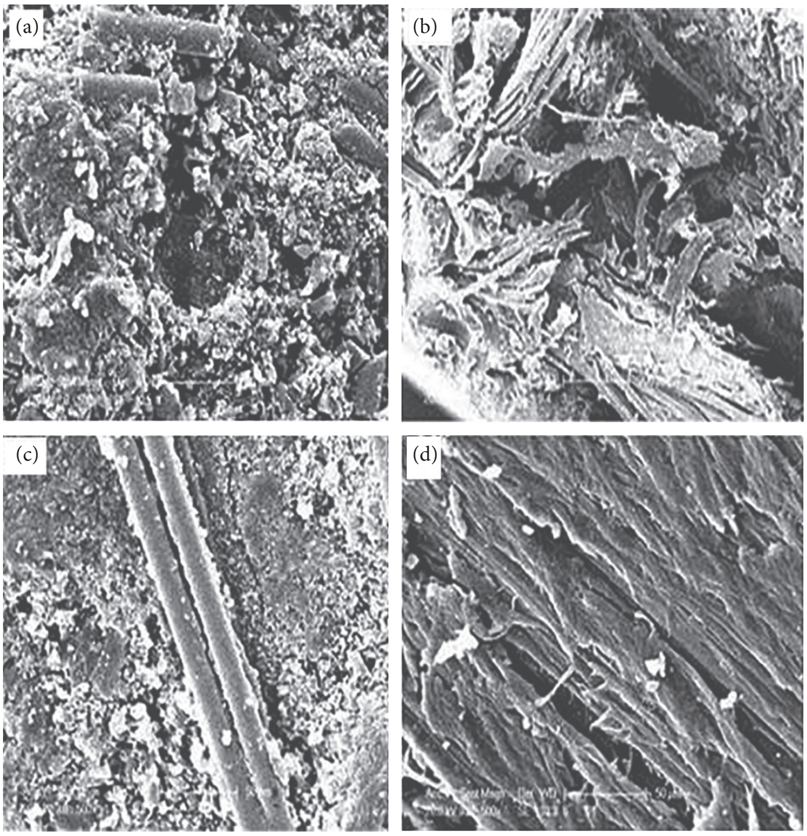


Figure 2: SEM images. (a) Untreated *Prosopis juliflora* fiber. (b) 15% NaOH-treated fiber. (c) 10% NaOH-treated fiber. (d) 5% NaOH-treated fiber [175].

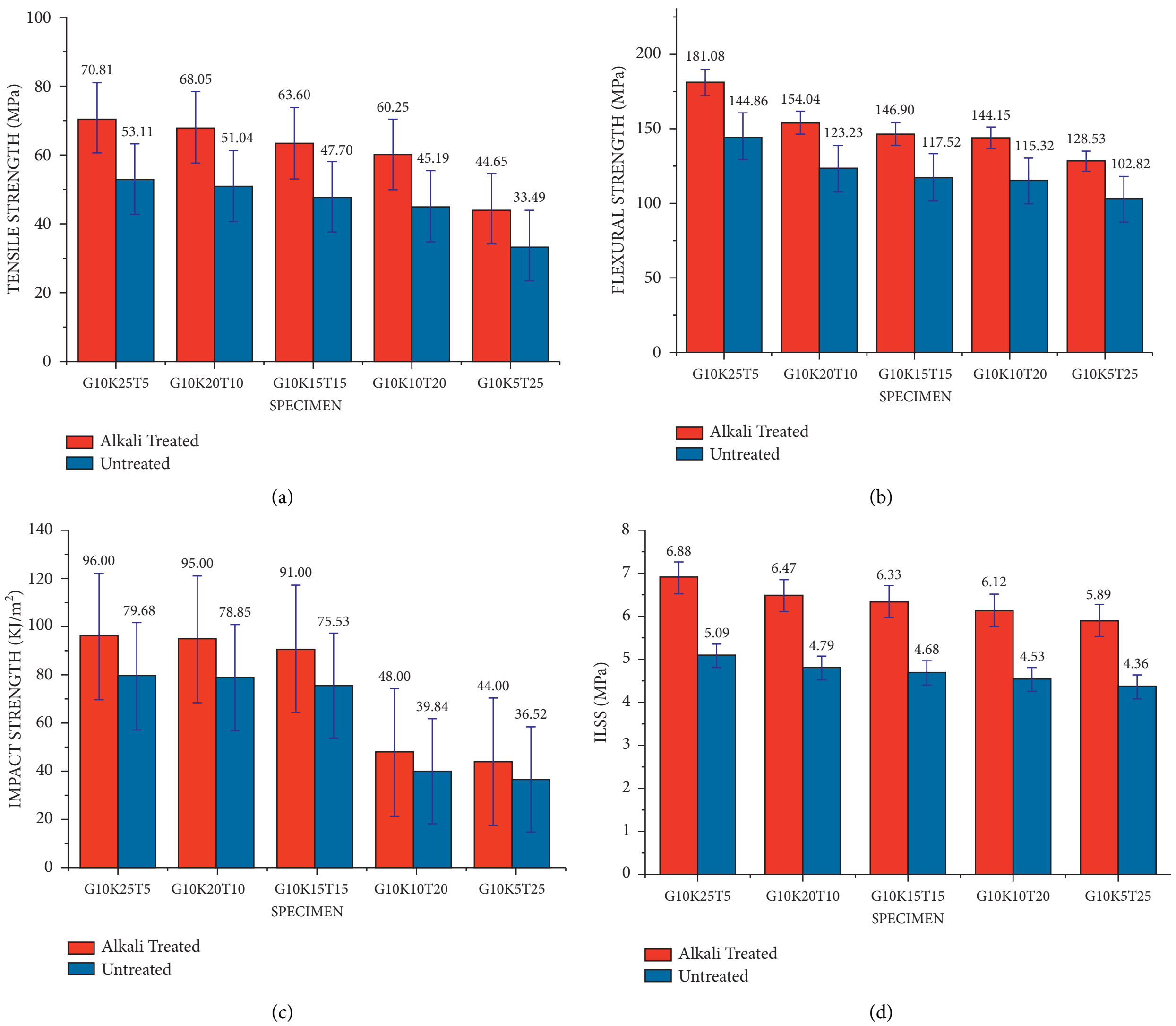


Figure 3: Tensile, flexural, impact, and interlaminar shear strength of glass/kenaf/tea leaf fiber-reinforced hybrid composites: effect of 5% NaOH alkaline treatment [12].

surface be rough enough has happened. Overall it was concluded that coconut sheath fiber-reinforced polyester composites could be a vital replacement for glass fiber-reinforced polymer composites. Kim et al. [179] studied the influence of bamboo fibers extracted by subjecting the fibers to steam explosion, alkalization, and chemical extraction. The conversion rate from raw source to extracted fiber was found to be significant for alkali extraction method. Then the extracted fibers were subjected to alkali, silane, and combined treatment with different proportion to study the optimal and suitable pretreatment type for bamboo fiber. The tensile strength and modulus of mercerized bamboo fibers were found to be superior when compared to silane and alkali/silane modified bamboo fibers. But the mechanical property of composites was found to be higher for alkali/silane-treated bamboo fiber-reinforced composites. Finally the water intake characteristics of alkali and alkali/silane-treated bamboo fiber-reinforced vinyl ester composites were better when compared to bamboo fiber-reinforced vinyl ester composites. Liu et al. [177] explored the silane coupling agent's influence on the tribological behaviour of corn stalk fiber-reinforced polymer composites. The silane treatment of fibril resulted in enhanced wear resistance; however, friction performance was not effective. The examination of worn surface morphology revealed the emergence of secondary plateau on the composite surface which enhanced the tribological characteristics of the composites. Lai et al. [180] investigated the possibility of fabricating fiber-reinforced polymer composites using coconut coir fiber as reinforcement material. The coconut coir fibers were exposed to mercerization followed by permanganate and stearic acid modifications to improve the effective adherence between the fiber and matrix. The fibrils were sized to 0.3 mm and 0.5 mm during the fabrication of the composites. Tensile and flexural strength results revealed that when the percentage of fiber loading increased, the strength values decreased. This was due to the inability of the fiber to support the stress shifting from the polypropylene

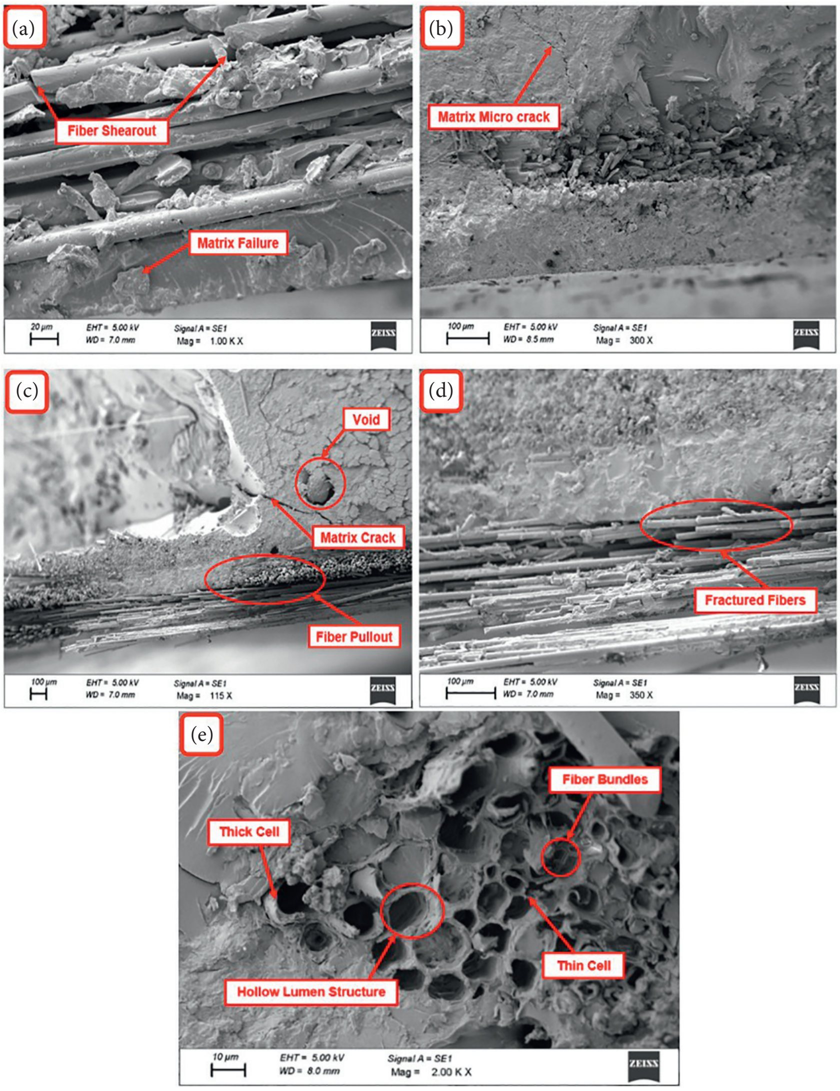
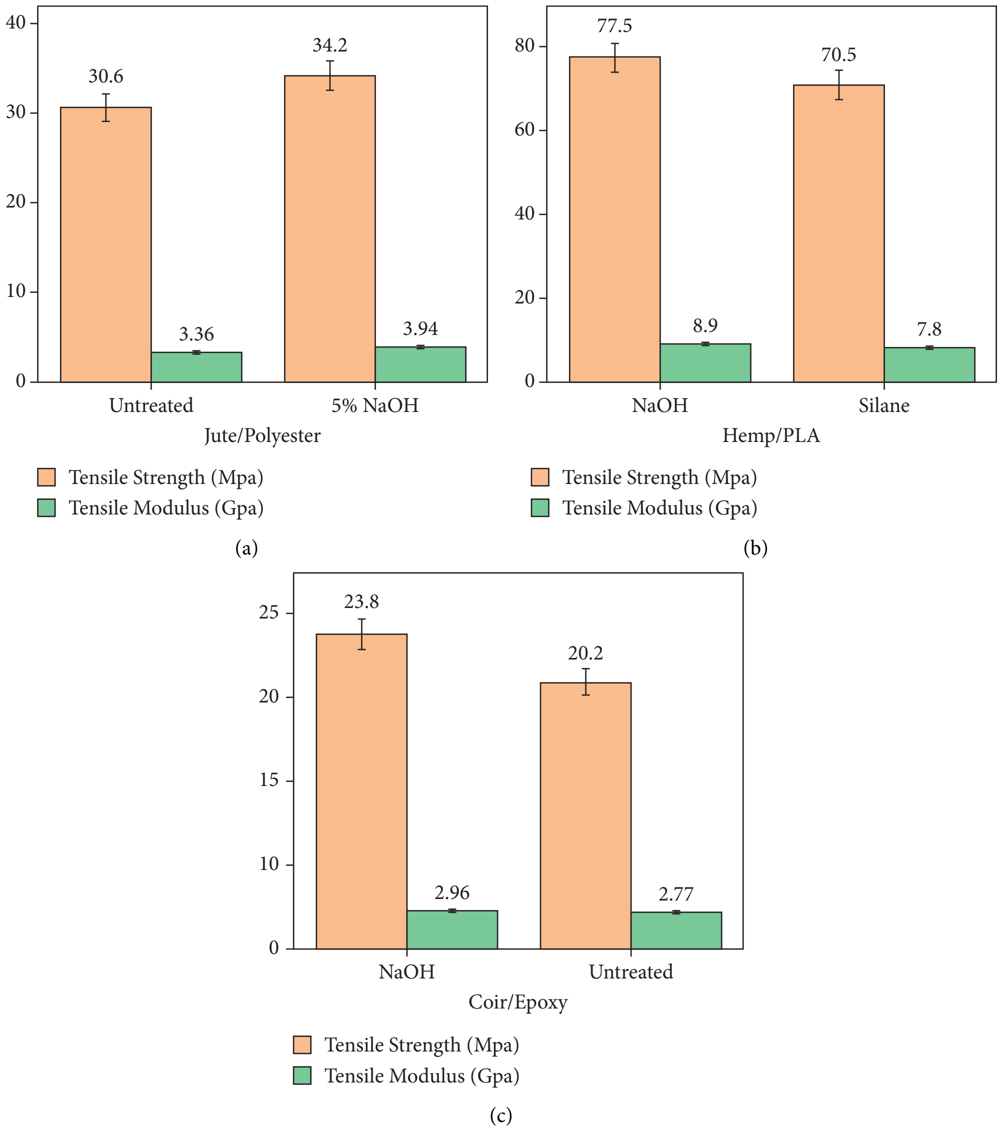


Figure 4: SEM micrographs of (a) tensile test of 5% NaOH-treated composites, (b) matrix microcrack, (c) fiber pull-out and void formation in the impact test specimen, (d) fractured surface of impact test specimen, and (e) hollow structure of natural hybrid fiber composites [12].

matrix and due to poor reinforcement property. Tensile and flexural modulus increased with increase in fiber loading, which was because of the higher fiber size, and this can also be inferred based on the aspect ratio of the fibrils. Zaman and Beg [119] evaluated the mechanical characteristics of banana fiber-strands-reinforced low density polyethylene matrix. The banana fiber strands were pretreated with methylacrylate (MA) solution combined with methanol and benzyl peroxide. The mechanical properties improved as a result of better adherence between the interface of banana fiber strand and polyethylene matrix. The banana fiber strands modified with starch solution showed improvement in composite properties against methylacrylate-treated fiber. Joseph et al. [161] studied the effects of benzoyl chloride treatment on sisal fiber and found maximum thermal stability compared to raw fiber composites. The treatment removes the hemicellulose and fatty substance in fiber surfaces for better mechanical and thermal properties. The sisal/glass/filler/epoxy reinforced composites were used in the frames, toys, and electronic panels. Sampathkumar et al. [181] analysed the influence of surface treatment on water absorption nature of *Areca* fiber. Due to the presence of hydroxide and other constituents in chemicals, the water absorption nature was higher and this leads to poor wettability. The results of their work concluded that there was a reduction in water absorption during acetylation of *Areca* fiber and increase in water absorption in treatment with alkali. Alfa fibers were kept under the various fiber surface treatments involving acetylating and it was confirmed that the treatment enhanced the resistance of fiber to moisture [182, 183]. Bisanda [184] reported that alkali-treated sisal fiber-reinforced polylactic acid composites removed lignin and waxy substances which led to better mechanical interlocking. The tensile, flexural, impact, and interlaminar shear strength (ILSS) of 5% NaOH-treated kenaf and tea leaf fibers-reinforced composites improved by 33.32%, 25%, 20.48%, and 35.16%, respectively, when compared with untreated composites due to removal of hemicellulose,



(a)

(b)

(c)

Figure 5: Tensile properties of 5% NaOH-treated jute/polyester and hemp/PLA composites [190].

lignin, pectin, and waxy elements which resulted in better interactions between hydrophilic fiber and hydrophobic matrix [12]. Figure 3 presents the mechanical properties of glass/kenaf/tea leaf fiber-reinforced hybrid composites.

The SEM analysis (Figure 4) shows that mechanical properties of untreated fiber composites were decreased by the problems such as fiber breakage, fiber pull-out, formation of micro voids, and nonuniform distribution of fiber and matrix. The 5% NaOH-treated kenaf and tea leaf fiber improved the adhesion between the fiber and matrix which resulted in better mechanical properties [12].

The biodegradability of chemically modified cellulosic fibers is associated with physical, chemical, mechanical, and thermal properties [5, 185, 186]. The agricultural waste fiber-reinforced composites are used in automobile, aerospace, construction materials, packaging applications, and medical applications. Sharma and Kumar [187] studied the tensile properties of sugar palm fiber obtained from different height of the palm plant. The tensile properties enhanced in the bottom part of the tree fiber due to their chemical composition, particularly cellulose, hemicellulose, and lignin [188, 189].

The tensile strength and tensile modulus of 5% NaOH-treated jute fiber-reinforced polyester composites improved by 5.2% and 17.2%, respectively, when compared with untreated composites due to better interaction between fiber and matrix (Figure 5). It was also revealed that 5% NaOH-treated hemp fiber-reinforced PLA composites exhibited outstanding results compared to silane-treated hemp fiber-reinforced PLA composites [190]. The tensile strength and tensile modulus of 5% NaOH-treated hemp fiber-reinforced PLA composites improved by 9.9% and 14.1%, respectively, when compared with silane-treated composites owing to better removal of unwanted substances such as hemicellulose and waxy elements. Likewise, 5% NaOH-treated coir fiber-reinforced epoxy composites exhibited superior tensile properties such as tensile strength (17.8%) and tensile modulus (6.8%) compared to untreated fiber composites. Based on various chemical treatments, alkaline treatment (5% NaOH) is the most economical and effective treatment in promoting better communications between fiber and matrix by removal of hemicellulose, lignin, and waxy elements due to disruption of hydrogen bonding in the fiber structure, thus resulting in better mechanical and thermal properties.

4. Conclusions

In this detailed review, the effects of various chemical treatments on different biofiber-reinforced composites were summarized. Furthermore, the effect of constitution of biofibers was also reported. The physical, mechanical, and thermal properties of various biofibers-reinforced composites were improved up on modification of fiber surfaces, while fiber swelling effect and water absorption rate were decreased by various chemical treatments like alkaline, silane, acetylation, permanganate, peroxide, benzoylation, acrylonitrile grafting, maleic anhydride grafted, acrylation, and isocyanate. Based on various chemical treatments, alkaline treatment (5% NaOH) is the most economical and effective treatment in promoting better communications between fiber and matrix by removal of hemicellulose, lignin, and waxy elements due to disruption of hydrogen bonding in the fiber structure, thus resulting in better mechanical and thermal properties. It was concluded that alkaline treatment of fibers with 5 wt% NaOH made the fibers more resistant to deformation and heat. The alkaline treatment has been one of the successful methods used to treat the natural fibers in order to achieve better results.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

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Research Article

Investigation of Weight Fraction and Alkaline Treatment on *Catechu Linnaeus/Hibiscus cannabinus/Sansevieria Ehrenbergii* Plant Fibers-Reinforced Epoxy Hybrid Composites

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The aim of the present work is to develop novel hybrid composites using areca, kenaf, and snake grass fibers as reinforcement and epoxy as the matrix. The areca, kenaf, and snake grass fibers were extracted from *Catechu Linnaeus, Hibiscus cannabinus*, and *Sansevieria Ehrenbergii* plants, respectively, and treated with 5% NaOH to improve the interfacial adhesion between the hydrophilic fiber and the hydrophobic matrix. Hybrid composites were developed by the compression molding technique and formulated based on the weight fraction of fibers. Tensile, flexural, and impact strength and hardness samples were prepared as per ASTM *D* 3039, ASTM *D* 790, ASTM *D* 256, and ASTM *D* 2240, respectively. The effects of alkaline treatment on developed hybrid composites were investigated. The developed hybrid composites with 20% wt. snake grass and 10% wt. areca fiber present interesting mechanical properties with a tensile strength of 58 MPa, flexural strength of 124 MPa, impact strength of 5.24 kJ/m^2, and hardness of 88. The results indicate that maximum mechanical properties were obtained for alkaline-treated fiber composites with 20% wt. snake grass fiber compared to untreated fiber composites owing to better adhesion between the treated fiber and the matrix. The effect of alkaline treatment was analyzed by Fourier transform infrared. The fractured surfaces of tested samples were analyzed by scanning electron microscopy.

1. Introduction

The role of natural fibers has been increasing in this world due to their outstanding properties, such as light weight, low energy consumption, renewability, worldwide availability, biodegradability, cost-effectiveness. Regardless of their properties, there are some limitations to natural fibers, such as high moisture content, low thermal stability, and incompatibility with the matrix. Reinforcement of natural fibers along with the polymer matrix has been emerging in this world due to its extended applications and higher specific properties [1]. Natural fibers are used in automobile structures because of their moderate tensile strength, better stiffness, and high damping capability. When natural fiber-reinforced composites are used in vehicles, it is expected that not only the weight of the component will be decreased, but also noise and vibration will be reduced. In addition to these, composites have high resistance to fatigue and corrosion [2]. The properties of natural fibers depend on the age, nature of soil, and environmental conditions [3]. Kenaf fiber (*Hibiscus cannabinus*) has been chosen for this work because it can grow under different climatic conditions. It shows properties such as low density, nonabrasiveness during processing, and high specific mechanical properties [4]. Lee reported that the tensile strength and modulus of both kenaf and jute-reinforced polypropylene composites strengthened with higher fiber loading, achieving maximum strength of 39 MPa and 1300 MPa, respectively, at 40% fiber weight fraction before declining at higher fiber weight fractions [5]. Singh stated that kenaf fibers assisted in enhancing the wear and frictional performance of the polyurethane thermoplastic composite by about 59 and 90% [6]. Areca (*Catechu Linnaeus*) and Snake fiber (*Sansevieria Ehrenbergii*) along with the kenaf fiber have been taken for this experimental work due to their attracting nature. Areca fiber is inexpensive, and it is used in medicine, paints, chocolates, etc. [7]. The areca evergreen tree grows linearly, reaching a height of 10 m to 20 m, and its stem is straight and slender, with a diameter of less than 15 cm. The leaves are 1.5 to 2 m long and have a large number of pinnately shaped leaves, with the upper part usually displaying 9 to 12 fronds. However, soil conditions have a large impact on the growth of these trees [8]. The areca husk accounts for roughly 60–80% of the overall weight and volume of the fresh fruit. The husk fiber is made up of cellulose with different amounts of hemicelluloses, 35–64.8%, lignin 14.0–26.0%, and pectin [9]. Snake grass-reinforced polymer composites may supplant sal wood and other wood species and save trees, which helps in diminishing environmental damage [10]. Besides their scope, there are some limitations to reinforcing natural fibers along with the polymer matrix, like low processing temperatures, high water intake, and improper stress transfer. Cellulosic fibers absorb more moisture content because of their hydrophilic nature. This nature of cellulosic fibers results in protrusion. When this fiber gets reinforced with the hydrophobic matrix, it results in improper bonding and poor interfacial shear strength [11, 12]. If fibers are not treated and if they are embedded in a polymeric matrix, it results in generating unstable interfaces and, therefore, the stress applied to the fiber/polymer composite is not efficiently transferred from the matrix to the fiber. These issues can be diminished by various treatment processes, thereby increasing the mechanical properties of the natural fiber-reinforced composites. Alkali treatment is a suitable method for enhancing the mechanical properties of natural fibers [13, 14]. This treatment will modify the surface by removing hemicellulose, lignin, pectin, and wax, thereby increasing the aspect ratio. A higher concentration of NaOH results in damage to the fiber and hence decreases the mechanical properties. So, optimum treatment percentage plays a crucial role in the modification of the fiber surface [15, 16].

Karsli and Aytac et al. [17] studied the tensile properties of flax fibers-reinforced PLA polycarbonate composites as function of 5% NaOH alkaline treatment for 20 min. The authors observed that 5% NaOH-treated flax fibers-reinforced PLA polycarbonate composites increases the flexural strength and flexural modulus by 9.1% and 62.5%, respectively, compared to untreated fiber composites. The maximum tensile strength, flexural strength, and modulus of the chopped snake grass fiber-reinforced polyester composite are achieved at 25% volume fraction for 30 mm of fiber length. The result indicates that, overall, 25% of the fiber fraction showed better mechanical properties [18]. Mazuki [19] conducted a study on kenaf fiber and highlighted its potential use in verities of applications such as panels of doors, seats, armrests, and dashboards. The potential of using kenaf-reinforced composite materials as sound barriers and acoustic absorbers was also indicated in the article. The tensile, flexural, impact, and interlaminar shear strength (ILSS) of 5% NaOH-treated kenaf and tea leaf fibers-reinforced composites were improved by 33.32%, 25%, 20.48%, and 35.16%, respectively, when compared with untreated composites due to removal of hemicellulose, lignin, pectin, and waxy elements which resulted in better interactions between hydrophilic fiber and hydrophobic matrix [20]. The treated surface of the fiber becomes rough, thereby enhancing the interfacial bonding and mechanical properties [21]. The tensile strength of randomly distributed snake grass fiber-reinforced composites was found to be lower than the tensile strength of longitudinal direction oriented composite material [22]. Satyanarayana et al. [23] investigated the chemical and physical characteristics of natural fibers. Jayabal et al. [24] studied randomly oriented coir fiber-reinforced polyester composites manufactured by hand layup technique for different fiber lengths of 20, 100, and 150 mm. The mechanical strength of polyester composites varies depending on the weight and length of the fibers. The findings revealed that fiber content had quite a greater impact on mechanical characteristics than fiber length [25]. Usually, the weight of the fiber content improves the tensile properties, and this is dependent on the type of matrix used [26]. A prolonged literature review has led to the selection of the optimum alkali treatment process. The experimental work emphasizes the importance of alkali treatment on the mechanical properties of snake grass, areca, and kenaf fiber-reinforced hybridized epoxy composites. No literature has reported data on kenaf-, snake grass-, and areca-reinforced hybridized epoxy composites. So, this hybridization combination was chosen and the main objectives of the paper were carried out as follows: (i) effect of alkaline treatment on kenaf, snake grass, and areca fiber and (ii) effect of weight fraction of snake grass fiber on mechanical properties of hybrid composites.

2. Experimental Work

2.1. Materials. Areca, kenaf, and snake grass fibers were collected from KCT, Tifac core, Coimbatore, India. For the preparation of composites, a widely available epoxy resin

(LY 556) and an amine-based hardener, triethylenetetramine (HY 951), were used. The hardener is used to enhance bonding between the fibers and the matrix. They were used in the mixing ratio of 10 : 1 as recommended by the supplier. The epoxy and hardener were purchased from Covai Seenu and Company, Coimbatore, Tamil Nadu. Epoxy resins (LY556) have prominent advantages over thermoplastic and other thermoset resins such as minimum shrinkage rate, excellent moisture and chemical resistance, and better damage tolerance [27]. Properties of epoxy resin (LY556) are presented in Table 1. The hardener (HY951) is made from polyamine monomers, such as triethylenetetramine. When these compounds are combined, the amine groups react with the epoxide groups to form a covalent connection. Amine hardeners react with epoxy resins and contribute to the ultimate properties of the epoxy resin treatment system.

Table 1: Properties of epoxy resin (LY556) (27).

Property	Epoxy resin
Density (g/cm^3)	1.1 to 1.4
Elastic modulus (GPa)	3 to 6
Tensile strength (MPa)	35 to 100
Compressive strength (MPa)	100 to 200
Elongation (%)	1 to 6
Cure shrinkage (%)	1 to 2
Water absorption (%)	0.1 to 0.4
Impact strength (J/m)	0.3

2.2. Process of Alkali Treatment. The alkaline treatment is also referred to as mercerization. This causes fibrillation, which leads to the breakdown of fiber bundles into smaller ones. The fiber surface becomes rough and the diameter of the fiber decreases, increasing aspect ratio and mechanical properties [28]. It breaks the hydrogen bond in the cellulose fibers and increases the number of reaction sites, which promotes better interfacial shear strength and stress transfer. Mercerization removes the noncellulosic contents like hemicellulose, lignin, and wax from the surface of the fiber and enhances the properties of the composite [29]. Hemicellulose in plant fiber has a more hydrophilic part, so alkali treatment decreases the moisture absorption of water. The reaction that occurred during the treatment is as follows:

$$\text{fiber} - \text{OH} + \text{NaOH} \longrightarrow \text{fiber} - \text{O} - \text{Na} + \text{H}_2\text{O}. \quad (1)$$

The hydrophilic nature of the fiber is reduced and the resistance to moisture increases. The amount of hemicellulose, pectin, lignin, and wax will be removed depending upon the concentration of treatment [16, 30, 31]. Among the various chemical treatments, alkali treatment has been identified as one of the best and most effective for removing impurities from the surface of the fibers. Reduction of the hydrophilic nature of the fibers has been carried out in this work by chemically treating the surface using NaOH. The fibers of areca, kenaf, and snake grass were soaked in 10 liters of distilled water for 24 hours to remove dusty layers and then hung to dry for 24 hours. For 3 hours, dust-free fibers were immersed in a 5% NaOH concentration solution (50 ml of NaOH in 10 liters of water). The fibers were then removed and washed several times in distilled water to remove the excess accumulation of NaOH solution on the surface of the fibers. The fibers were then air dried for 24 hours.

2.3. Fabrication of Hybrid Composites. Initially, treated fibers were cut into 28 cm lengths as per mold requirement with the help of a cutter. For preparing the epoxy matrix, both the epoxy resin and epoxy hardener (LY 556 and HY 951) were mixed with the aid of a stick in the ratio of 10 : 1. Following the preparation of matrix materials and reinforcement, the composites were manufactured using the compression molding technique (Supplier: Modern Plastics Pvt Ltd., Coimbatore, India). In the fabrication process, 300 * 300 mm aluminum plates were used. Aluminum plates and frames are first cleaned, and then white grease is applied to the aluminum plates to reduce the friction between them. Then the epoxy resin is poured onto the aluminum plate, and the fibers are arranged on the plate bidirectionally, with the areca fiber placed on the bottom surface, the middle layer of the laminate is occupied by kenaf fiber, and the top layer is occupied by snake grass fiber. The epoxy matrix is poured between each layer of fiber in order to achieve uniform dispersion [32]. Five samples were prepared in the same order using different fiber content. Then the completed laminates were placed inside the modern compression molding machine and maintained at a temperature and pressure of 120 °C and 35 bar for 45 minutes and the laminates were cured for another 45 minutes. After solidification, the final dimension of the composite is 280 mm × 280 mm *x* 5 mm, obtained from the mold cavity. The diamond cutter was used to cut the samples for mechanical characterization tests as per ASTM Standard. Table 2 presents the designation of hybrid composites. Figure 1 shows graphical procedure of composite fabrication process.

2.4. Mechanical and Morphological Analysis. It is one of the simplest and most commonly used mechanical tests. The samples were tested on a computerized universal testing machine (UTM) (Supplier: Aimil Ltd., India) with a crosshead speed of 2 mm/min. The tests were carried out in accordance with ASTM *D* 3039 standards with a specimen size of 250 × 25 × 5 mm [33]. Each composite was tested with five specimens, and their values are noted. Figure 2 shows tensile gripper and tested samples. Flexure tests are generally used to determine the flexural modulus or bending of a material. The samples were tested using a three-point bending test on a computerized UTM with a crosshead speed of 2 mm/min. The tests were carried out in accordance with ASTM *D* 790 standards with a specimen size of 125 × 12.7 × 6.5 mm [33]. Each composite was tested with five specimens, and their values are noted. This test is used to determine a material's impact resistance. In this experiment, a hybrid combination was examined for the ability to absorb energy without breaking. The samples were tested on a digitalized Izod impact tester.

The tests were carried out in accordance with ASTM *D* 256 standards with a specimen dimension of

Table 2: Designation of hybrid composites.

Sample designation	Kenaf fiber (% wt.)	Snake grass fiber (% wt.)	Areca fiber (% wt.)	Epoxy resin (% wt.)
A	10	5	25	60
B	10	10	20	60
C	10	15	15	60
D	10	20	10	60
E	10	25	5	60

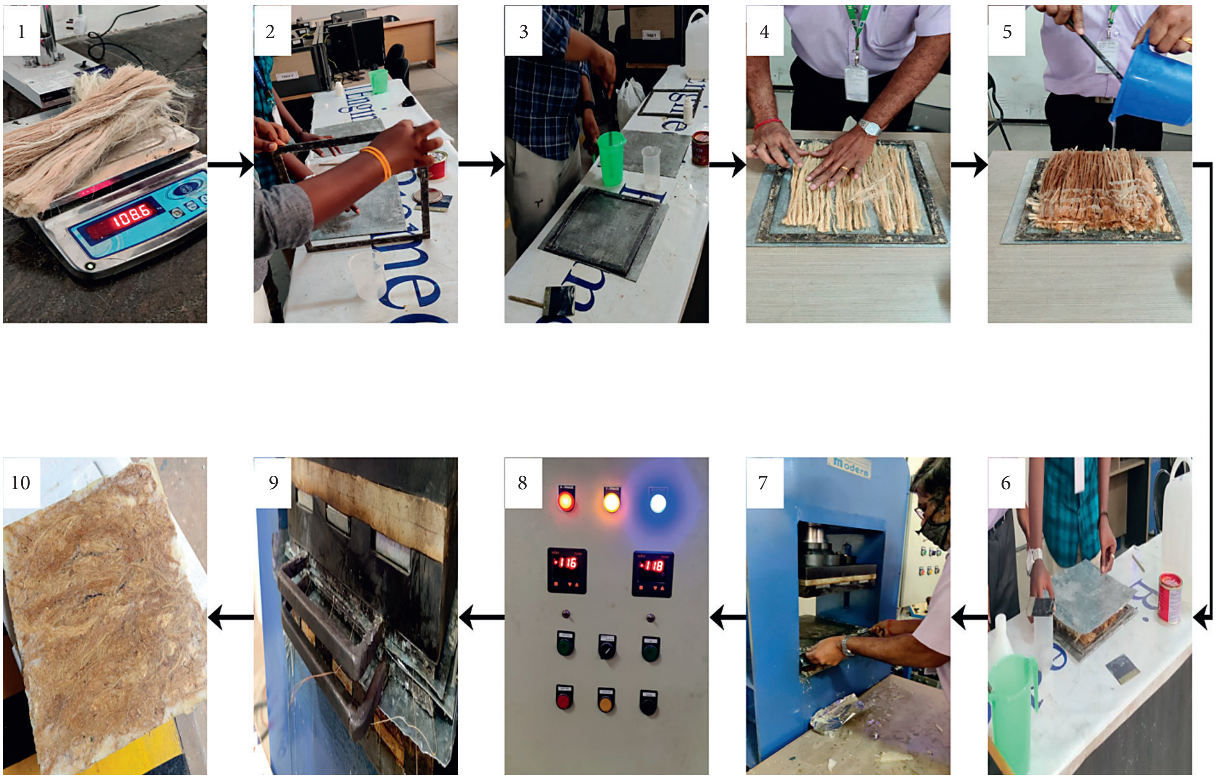


Figure 1: Graphical procedure of fabrication process.

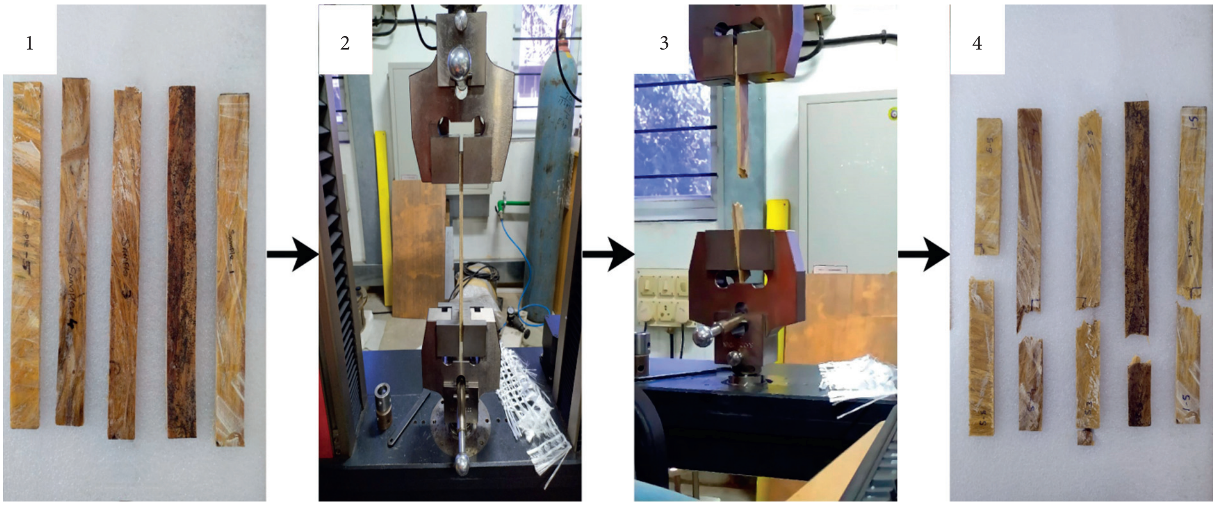


Figure 2: Tensile setup and fractured samples.

65 × 12.7 × 6.5 mm [33]. Each composite was tested with five specimens, and their values are noted. Hardness is determined by how little deformation it offers under localized mechanical or abrasive pressure. The samples were tested on a Shore *D* Durometer. The tests were carried out in accordance with ASTM *D* 2240 standards with a specimen

dimension of 20 × 20 × 6.5 mm. Five specimens were tested, and six readings at different points were taken. A mean value was determined for the specimens. A scanning electron microscope was used to examine the morphology of the modified fibers using (SEM JEOL JSM-6510LA). After fabrication, the surfaces characteristics of the composite materials are investigated using SEM. Scanning electron micrographs clearly show the interfacial adhesion between the matrix and the fiber. The SEM operates at an accelerating voltage of 25 kV.

3. Results and Discussion

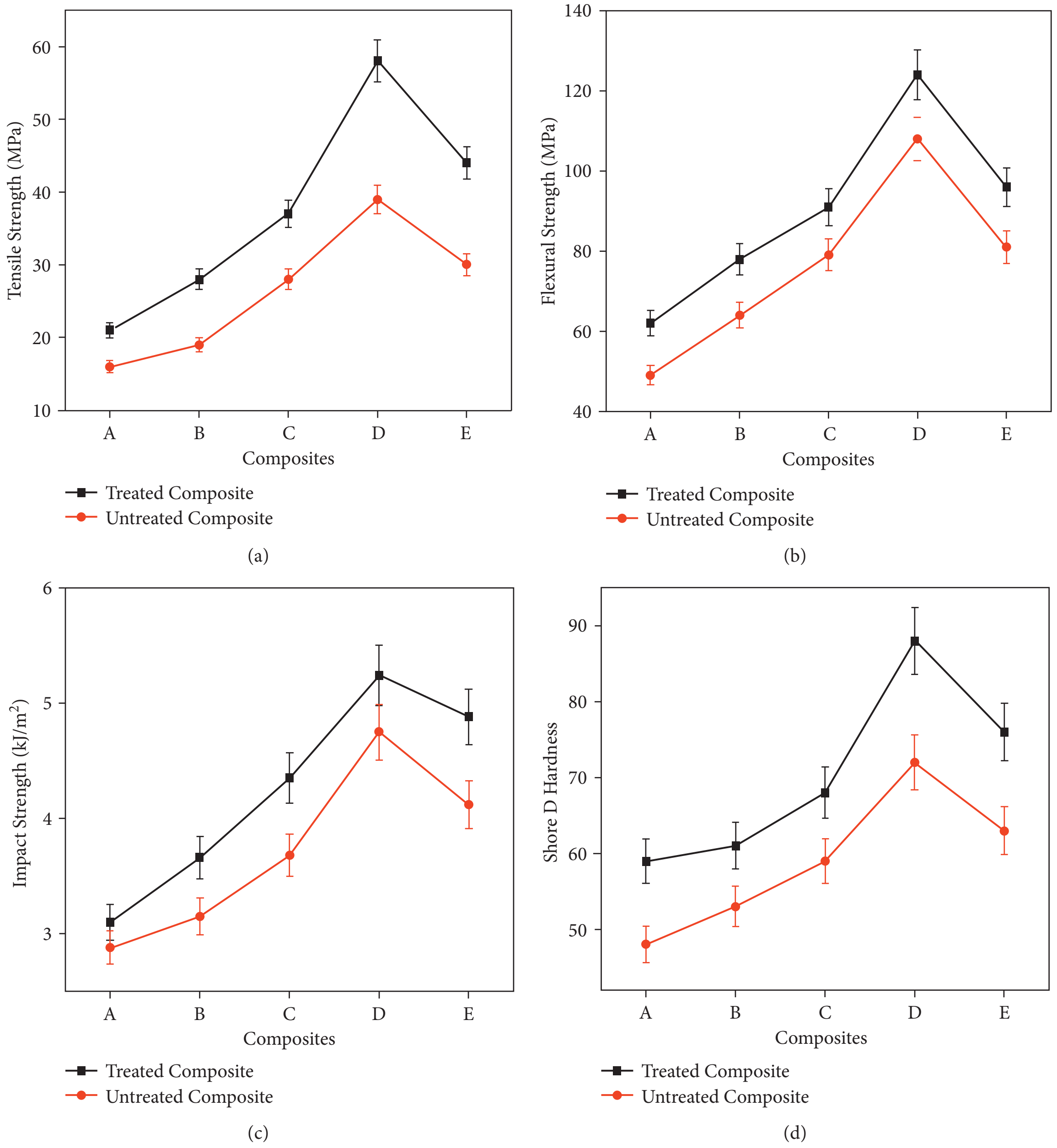
3.1. Mechanical Characterization. Fiber content and strength are influencing parameters in determining the properties of the composites. Specimen *D* gave a higher tensile strength of 58 MPa, followed by specimen *E* (44 MPa), specimen C (37 MPa), specimen B (21 MPa), and specimen A (21 MPa). Figure 3(a) shows the tensile strength of treated and untreated composites. Among the hybrid composites, specimen *D* has 20% wt. snake grass fiber which showed enhanced tensile strength and further increase in snake grass fiber led to a sudden decrease in tensile strength. From the results, it can be inferred that the alkali treatment increases the elastic behavior and makes it able to withstand failure. At a lower strain rate, the stress increases linearly, forming the elastic region, and above this zone, the specimen exhibits plastic deformation. This behavior is due to the reason that resin in the region starts to deform plastically, and it leads to the formation of minor cracks in the resin [33]. When the fibers propagate through these cracks, it leads to a decrease in tensile strength. The tensile strength of specimen A is much lower than specimen D. This is due to the reason that the 20% wt. snake grass fiber in specimen A bonded strongly with the epoxy matrix, thereby enhancing the tensile properties and interfacial shear strength, whereas specimen A has 25% wt. of areca fiber which does not properly interact with the matrix when compared to other composites. According to Maslinda et al. [34], the tensile strength of the hybrid composites extremely weakened and their results showed that mechanical properties of the hybrid composites were influenced by weight fraction of cellulosic fiber. With the increase of cellulosic fiber beyond 40% wt. in composites, the adhesion between the hydrophilic fibers and the hydrophobic matrix deteriorated leading to poor strength. The improper bonding is due to the higher amount of hemicellulose percentage, which is responsible for moisture content. 5% alkali treatment on areca is not efficient enough to remove the noncellulosic contents to a better extent. The tensile strength of all untreated fiber composites is lower as compared to alkaline-treated fiber composites. This indicates that 5% NaOH reagent increases the roughness of fibers and decreases the fiber diameter, resulting in greater tensile properties.

Flexural strength determines how the material will resist bending force. Specimen *D* shows a higher flexural value of 124 MPa when compared to other specimens. The causes of the higher flexural strength of the specimen *D* are stated as follows: (i) 20% wt. snake grass fiber is properly bonded with the matrix, which means that 5% NaOH has effectively removed the hydrophilic nature of the fiber. (ii) The diameter of the fiber decreases so that crack generation will not occur. (iii) Due to this, the interfacial bond between the fiber and the matrix becomes strong and fiber pullout will not be generated on the surface. (iv) Enhanced interfacial bonding leads to proper stress distribution. Specimen *E* has the highest flexural strength of 96 MPa, followed by specimen C (91 MPa), specimen B (78 MPa), and specimen A (62 MPa). Figure 3(b) shows the flexural strength of treated and untreated composites. The decrease in flexural strength can be due to the presence of voids in the surface [33]. Specimen A has more holes on the surface. That is why the flexural strength is less when compared to other specimens. Delamination of specimen A occurs at a faster rate when compared to other specimens. The weaker fiber/matrix adhesion could not offer adequate stress transport; thus, the flexural strength of the hybrid composites reduced considerably [35]. Alkaline-treated fiber-reinforced composites display the highest flexural strength, while untreated fiber composites reveal a marginal loss in flexural strength due to poor adhesion between fiber and matrix.

Impact strength determines how much energy the material can withstand when a load is applied to it. Specimen *D* has the highest impact strength of 5.24 $\mathrm{kJ/m^2}$ because of its low brittleness. This is so because the stress distribution between each lamina has been distributed equally. The reasons are stated as follows: (i) proper alkali treatment reduces the diameter and makes the surface rough. (ii) In addition to that, all the noncellulosic components are removed to an optimum extent. (iii) Due to this, the material's ductile-brittle transformation will not happen soon [36]. The brittle behavior in all other specimens will occur soon. Figure 3(c) shows impact strength of treated and untreated composites. From the results, it can be concluded that other specimens will not absorb much energy when compared to specimen D. At higher luffa cylindrica fiber volume fraction, the accretion of reinforcement in the composites causes poor matrix regions leading to inadequate adhesion between the luffa cylindrica fiber and epoxy matrix and results in reduced impact strength [37].

Low energy absorption of the specimen is due to protrusion of the fiber surface, which leads to the onset of brittle behavior [36]. As a result of the impact test, it can be indicated that 5% NaOH solution is optimum for enhancing the impact strength of the composites compared to untreated fiber composites owing to minimized voids, formation of pores, and removal of moisture content, hemicellulose, lignin, and wax.

The hardness test was done using the Shore *D* Durometer. The ability of a material to resist deformation is referred to as hardness. Among those five specimens, specimen *D* has the highest hardness value of 88. This is because the interlaminar strength between the fibers and the matrix is strong [33]. As a result, the composite becomes finer and more capable of resisting applied force, so the highest value is obtained for specimen *D*, followed by specimen *E* with a hardness value of 76, specimen C with a hardness value of 68, and specimen B with a hardness value



(a)

(b)

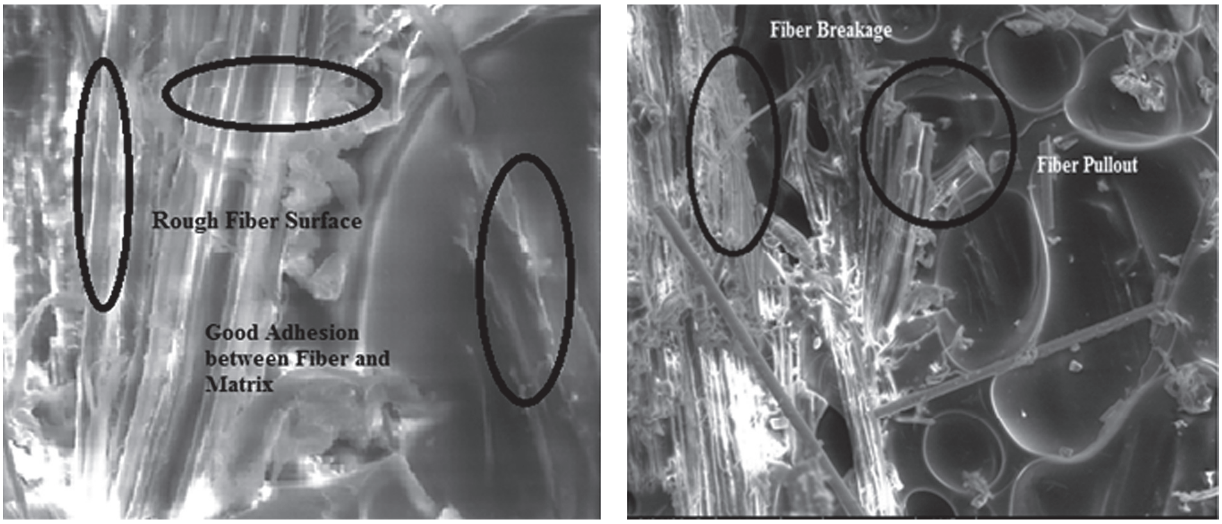
(c)

(d)

Figure 3: Alkaline-treated and untreated composites: (a) tensile strength, (b) flexural strength, (c) impact strength, and (d) shore D hardness.

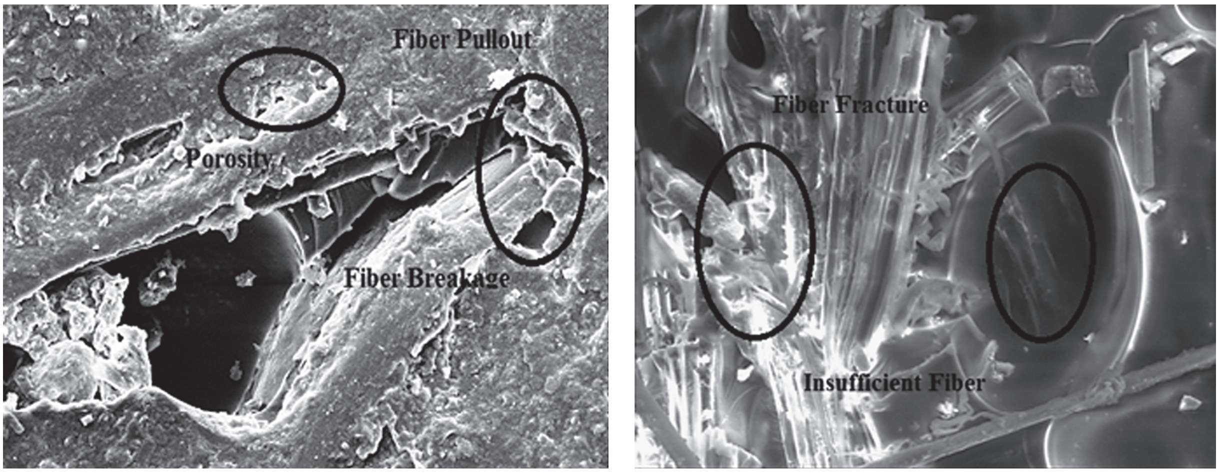
of 61. Specimen A has the lowest hardness value among the five specimens, which is due to improper fiber-matrix adhesion (Figure 3(d)). Because 25% wt. of the areca fiber did not remove the noncellulose compound more effectively, the fiber-matrix adhesion was poor, resulting in a lower hardness value. When there are fewer fibers in the region, the hardness value is low. When there are more fibers focused on a specific location, then the hardness value is high. The hardness values are influenced by the strong fiber-matrix bonding. The hardness of alkaline-treated composites indicates noticeably better interaction between fibers and matrix which resists indentation or penetration as compared to untreated fiber composites [20].

3.2. SEM Analysis. Among the various hybrid composite specimens, specimen *D* has 20% wt. of snake grass fiber, 10% wt. of kenaf fiber, 10% wt. of areca fiber, and 60% wt. of resin showing improvement in fiber-matrix adhesion. The surface of specimen *D* has no fiber pullout and, from this, it can be concluded that specimen *D* has bonded properly with the epoxy matrix and the surface of the composite produced a rough nature (Figure 4(a)). The rough nature is due to the removal of hemicellulose, which is responsible for moisture content. Due to this, there is no generation of microcracks and this leads to proper stress distribution by carrying the loads efficiently [36]. In Figure 4(b), specimen *E* showed some microcracks in the surface of the fiber, which is due to



(a) (b)

Figure 4: SEM images: (a) 20% wt. of snake grass fiber-reinforced composite; (b) 25% wt. of snake grass fiber-reinforced composite.



(a) (b)

Figure 5: SEM images: (a) 5% wt. of snake grass fiber-reinforced composite; (b) 10% wt. of snake grass fiber-reinforced composite.

the fact that 25% wt. of the snake grass fiber did not bond properly with the epoxy matrix. Figures 5(a) and 5(b) show the SEM images of specimens A and B which depict more fiber breakage, fiber pullout, and insufficient distribution of fiber in the matrix leading to failure of composite under minimum load.

4. Conclusion

This paper investigated and reported the effect of alkali treatment on hybridized areca, kenaf, and snake grass fiber-reinforced epoxy composites. Fibers were treated with 5% NaOH to improve the interfacial adhesion between the fiber and the epoxy matrix. The compression molding technique was used to fabricate hybridized fiber-reinforced epoxy composites by keeping the weight percent of kenaf fiber content constant at 10% wt. and changing the weight percent of areca and snake grass fiber to 25A-5SG, 20A-10SG, 15A-15SG, 10A-20SG, and 5A-25SG. The mechanical properties of 5% NaOH treated areca, kenaf, and snake grass fibers-reinforced composites with varying fiber content were tested. The results of the tests lead to the subsequent conclusions:

(i) The composite containing 10% wt. areca, 10% wt. kenaf, and 20% wt. snake grass fibers had the highest tensile, flexural, impact, and hardness values. This is due to the fact that 20% of alkaline-treated SG fibers remove hemicellulose more effectively, improving fiber-matrix adhesion compared to untreated fiber composites.

(ii) 5% NaOH alkaline treatment of fibers reduced their hydrophilic nature, which improved fiber-matrix adhesion and resulted in good mechanical properties.

(iii) The SEM micrograph of (10% wt. A–10% wt. K–20% wt. SG fibers) clearly shows no fiber pullout, indicating that the fiber and matrix were well bonded.

(iv) Based on the mechanical properties, it was concluded that the areca, kenaf, and snake grass fibers are alternative reinforcements for the development of hybrid composites for production of less weight products used in aircraft, automobile, building and constructions, sports and home appliances, etc.

Data Availability

The data used to support the findings of this study are included in the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

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Research Article

Experimental Study on the Sound Absorption Properties of Finger Millet Straw, Darbha, and Ripe Bulrush Fibers

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Nowadays, emerging noise pollution by external factors causes harmful diseases in human beings. The development of a bio-based filler or panel will help to eliminate some unwanted noise in working places and living rooms. This work aimed to develop an ecowaste fiber (leftover after harvesting)-based sound absorber and analyze its capabilities for sound absorption. The ecowaste fibers are collected by the gleaning process, i.e., the process of collecting leftovers from fields. The sound absorption capabilities of three natural fibers extracted from *Eleusine coracana* (Finger millet) straw, *Desmostachya bipinnata* (Darbha), and *Typha domingensis* (Ripe bulrush) plants are investigated in this study, both individually and in hybrid combinations. The sound absorption property mainly depends on factors such as porosity, flow resistivity, thickness, density, and tortuosity. Fiber length and fiber type play a significant role when fibers are arranged individually or in hybrid combinations. The stacking effect on the sound absorption coefficient of hybridized fiber arrangement was experimentally analyzed. The sound absorption coefficient (α) was found to be lower in the range of 1000 Hz–2500 Hz for all the combinations. As a homogenous fiber arrangement, the darbha fiber exhibited the better NRC (noise reduction coefficient) of 0.86 for 50 mm thickness among three different fibers and as a hybrid composition, ripe bulrush and darbha fibers exhibited NRC of 0.90 which is more capable of absorbing sound in the critical frequency range of 500 to 2000 Hz. These types of natural fiber fillers are highly capable of better sound absorbing and used in the applications such as classrooms, sound recording rooms, and theatres.

1. Introduction

In the modern era, one such issue is noise, and it is considered undesirable. Continuous exposure to noise levels of 80 dB or higher for more than eight hours a day increases tension and alters breathing patterns [1]. Since the inventions of new machinery and automobiles, noise control has become a significant concern. The initial solution to address these noise problems is to develop sound absorbers, barriers, and diffusers. Recently, researchers are working to develop more cost-effective and environmentally sustainable acoustic materials to address the concerns mentioned earlier.

Initially, manufacturers of sound absorbers used asbestos for cost-effectiveness. Later, it was proven that it has a carcinogenic dangerous emission on both humans and animals. Since then, the majority of industries have limited the use of asbestos and utilized synthetic fibers as a partial replacement. Besides asbestos, other materials were used in the past for sound absorption properties. However, it is also found to be hazardous when inhaled and results in lung diseases [2]. Numerous researchers have recently investigated the acoustic absorption properties of different natural biofibers, including kenaf fiber [3], coconut coir [4], oil palm fruits [5], and pineapple leaf fiber [6–8]. These studies demonstrated a significant potential for natural fibers to be used as insulating materials. Additionally, some researchers used kenaf/polypropylene nonwoven composite [9], coir composite [10], and oil palm fiber based composite [11] to establish its acoustic capabilities. Natural fibers performed exceptionally well when combined with other fibers and matrices in various composites. *Camellia sinensis*/*Ananas comosus*/glass fiber based composites exhibited good sound absorption properties for 25% by weight of *Camellia sinensis* because of its porous nature [12]. The chicken feather fibers are added by different weight percentages to the wood particleboard, and the result showed that the 5% chicken feather content was found to be a reasonable combination for maintaining acceptable fire characteristics in panels [13]. Coir-banana-polypropylene hybridization was found to have a lower sound transmission loss than individual fiber composites [14] and short ramie fiber composites had a higher SAC value than ramie fabric-based composites [15, 16]. The use of natural fibers as filler materials in sound absorption applications yields good results. The porous absorbers, panel absorbers, and membranes are the types of sound absorbers that permit the passage of sound and airwaves through materials with channels and cavities. According to the literature, sound absorbers or proofers are fibrous, cellular, or granular [17]. The fibers are pretreated with some chemical agents to remove their foreign substances for enhancing better adhesion. The results indicate that fibers that have been physically and chemically treated with some chemical agents have a higher NRC (noise reduction coefficient) than those that have not been treated. The surface modification and volume fraction of fiber increase interfacial adhesion, which improves both mechanical and acoustic absorption properties [12].

The sound absorption coefficient (SAC) results support the use of insulation panels made of tree bark as structural elements for noise reduction in residential structures, while also offering new thrust areas for further research in this subject [18]. Tudor et al. (2021) [19] have demonstrated that bark-based boards with fine-grained particles perform better in terms of sound absorption coefficient values than boards with coarse-grained particles. It is required to consider bark boards greater than 50 mm in thickness for their capability of performing an acoustic function in border structures. At less than 50 mm in thickness, the individual layers of the bark pieces are not overlapped, resulting in huge air spaces and an ineffective sound absorber.

Olcay and Kocak (2020) [20] investigated the effects of alkali treatment (NaOH) and fiber reinforcement ratios on the mechanical and sound absorption of PU foam-based composites. These fibers were pretreated with 10% alkali for 15 minutes. The composition with 5% by weight of the fiber exhibits a higher SAC of 0.41. These composites can be applied in the construction field and automotive parts where noise reduction is more desirable. The results reveal that agricultural waste products can be used as an additional alternative to increase the SAC of material without an increase in thicknesses [21]. To enhance the composite's acoustic properties, natural fiber coir was added with different blend ratios of reclaimed viscose (in percent), namely, 70 : 30; 60 : 40; 50 : 50; and 60 : 40, using a needle-punching technique [22]. The results reveal that when the viscosity content of the product increases, acoustic absorption also increases linearly. Also, the increased viscosity results in increased moisture absorption and the addition of fiber weight. Because of the presence of unidirectional coir fiber, the air resistivity increased with the denser fiber. The purpose of the layer's thickness is to increase the longer path for incident sound waves to pass through the material to lose more energy.

Flax has superior mechanical qualities when compared to other natural fibers [22] and is the strongest natural fiber in terms of properties, namely, tensile strength and crack inhibitor [23]. Additionally, flax fiber mixed with the epoxy demonstrated much greater vibration and sound dampening at low densities. In general, low permeability is considered a positive factor in enhancing the acoustic absorbance capacity in the low-frequency region [24]. Numerous researchers have already created natural fiber based composites using polymeric granules and fibers as an additional reinforcement that increases sound and physical properties [25]. Mamtaz et al. [26] have manufactured and analyzed novel composites comprised of natural fibers such as unidirectional coconut coir fibers and flakes form of rice husks. The results indicated that the produced composites exhibit an excellent sound absorption performance (SAC of 0.73) below 1500 Hz. This was attributable to the fact that adding rice husk filler to composites leads to the filling of the pores, lowering the porosity and increasing the surface contact area. These factors contribute to an increase in flow resistivity, which improves the SAC in low-frequency bands.

Berardi and Iannace [27] measured the SAC of kenaf samples by varying thicknesses and densities at 50 to 2500 Hz. When the density of fibrous increases from 45 to 110 kg/m^3, the SAC reaches 0.92 at a frequency of 2300 Hz. Lim et al. [28] investigated the SAC of kenaf fiber based fillers at 530 Hz to 4600 Hz with a sample thickness of 25 mm to 30 mm and a density of 160 kg/m^3. The result reveals that SAC is greater than 0.5 above 600 Hz, while the SAC exceeds 0.87 above 1750 Hz. Similarly, the investigation on the sound absorption of the kenaf fiber based composite sample also showed a better SAC of 0.89 by varying thicknesses and densities [29]. The SAC was measured using both impedance tube and reverberant chamber methods. The results indicate that samples having a thickness of 35 mm with a bulk density of 150 kg/m^3 exhibited better SAC and NRC of 0.65 and 0.53, respectively. Hao et al. [30] investigated the sound absorption characteristics of 50% kenaf and 50% polypropylene blended composite having 6 mm of thickness. It has been reported that SAC increases as the frequency of sound intensity increases.

The sustainable reuse of waste biomaterials in recent years has become crucial for environmental and economic preservation. Rice husk, ripe bulrush, and darbha are eco-waste materials (leftover fibers) found to be abundant in many regions. The present study aims to investigate the sound absorption properties (SAC-α and NRC) of finger millet straw, darbha, and ripe bulrush fibers for different thicknesses. Additionally, their hybrid combinations are investigated to understand the effect of hybridization and increase of thickness on the sound absorbing properties.

2. Materials and Methods

2.1. Materials. Straws of finger millet (*Eleusine coracana*) were collected from a harvesting site in the district of Mysuru, India. Finger millet straw fibers were obtained upon the chemical treatment of finger millet straws and the extraction process as shown in Figure 1. Darbha fibers were extracted from darbha plants (*Desmostachya bipinnata*) grown along the Cauvery riverbanks and the ripe bulrush fibers from *Typha domingensis*, a weed plant that grows along the banks of lakes in the Mysuru district. Chetana Chemicals, Mysuru, supplied chemicals, namely, sodium hydroxide, hydrogen peroxide, acetone, and double-distilled water to carry out the chemical pretreatments.

2.2. Fiber Extraction and Chemical Treatment. Finger millet straws were collected at the harvesting site during the extraction process of finger millet grains from the harvested finger millet plant. As illustrated in Figure 1, the collected finger millet straws were chopped to remove the interconnecting straw buds. The resulting bud fewer straws were washed five times with double-distilled water. This facilitated the removal of dust and dirt particles that adhered to the straw surface. The water-washed straws were then sun-dried for 12 hours appropriately. Darbha fibers were extracted from the plant by hand separation, after washing with double-distilled water and sun drying as shown in Figure 2. Lastly, ripe bulrush fibers were extracted from ripe bulrush grass using the combined retting process [31] and prewashed with distilled water before being dried in the sunlight as shown in Figure 3. Following sun-drying, the fibers and straws were alkali-treated (10% NaOH treatment for 24 hours) to remove any remaining dust and impurities [32]. They were washed with double-distilled water to remove any alkali substances that remained on the fiber surface. The obtained fibers and straws were sun-dried until moisture content was decreased to less than 2%. Additionally, the fibers of darbha and ripe bulrush were used to prepare samples. However, the finger millet straws were further treated with hydrogen peroxide and acetone (5 ml of hydrogen peroxide and 95 ml of acetone in a 100 ml solution) to obtain fine straw fibers of finger millet for sample preparation.

2.3. Fiber Properties. The fiber properties, such as fiber length, diameter, and density of all three fibers, are determined for the three different natural fibers used are listed in Table 1.

2.4. Preparation of the Testing Specimens. Chemically treated fibers of 300 mm in length were filled into plastic mesh for subsequent insertion into an impedance test tube as shown in Figure 4. The plastic mesh helps to hold the fibers tightly and has a negligible effect on the sound absorption of fibers. Samples are coded as S1–S3 (individual fibers), S4–S9 (stacked hybrid combinations), and S10–S12 (stacked hybrid combinations). The thickness of the samples containing individual fibers (S1, S2, and S3) are kept constant as 50 mm and hybrid combinations (S4 to S12) are kept totally as 50 mm and 25 mm individually. As shown in Table 2, S4–S9 are stacked hybrid combinations of fibers, whereas S10, S11, and S12 are homogenous hybrid combinations. All these samples (from S4 to S12) contain two distinct fibers, each contributing 25 mm in thickness, resulting in the formation of a 50 mm thick hybrid fiber combination. Thus, all samples from S1 to S12 were examined for their sound absorption properties—individual fibers (S1, S2, and S3), stacked hybrid fibers (S4–S9), and homogenous hybrid fibers (S10, S11, and S12), as illustrated in Figure 5.

2.5. Experimental Setup. The sound absorption coefficients (α) of individual and hybrid fibers were determined using an impedance tube according to ISO 10534 (2) 1998 standard [33]. Figure 4 depicts the experimental setup with an impedance tube, which includes an impedance tube, a data analyzer, and a data acquisition system. The sample holder has a diameter of 45 mm, the microphones are 30 mm apart, and the distance between the test sample and the nearest microphone is 90 mm. The sound absorption properties were determined over a frequency range of 100 Hz to 4500 Hz and at sample thicknesses of 10 mm, 20 mm, and 50 mm. To investigate the effect of air gap on the sound absorption coefficient of fiber, a 10 mm air gap is provided between the fibers and the sample holder. Because the fibers are held in a net, a 10 mm air gap can be maintained behind fiber samples. To begin, fibers (wrapped in a net) measuring 50 mm in thickness are inserted into the sample holder in such a way that they are entirely in contact with the sample holder's innermost surface. Using the scale engraved on the sample holder, precisely move the contact surface of the sample holder back to ensure a 10 mm air gap. The average sound absorption coefficient ($\mathrm{SAC}_{\mathrm{avg}}$) and noise reduction coefficient (NRC) were calculated from (1) and (2), respectively,

$$\mathrm{SAC}_{\mathrm{avg}} = \frac{\alpha_{125} + \alpha_{250} + \alpha_{500} + \alpha_{1000} + \alpha_{2000} + \alpha_{4000}}{6}, \tag{1}$$

$$\mathrm{NRC} = \frac{\alpha_{250} + \alpha_{500} + \alpha_{1000} + \alpha_{2000}}{4}, \tag{2}$$

where SAC is sound absorption coefficient, NRC is noise reduction coefficient, and α_n is sound absorption coefficient of "n^{th}" frequency.

3. Results and Discussion

3.1. Sound Absorption Properties of Individual Homogenous Fibers

3.1.1. Effect of Increasing Fiber Thickness on Sound Absorption Properties of Fibers. Figure 6 shows a consistent increase of SAC in the frequency range 100 Hz–1000 Hz across all sample thicknesses of finger millet straw fiber (F), namely, 10 mm, 20 mm, and 50 mm. SAC decreases in the frequency range 1000 Hz–2500 Hz; however, it resumes its upward trend in the frequency range 2500 Hz–3500 Hz. SAC further suffers fall in

Figure 1: Fiber extraction from finger millet straws at Bandipalya.

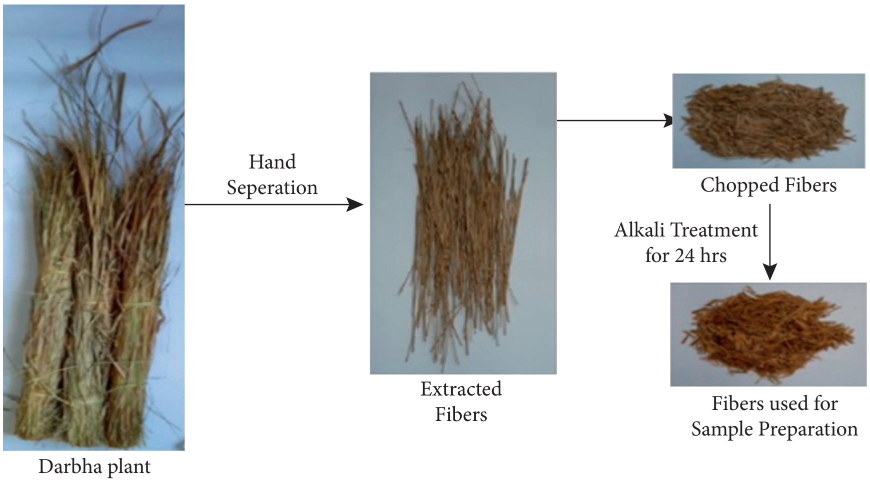


Figure 2: Darbha fiber extraction from darbha plant.

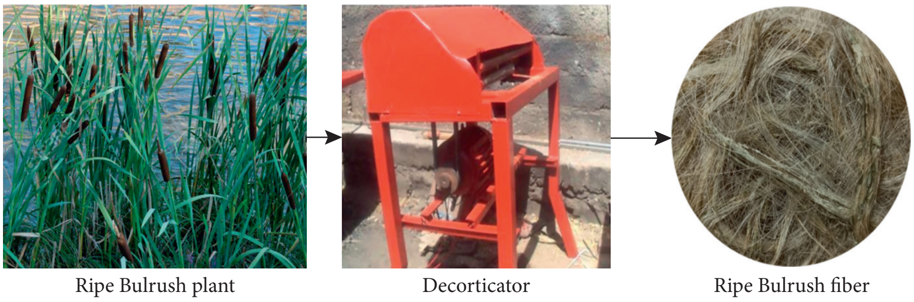


Figure 3: Fiber extraction from ripe bulrush.

the frequency range 3500 Hz–4500 Hz which may be related to fiber properties of finger millet straw fibers (F). Similarly, SAC is noticed for darbha (D) and ripe bulrush (R) fibers. However, with an exception in SAC value of D and R in the frequency range 3500 Hz–4500 Hz, SAC is consistently increasing higher values of 0.91. The possible reason for such an exceptional behavior of D and R fibers when compared to F fibers may be owed to the fact that F fibers have undergone double chemical treatment, i.e., 10% NaOH as common with D and R, along with peroxide treatment (performed only on F fibers). Thus, it can be summarized that all the fibers of F, D, and R have poor sound absorption properties in the common frequency range 1000 Hz–2500 Hz and hence this frequency range is considered a critical frequency range for analysis in this study.

The noise reduction coefficient (NRC) of F fibers has increased from 0.24 to 0.44 when the thickness was increased from 10 mm to 20 mm. Similarly, it increases from 0.44 to 0.78 when the thicknesses of the fibers are increased from 20 mm to 50 mm.

Table 1: Physical properties of fibers [14].

Fiber properties	Finger millet straw fiber (F)	Darbha fiber (D)	Ripe bulrush fiber (R)
Fiber length (mm)	80 ± 2.5	80 ± 4	80 ± 3
Fiber diameter (μm)	70 ± 15	60 ± 5	64.8 ± 12
Density (g/cm^3)	1.33 ± 0.1	1.07 ± 0.1	1.23 ± 0.1

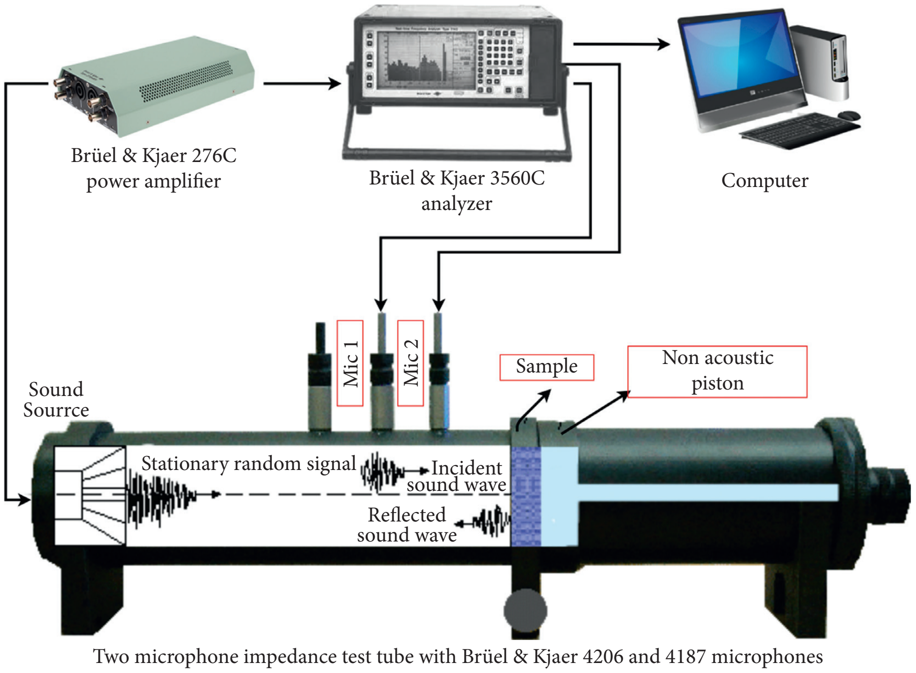


Figure 4: Impedance tube setup for testing sound absorption properties.

Table 2: Nomenclature of fiber combination.

Sample code	Fibers	Sample thickness
S1	Finger millet straw fiber (F)	50 mm
S2	Darbha fiber (D)	50 mm
S3	Ripe bulrush fiber (R)	50 mm
S4	Finger millet straw fiber/ripe bulrush fiber (F/R)	25 + 25 = 50 mm
S5	Ripe bulrush fiber/finger millet straw fiber (R/F)	25 + 25 = 50 mm
S6	Finger millet straw fiber/darbha fiber (F/D)	25 + 25 = 50 mm
S7	Darbha fiber/finger millet straw fiber (D/F)	25 + 25 = 50 mm
S8	Darbha fiber/ripe bulrush fiber (D/R)	25 + 25 = 50 mm
S9	Ripe bulrush fiber/darbha fiber (R/D)	25 + 25 = 50 mm
S10	Finger millet straw fiber + darbha fiber (F + D or D + F)	50 mm
S11	Finger millet straw fiber + ripe bulrush fiber (F + R or R + F))	50 mm
S12	Darbha fiber + ripe bulrush fiber (D + R or R + D)	50 mm

Note. / represents the position of the fiber; + represents mixed up fibers.

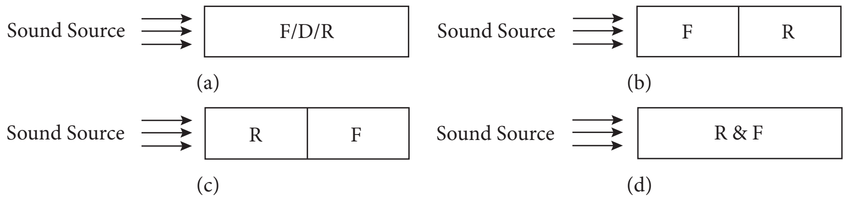
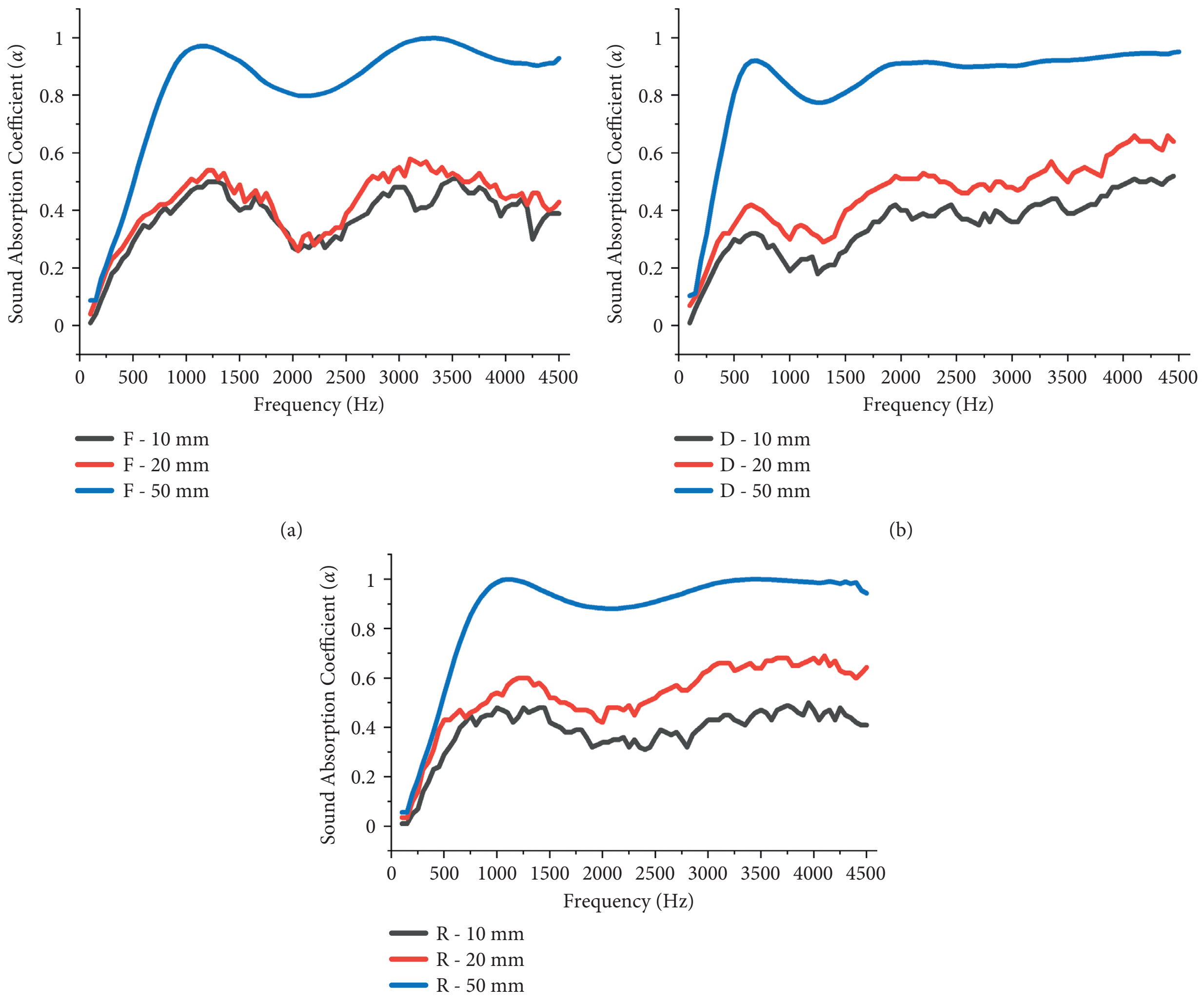


Figure 5: Fiber placement concerning the sound source. (a) Individual fibers: F or D or R. (b) F facing sound source and R behind F. (c) R facing sound source and F behind R. (d) R and F forming homogenous hybrid fiber combinations.



(a)

(b)

(c)

Figure 6: Sound absorption properties of individual fibers for a sample thickness of 10 mm, 20 mm, and 50 mm. (a) Finger millet straw fiber. (b) Darbha fiber. (c) Ripe bulrush fibers.

In the same way, the NRC was found for D fibers too. It was observed that NRC has risen from 0.23 to 0.38 for 10 mm to 20 mm. Again it gets peaked at 0.86 for D fibers when the thickness was increased to 50 mm. For R fibers, when fiber thickness is increased from 10 mm to 20 mm, the NRC was found to be 0.24 to 0.36, respectively. Hence, for 50 mm R fibers, an NRC of 0.84 was achieved. Considering the SAC values in the entire frequency range of 500 Hz–4500 Hz, the sound absorption coefficient for the sample with 50 mm thickness is found to be more than 0.8 when compared with SAC values of around 0.6 for 20 mm thickness and around 0.4 for 10 mm thickness for all the three types of individual fibers of F, D, and R. This has proven that the increase of thickness increases NRC values.

3.1.2. Effect of Adding Air Gap on Sound Absorption Properties of Fibers. The NRC for 50 mm fiber arrangements were found to be more efficient in absorbing sound than the 10 mm and 20 mm thicknesses samples. So, in Figure 7, the comparison on SAC of individual fibers without air gap and with an air gap of 10 mm was only depicted for 50 mm samples. The test results indicate a slight increase in values for all three individual fibers when a 10 mm air gap is provided between the fiber sample and the nonacoustic piston. Also, the NRC of fibers increased to 0.80 (for F fibers), 0.89 (for D fibers), and 0.87 (for R fibers). These results proved that the SAC value increased when an air gap was provided between the test sample and the sample holder. The same is true in the case for F, D, and R fibers also.

3.2. Sound Absorption Properties of Hybrid Combinations of Fibers

3.2.1. Sound Absorption Properties of Stacked Hybrid Combinations. This study aims to explain the effect of fiber type (F, D, and R fibers), thickness (50 mm), and stacking order on SAC values. As illustrated in Figure 8(a), the hybrid combination S4 (F/R) exhibits superior values of 0.87 SAC in the frequency range of 1000 Hz–2500 Hz when compared to the individual fiber samples S1 and S3. However, S5 (R/F) exhibits similar

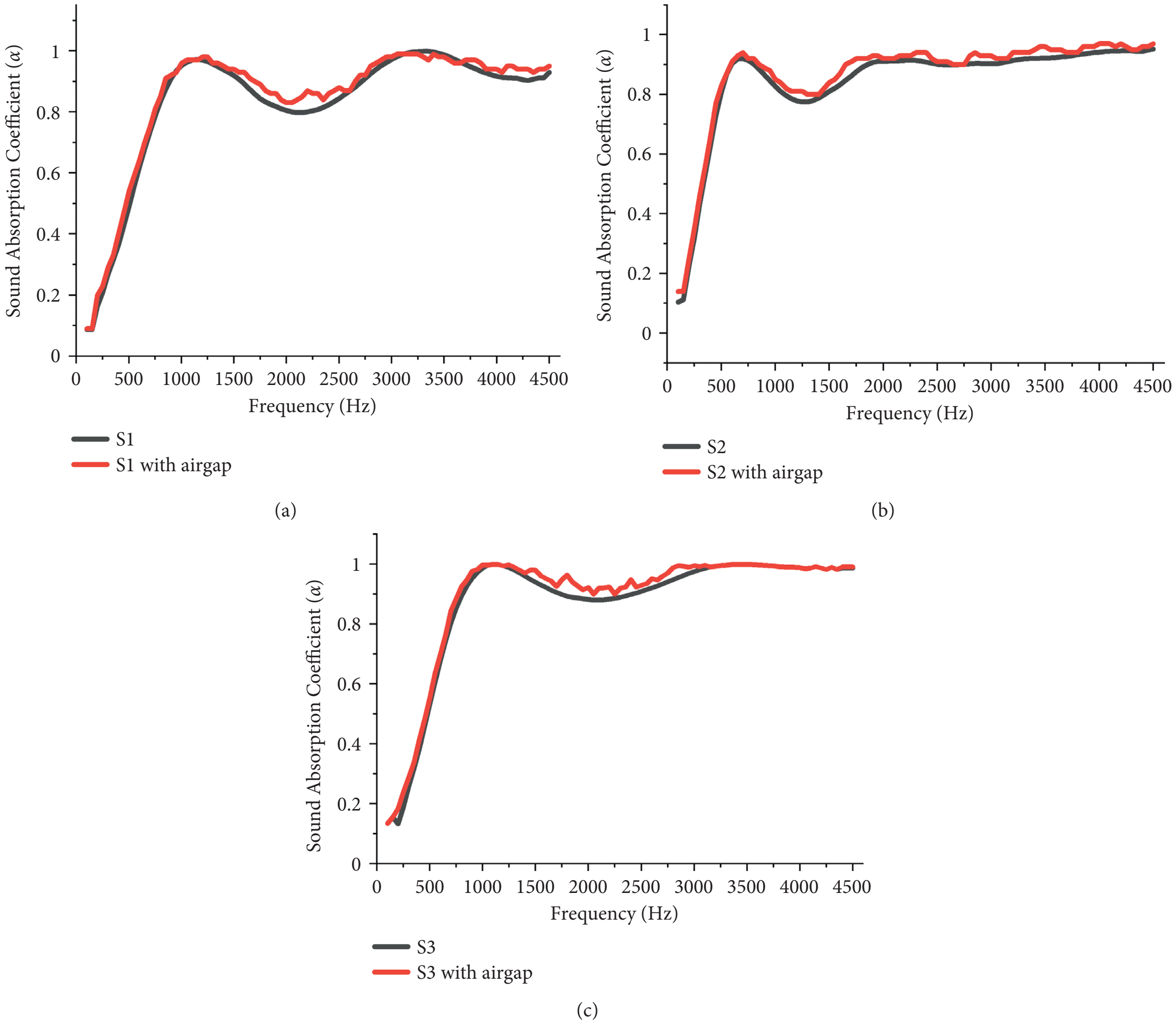


Figure 7: Sound absorption properties of individual fibers with and without air gap of 10 mm. (a) Finger millet straw fiber. (b) Darbha fiber. (c) Ripe bulrush fibers.

values to S1 and S3 in the frequency range of 1000 Hz–2500 Hz. In this combination, it is understood that if the finger millet faces the sound source, the SAC will tend to increase. Similarly, in S6 and S7 combination, S6 exhibited excellent absorption compared to S7 of 0.89 SAC in the frequency range 1000 Hz–2500 Hz as illustrated in Figure 8(b). The primary reason for this behavior of hybrid samples is that the absorption properties of the samples are dependent on the fiber type and stacked arrangement of the fibers exposed to the sound source. In the frequency range of 1000 Hz–2500 Hz, S8 and S9 exhibit similar acoustic properties of S2 and S3 samples, respectively, as illustrated in Figure 8(c). According to Figures 8(d) and 8(f), S9 (R/D) and S8 (D/R) have achieved higher sound absorption value than S5 (R/F) and S7 (D/F), respectively, for the frequency range 1000 Hz–2500 Hz. While both S9 (R/D) and S5 (R/F) exhibit similar absorption characteristics when R fibers are exposed to a sound source, they achieve greater absorption when D or F are exposed to a sound source. The reason for this difference in absorption behavior between S9 and S5 can be attributed to the fiber type (F or D) that supports the R fibers. However, there is only a slight difference in the sound absorption behavior of S7 (D/F) and S8 (D/R) and no significant difference in the sound absorption behavior of S4 (F/R) and S6 (F/D) for the aforementioned critical frequencies. From the SACs acquired for the different samples (S4 to S9), the NRC was calculated as 0.86 (S4 sample), 0.865 (S5 sample), 0.89 (S6 sample), 0.88 (S7 sample), 0.88 (S8 sample), and 0.90 (S9 sample), respectively.

3.2.2. Sound Absorption Properties of Homogenous Hybrid Fiber Combinations. As illustrated in Figures 9(a) and 9(c), the sound absorption performance of homogenous hybrid combinations (S10 and S12) are superior to that of individual fibers (S2) over the frequency range of 1000 Hz to 2500 Hz. The test results indicate that darbha fibers (D) have low sound

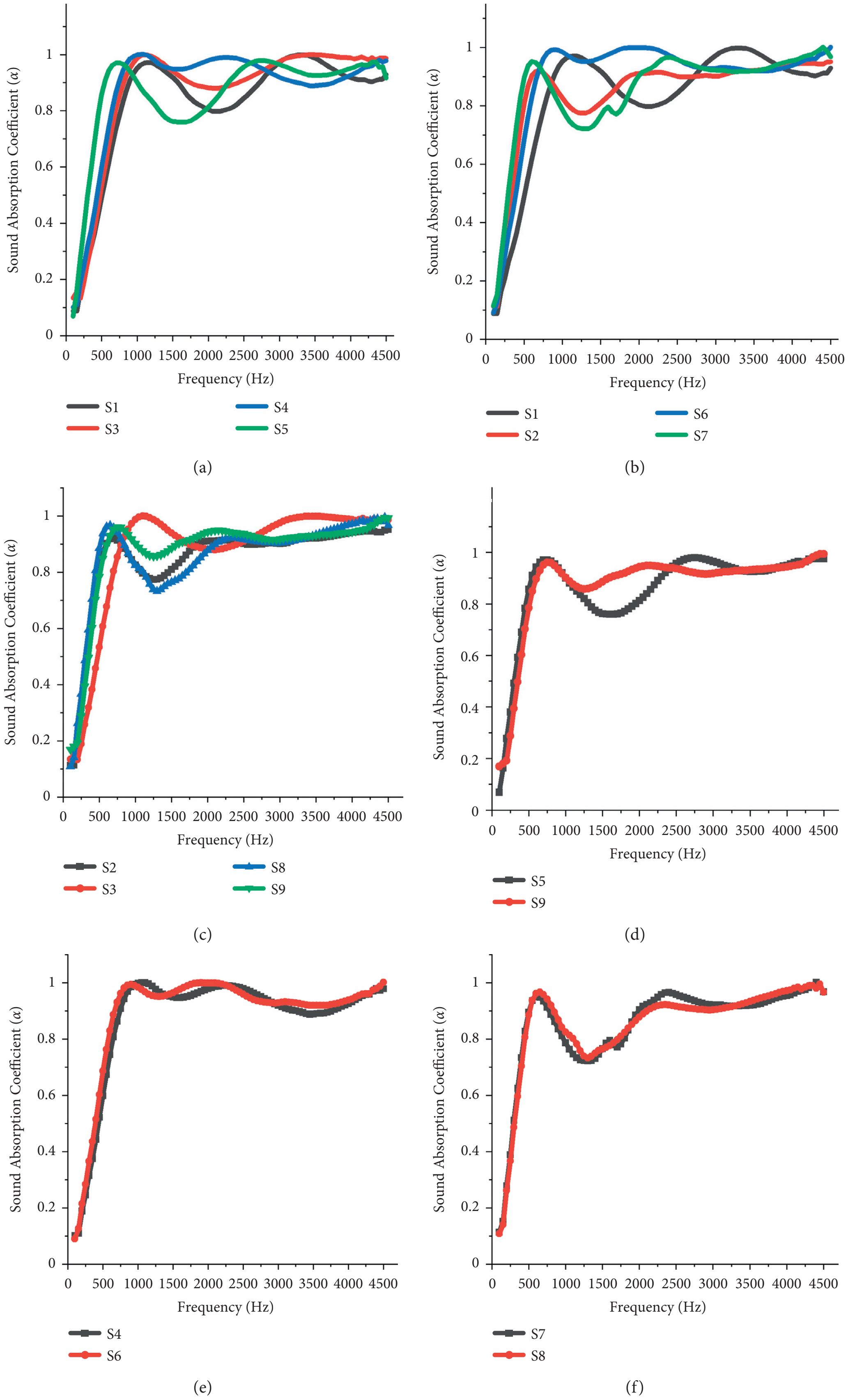


Figure 8: Comparison of sound absorption properties of various stacked hybrid combinations. (a) S4 and S5 with S1 and S3, (b) S6 and S7 with S1 and S2, (c) S8 and S9 with S2 and S3, (d) S5 with S9, (e) S4 with S6, and (f) S7 with S8.

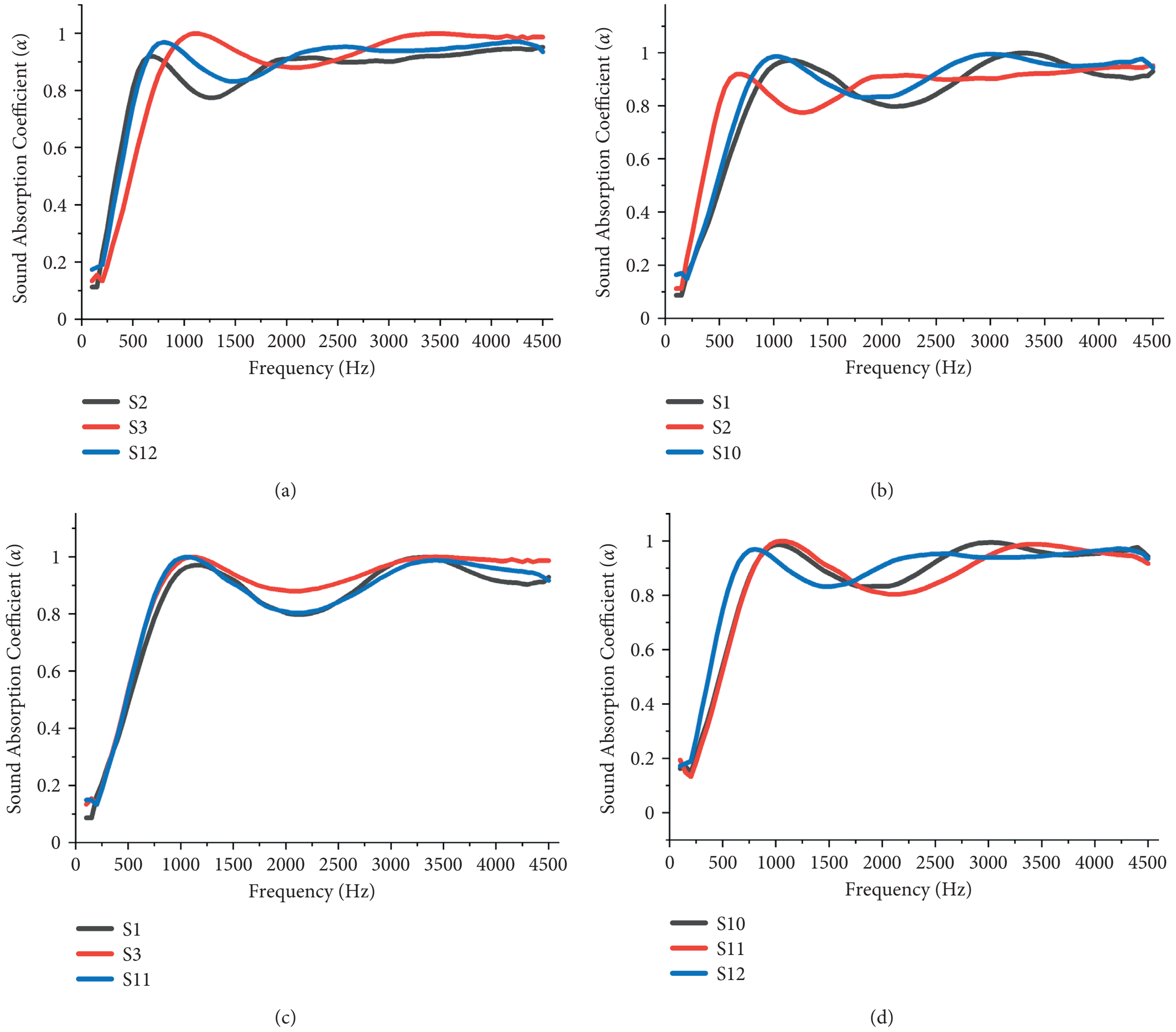


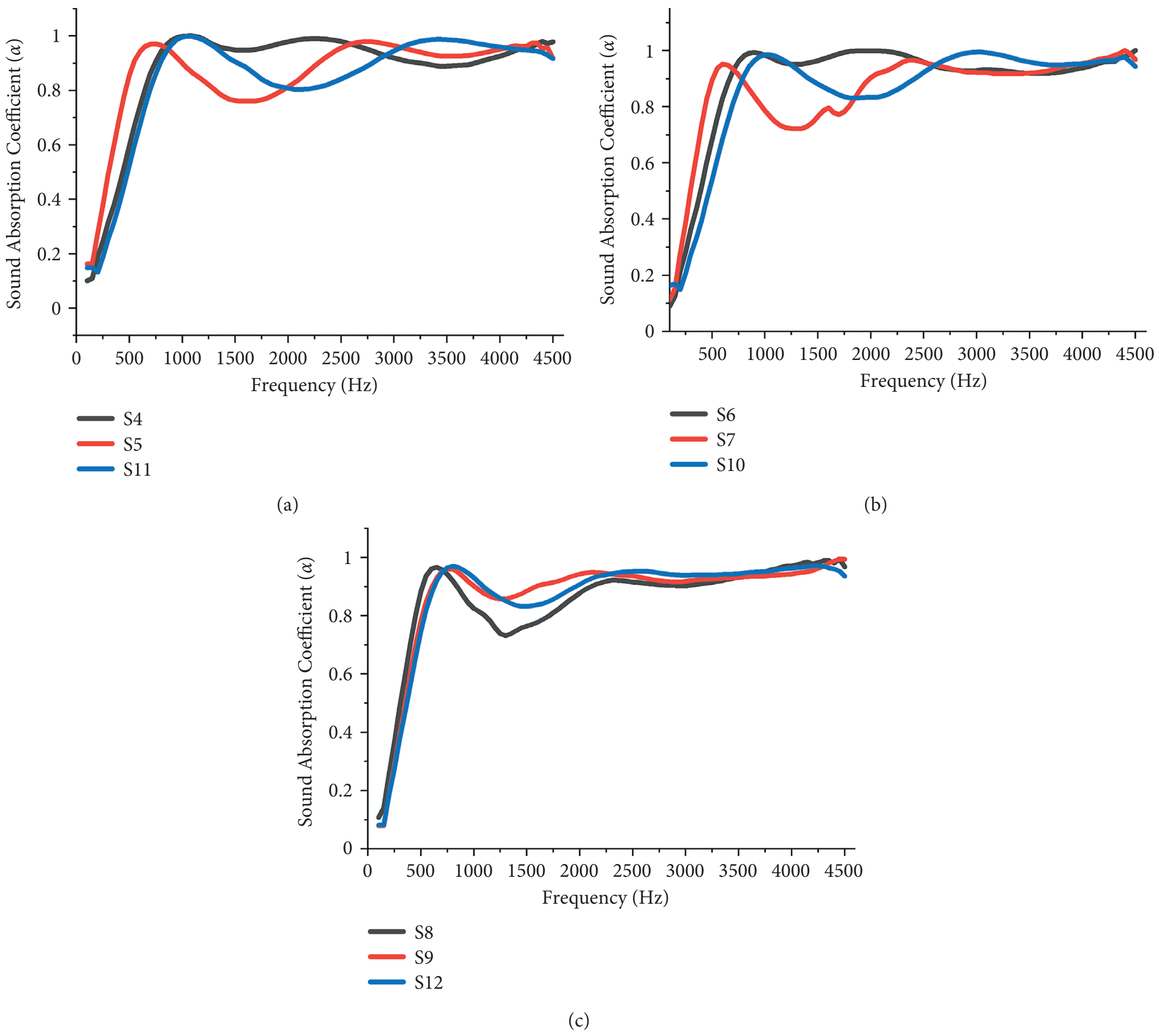
FIGURE 9: Comparison of sound absorption properties of homogenous hybrid fiber combinations with individual fibers: (a) S10 with S1 and S2, (b) S11 with S1 and S3, (c) S12 with S2 and S3, and (d) S10, S11, and S12 combinations (S1, S2, and S3). Furthermore, the sound absorption properties of homogenous hybrid combinations are comparable to those of stacked hybrid combinations.

absorption of 0.78 for individual fiber arrangement but exhibited superior acoustic properties when combined with F and R fibers. This proves that hybridization will tend to increase the SAC and NRC of the fibers. It can be concluded from Figures 9(a) and 9(b) that there is no significant difference in the sound absorption properties of finger millet straw fibers (F fibers) when used alone but in combination with D and R fibers it exhibited better SAC and NRC for the frequency range 1000 Hz–2500 Hz. This is experimentally studied also for the ripe bulrush fibers (R), whose results revealed that it exhibited superior sound absorption with F and D fibers than individual fiber arrangements as illustrated in Figures 9(b) and 9(c) for the critical frequency range of 1000 Hz–2500 Hz. However, the homogenous hybrid combination exhibits acceptable NRC values of 0.90, 0.91, and 0.93 for S10, S11, and S12.

3.2.3. *Comparison of Sound Absorption Properties of Stacked Hybrid Fiber Combinations with Their Homogenous Hybrid Fiber Combinations.*

NRC was found to be more for the homogenous combinations (S10, S11, and S12) in the critical frequency range than their individual fibers (S1, S2 and S3). As illustrated in Figure 10(a), S11 has superior absorption properties when compared to S4 and S5. Similarly, S10 and S12 have better sound absorption characteristics when compared with other combinations (S6-S7 and S8-S9), respectively. This happens due to the better interlocking of the fibers and the sound source finds it a critical path to travel along. Finally, the sound gets arrested or absorbed. The NRC for all the combinations is listed in Table 2 for better clarity. Table 3 lists the statistical data that represent the increase of NRC in percentage for all the combinations in Table 4.

The noise reduction coefficient (NRC) of previously published articles were compared with the present work for a better understanding of the increase in sound absorption properties as shown in Figure 11.



(a) (b) (c)

Figure 10: Comparison of sound absorption properties of stacked hybrid combinations of fibers with their homogenous hybrid combinations: (a) S4 and S5 with S11, (b) S6 and S7 with S10, and (c) S8 and S9 with S12.

Table 3: NRC for different fiber combinations.

Sample code	Fibers	Sample thickness	NRC
S1	Finger millet straw fiber (F)	50 mm	0.78
S2	Darbha fiber (D)	50 mm	0.86
S3	Ripe bulrush fiber (R)	50 mm	0.84
S4	Finger millet straw fiber/ripe bulrush fiber (F/R)	25 + 25 = 50 mm	0.86
S5	Ripe bulrush fiber/finger millet straw fiber (R/F)	25 + 25 = 50 mm	0.865
S6	Finger millet straw fiber/darbha fiber (F/D)	25 + 25 = 50 mm	0.89
S7	Darbha fiber/finger millet straw fiber (D/F)	25 + 25 = 50 mm	0.88
S8	Darbha fiber/ripe bulrush fiber (D/R)	25 + 25 = 50 mm	0.88
S9	Ripe bulrush fiber/darbha fiber (R/D)	25 + 25 = 50 mm	0.90
S10	Finger millet straw fiber + darbha fiber (F + D or D + F)	50 mm	0.90
S11	Finger millet straw fiber + ripe bulrush fiber (F + R or R + F))	50 mm	0.91
S12	Darbha fiber + ripe bulrush fiber (D + R or R + D)	50 mm	0.93

Table 4: Statistical data analysis: NRC increase percentage with comparison to all combinations.

Sample code	NRC	Increases in NRC values (%)											
		S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
S1	0.78	0	9.30	7.14	9.30	9.83	12.36	11.36	11.36	13.33	13.33	14.29	16.13
S2	0.86	10.26	0	2.38	0.00	0.58	3.37	2.27	2.27	4.44	4.44	5.49	7.53
S3	0.84	7.69	2.33	0	2.33	2.89	5.62	4.55	4.55	6.67	6.67	7.69	9.68
S4	0.86	10.26	0	2.38	0	0.58	3.37	2.27	2.27	4.44	4.44	5.49	7.53
S5	0.865	10.90	0.58	2.98	0.58	0	2.81	1.70	1.70	3.89	3.89	4.95	6.99
S6	0.89	14.10	3.49	5.95	3.49	2.89	0	1.14	1.14	1.11	1.11	2.20	4.30
S7	0.88	12.82	2.33	4.76	2.33	1.73	1.12	0	0	2.22	2.22	3.30	5.38
S8	0.88	12.82	2.33	4.76	2.33	1.73	1.12	0	0	2.22	2.22	3.30	5.38
S9	0.90	15.38	4.65	7.14	4.65	4.05	1.12	2.27	2.27	0	0	1.10	3.23
S10	0.90	15.38	4.65	7.14	4.65	4.05	1.12	2.27	2.27	0	0	1.10	3.23
S11	0.91	16.67	5.81	8.33	5.81	5.20	2.25	3.41	3.41	1.11	1.11	0	2.15
S12	0.93	19.23	8.14	10.71	8.14	7.51	4.49	5.68	5.68	3.33	3.33	2.20	0

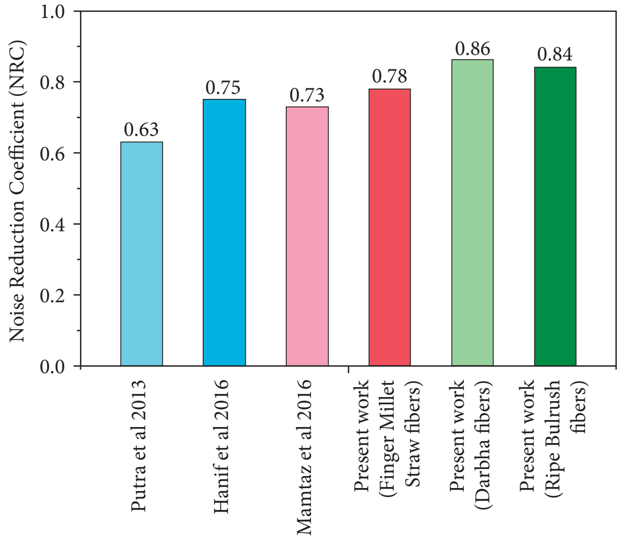


Figure 11: Comparison of sound absorption properties of present work with previously published work [6, 26, 34].

4. Conclusion

The sound absorption properties of three plant-based natural fibers, finger millet straw fiber (F), darbha fiber (D), and ripe bulrush fibers (R), are investigated in this research study. At first, fibers are studied individually, secondly in stack-up hybrid combination, and finally in homogenous hybrid combinations. The SAC values of individual fibers F, D, and R increased significantly with an increase in sample thickness from 10 mm to 20 mm and then for 50 mm and also increased with the addition of an air gap of 10 mm between fiber sample and sample holder. Additionally, test results indicate that the fiber type and stack-up arrangement of the fibers play a significant role in determining the sound absorption properties (SAC and NRC). Darbha fibers (D) exhibited superior sound absorption of 0.86 NRC as an individual fiber arrangement. Also, darbha fibers when added with ripe bulrush fibers either in stacked up and homogenous hybrid combinations exhibited superior sound absorption compared to the other combinations in the critical frequency range of 500 Hz–2000 Hz. From these studies, it was concluded that the darbha fiber will help to enhance the sound absorption properties either individually or in hybrid combinations.

Data Availability

The data used to support the findings of this study are included in the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this article.

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