

A Review of Cellulosic Fibre Surface Modification Techniques - The Case of Ramie

Bewuket Teshome Wagaye, Luc The Nguyen, Biruk Fentahun Adamu, Adane Dagnaw Gudayu, Jiansheng Guo

How to cite: Wagaye BT, Nguyen LT, Biruk Adamu BF, Gudayu AD, Guo J. A Review of Cellulosic Fibre Surface Modification Techniques - The Case of Ramie. Textile & Leather Review. 2024; 7:1061-1095. <https://doi.org/10.31881/TLR.2024.069>

How to link: <https://doi.org/10.31881/TLR.2024.069>

Published: 11 June 2024



A Review of Cellulosic Fibre Surface Modification Techniques - The Case of Ramie

Bewuket Teshome WAGAYE^{1,2}, Luc The NGUYEN³, Biruk Fentahun ADAMU^{1,2}, Adane Dagnaw GUDAYU², Jiansheng GUO^{1*}

¹Key Laboratory of Textile Science & Technology, Ministry of Education, College of Textiles, Donghua University, Shanghai, 201620, China

²Textile Engineering Department, Ethiopian Institute of Textile and Fashion Technology, Bahir Dar University, Bahir Dar, Ethiopia

³Faculty of Garment Technology and Fashion Design, Hung Yen University of Technology and Education, Hai Duong, Vietnam

*jsguo@dhu.edu.cn

Review

<https://doi.org/10.31881/TLR.2024.069>

Received 16 April 2024; Accepted 7 June 2024; Published 11 June 2024

ABSTRACT

The demand for cellulosic fibres as a reinforcement in polymers is growing due to their renewable, biodegradable, and ecologically favourable properties. In addition, they are lower in density than e.g. glass fibres, which will help in making lightweight composites. Besides, there is no health hazard with ease of processing and minimum damage to the processing equipment. Despite their advantages and high demand, their utilization in industrial applications is still limited. This is due to their inherent polar and hydrophilic nature and their consequent incompatibility with hydrophobic polymers. Therefore, it is often necessary to do hydrophobic modification treatment. The review examines a range of strategies for modifying ramie surfaces, encompassing physical, chemical, and biological processes. It also analyzes the procedures and techniques involved in the preparation and manufacturing of ramie-reinforced polymeric biocomposites. Moreover, an investigation is conducted to evaluate the performance of the composites produced. The performance evaluation includes both before and following fibre treatment, covering surface morphology, wettability, chemical composition, mechanical properties, and flame retardancy. Most of the researchers reported that alkaline and silane coupling agent treatments are the most prevalently applied pretreatments. Compared to biological and physical methods, the application of chemical agents enables excellent adhesion of fibre to polymer resins. However, the utilization of solvents such as ethanol and methanol increases the amount of waste produced and toxic compounds released.

KEYWORDS

ramie, surface modification, adhesion, biocomposites, properties

INTRODUCTION

The depletion of nonrenewable resources, demand for lightweight materials, and environmental pollution drive interest in renewable and biodegradable raw materials [1-4]. Natural fibre-reinforced composites are promising substitutes for petroleum-based composites in structural applications [5]. In this regard, cellulosic fibres like ramie, flax, hemp, and sisal gain attention as polymer

reinforcements due to their renewability, biodegradability, abundance, high strength- to-weight ratio, thermal stability and acoustic insulating properties [6,7]. With favourable functional properties and economic implications [8], they are suitable for biocomposites [3,9-12]. Moreover, increasing demand for eco-friendly goods in various applications, including electrical, car, marine, and aerospace, prompts researchers to use natural fibres in polymer-based composites [13-15].

Biocomposites contain either a bio-fibre dispersion phase or a matrix phase derived from renewable resources. It is fully bio-based when both phases come from biological sources, which is considered to be a green composite [16]. Green composites, featuring moulding ability and recyclability, combine biodegradable fibre with biodegradable resin, representing a focus in new material fields [17].

Several factors affect biocomposite production. Crucial factors include fibre-matrix adhesion, matrix and fibre modification, hybrid strategy, and processing approach for high-performance biocomposites [18]. Excellent interfacial adhesion is vital for fibre reinforcing efficiency and bulk mechanical performance of composites [14]. Natural fibres have limitations such as inherent polar and hydrophilic nature, limiting compatibility with hydrophobic polymeric matrices [12,19-23]. This hampers uniform fibre dispersion, leading to efficiency restrictions. Moisture absorption by natural fibres causes swelling, voids, and poor mechanical properties in composites [21]. Water absorption may create aggregates, weakening biocomposites' dimensional stability [16]. Furthermore, there are issues regarding decreased thermal stability and inconsistent quality [10]. Lignin and waxy compounds in natural fibres cover reactive groups, hindering interlocking with polymer matrices [24]. Thermal decomposition of natural fibres starts above 220 °C, limiting processing temperature and choices of polymer matrices [19,21]. Modifications aim to enhance natural fibre adherence by altering surface characteristics. Exemplary strength and stiffness could be achieved with a strong interface between fibre and matrix. Matrix-to-fibre stress transfer efficiency could be reduced with a weaker interface [25].

A diffusion or reaction zone forms at the fibre-resin interface in either chemically or mechanically mixed phases. Interfacial adhesion significantly influences composite mechanical properties. Inadequate adhesion across the phase boundary results in weak force dispersion and poor mechanical characteristics [24,26]. Efficient modification of cellulose is crucial for controlling properties like hydrophobicity and adhesion to synthetic polymers [11]. Surface energy, indicating the fibre's adhesive qualities, is commonly used to assess interface adhesion [27].

Various methods-chemical, physical, and biological-can modify natural fibre surfaces [28,29]. Mechanisms include associating polar groups or mechanical interlocking with the polymer backbone. Chemical treatments like mercerization, acetylation, etherification, silanization, benzylation, grafting,

and the insertion of compatibilizing agents improve cellulose fibres by chemically modifying their structure through reactions with the fibres [22,30-32]. Enhancing composites' mechanical performance involves adding high specific surface area to natural fibres in polymer matrices [33]. Chemical treatments are designed to improve hydrophobicity and surface roughness, hence enhancing the bonding between the matrix and fibres [34]. Moreover, methods to improve the adherence between the two phases (adhesion promoters) are frequently needed because of limited compatibility between the reinforcement and resin [16]. Adhesion promoters reduce water absorption and enhance process-ability. Physical processes like stretching, calendering, thermo-treatment, and electric discharge (corona, cold plasma) alter fibre properties, enhancing mechanical attachment without changing chemical composition [21].

This paper provides a comprehensive review of the physical and chemical properties of ramie fibre. It explores various surface modification techniques for ramie fibre, evaluating chemical, physical and biological methods. The study also explores the fabrication methods employed in the development of ramie/polylactic acid (PLA) biocomposites. Additionally, a thorough comparison is made to assess the enhancements achieved in the physical, mechanical, thermal, and chemical properties of these biocomposites. The study also looks at how well the matrices bond with the interfacial surfaces, giving a full picture of the performance of ramie/PLA biocomposites.

RAMIE FIBRE PROPERTIES

Ramie is an enduring plant that naturally occurs in China, Japan, and the Malay Peninsula serving as a textile fibre for generations. Ramie belongs to the nettle family, genus *Boehmeria nivea* [35]. A 1-hectare farm produces 880–2200 kg of ramie, which has 3.5% fibre by weight. Growing in hot and humid areas takes 45–55 days. The plant has large, heart-shaped leaves on an almost branchless stem, reaching 1–2.5 m tall. Figure 1 illustrates the image of the ramie plant. The plant's stem and leaves contain approximately 80% moisture [36-38]. Fishing nets, sewing thread, canvas, upholstery, fire hoses, and garments have been made from ramie. China annually exports a substantial volume of ramie yarn and fabrics. Japan and Europe are the primary export markets for ramie products [39]. Additionally, ramie finds use in polymer composites for reinforcement and in ballistic applications [40]. The chemical structure of cellulose, hemicellulose, and lignin is presented in Figure 2. Cellulose is the major component of natural bast fibres. It produces fibres that are surrounded by a mixture of structural biopolymers, primarily hemicellulose and lignin. Cellulose polymer formation involves linked repeat units, termed the degree of polymerization (DP), a critical property in cellulosic fibres. DP varies among fibres; cotton (7000 DP), ramie (6500 DP), and flax (8000 DP) are common, with the high DP

[41]. Ramie fibres exhibit strong but inextensible characteristics due to well-oriented crystallites parallel to the fibre axis [42]. The tensile strength of natural fibres is determined by the DP of the cellulose, the overall cellulose content in the fibres, and the micro-fibril angles [2]. Ramie surpasses flax and jute in tensile strength, nearing C-glass fibre in modulus and tensile strength [43], with comparable specific modulus and strength to synthetic fibres for composites [37,38]. The mechanical properties of ramie are similar to those of sisal [44]. It resists bacteria, fungi, mildew, and insects, and exhibits stability in alkaline and mild acid conditions [38,45].

Ramie, the longest and strongest vegetable fibre, exhibits a flat and ribbon-like shape [46] with multiple rounded cross-sections instead of a simple round, oval, or flat shape either with or without a lumen [47]. Available in continuous form, its length ranges from 60 to 250 mm [48]. Comparatively longer and coarser than flax, ramie possesses outstanding thermal conductivity but its strong polarity of the fibre leads to increased hydrophilicity, and larger expansion in humid conditions [49].



Figure 1. Ramie plant image [38]

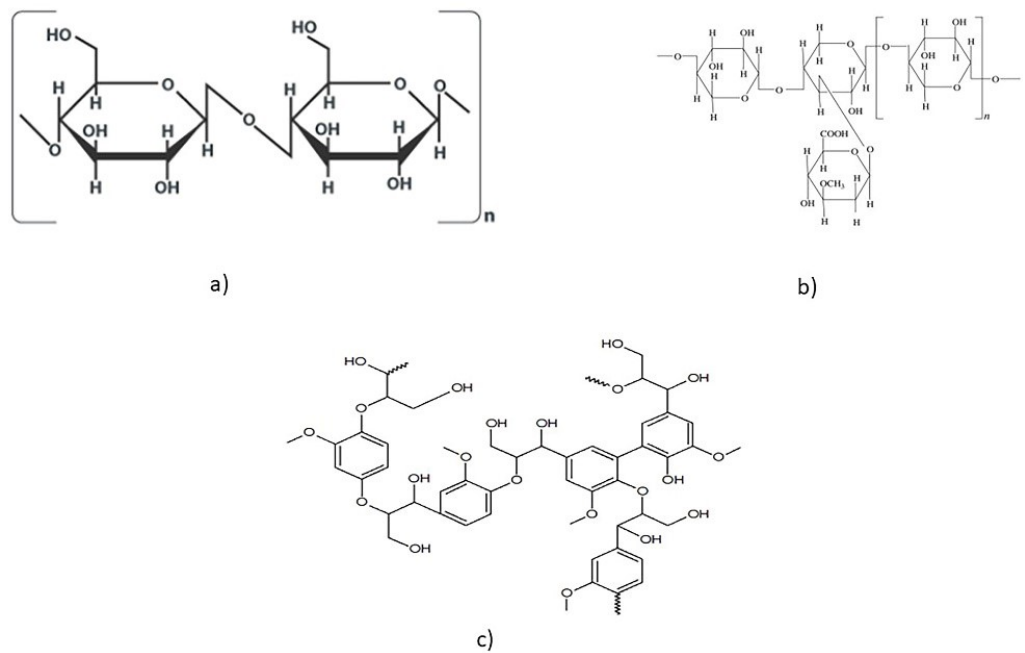


Figure 2. Chemical structures of (a) cellulose, (b) hemicelluloses, and (c) lignin

Ramie is a suitable reinforcement in fibre-reinforced composites among natural cellulosic fibres, offering low specific density, good mechanical properties, high tenacity, crystallinity, and moisture resistance [2, 50-58]. The key properties of ramie fibres are given in Table 1.

Table 1. Physical, chemical and mechanical properties of Ramie fibre [2,50-57]

Properties	Ramie
Cellulose content (wt.%)	68.6-76.2
Hemicellulose content (wt.%)	13.1-16.7
Lignin content (wt.%)	0.6-0.7
Pectin (wt.%)	1.9
Waxes (wt.%)	0.3
Extractives (%)	1.9-2.2
Moisture (wt.%)	12–17
Cellulose crystallinity	64
Microfibrillar angle (°)	8
Mean length (mm)	12-15
Diameter (µm)	25.5
Density (g/cm³)	1.4-1.55
Tensile strength (MPa)	200-1000
Elongation (%)	1.2-4
Tensile Modulus (GPa)	41-130
Specific modulus (GPa cm³/g)	27.3-81
Aspect ratio	2000-6000
Degree of crystallinity (%)	70

SURFACE MODIFICATION OF RAMIE FIBRES

Good interfacial adhesion can be achieved by fibre modification, the use of interface-active additives, or matrix modification [59]. The classification of surface modification treatments for cellulosic fibres is presented in Figure 3, which may be physical, chemical, or biological. The chemical methods involve the use of chemical agents such as acids, alkalis, and coupling agents. The physical methods include plasma treatment, corona discharge, and ultraviolet (UV) irradiation. The biological methods involve the use of enzymes. Common chemical, physical and biological modification treatment systems used to improve the adhesion between ramie fibre and polymeric matrices are shown in Table 2 and Table 3. It is important to remember that not all treatments are sustainable [60].

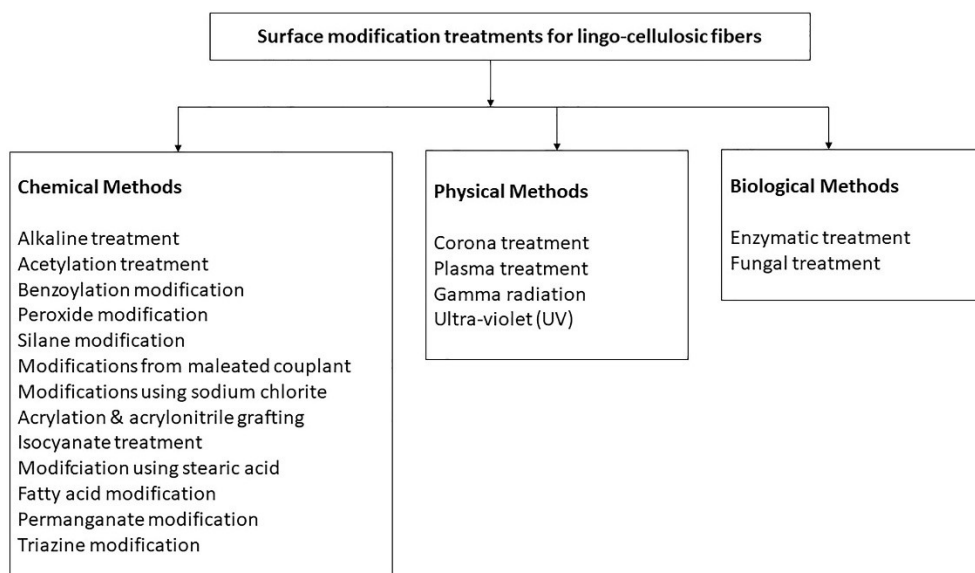


Figure 3. Classification of the surface modification treatments

Table 2. Chemical treatments summarized for improving interfacial adhesion between ramie and polymer matrices

Chemical treatment on ramie fibre	Matrix used	Improvements in the properties	References
<i>Alkali treatment</i> (5% alkali with DAP for 2 h)	PLA (thermoplastic)	H decreased from 5.9 ± 1.3 to 2.0 ± 0.8 mJ/m ² , W of alkali/silane/DAP-treated was higher ($95.4^\circ \pm 1.3^\circ$)	[27]
<i>Alkali treatment</i> (1% alkali for 1 h, 3% silane for 2 h)	PLA (thermoset)	CA increased by 14.26% (silane) and 33.12% (alkali-silane), A decreased by 23.70%	[49]
8% alkali for 4 h	PLA (thermoset)	Increased 39% TS, IS 43% and FS 35% as ramie content increases from 37 to 48%	[50]
0.4% KMnO ₄ and 0.08% silane in acetone solution	PLA (thermoplastic)	Increased 20% TS, FS also improved	[61]

Chemical treatment on ramie fibre	Matrix used	Improvements in the properties	References
<i>Alkali treatment</i> (5% alkali for 3 h and 6% silane in acetone for 24 h)	PLA (thermoplastic)	TS were 64.24 MPa (composite treated by NaOH)	[43]
0.08% permanganate and silane	PLA (thermoplastic)	Fibre surface becomes rougher	[52]
<i>Alkali treatment</i> (5% alkali for 1 h and cyclic loading)	PLA (thermoplastic)	Considerable tensile failure load and smallest failure deformation	[35]
10% NaOH for 24 h, 10% KH550 and KH570	Vinyl resin	TS improved by 49.29% and IS increased by 38.28%	[62]
1% APP	Benzoxazine	Increased limiting oxygen index (LOI)	[63]
5% NaOH for 1 h, 1% silane for 1 h, and 4% of dicumyl peroxide	PLA (thermoplastic)	Increased thermal stability	[64]
ESO at 160 °C in argon gas	Polypropylene	Better interfacial bonding	[65]
Acetylation for 1 h, 1% silane, 4% peroxide for 30 min	AESO	Acetyl groups substituted the OH groups and the fibre surface was covered with nonpolar groups	[66]
Silanized CNT (0.1% - 1%)	Epoxy	The Fibre surface showed more flaws and roughness	[67]
Compatibilized by maleic anhydride	PLA (thermoplastic)	Improvements in mechanical properties	[68]
10% NaOH at 60 °C, 4 h	Cellulose	Three crystal peaks shifted to 12.1°, 19.8° and 22.0°	[69]
2, 5 and 10% NaOH	Polyester	A peak at 1730 cm ⁻¹ disappeared	[70]
5% NaOH for 2 h, 6% silane, 5% APP for 2 h	Epoxy	Increased surface energy and dispersed component of ramie fabric, showing enhancement in non-polarity	[71]
5% NaOH for 2 h	Epoxy	Ramie fibres become looser	[72]

A: Absorption; W: Wettability; H: hydrophilicity; CA: Contact angles; TS: Tensile strength; FS: flexural strength; IS: impact strength; DAP: Diammonium phosphate; APP: Ammonium polyphosphate; ESO: Epoxy-silicone oil; AESO: Acetylated epoxidized soybean oil; CNT: Carbon nanotube

Table 3. Physical treatments summarized for improving interfacial adhesion between ramie and polymer matrices

Physical treatment of ramie fibres to improve adhesion	Matrix used	Improvements in the properties	References
Trans-crystallization by Stereocomplex Crystals	Poly lactide	Higher crystallinity than the common spherulites	[23]
Atmospheric pressure plasma jet with ethanol pretreatment for 10 min	Polypropylene	Larger advancing contact angles, 50% increased IFSS	[73]
Low-temperature plasma treatment for 1 min-300 W and 3 min-100 W	-	Increased surface energy 117.0 and 122.9% respectively	[74]
Physical treatment of ramie fibres to improve adhesion	Matrix used	Improvements in the properties	References

Atmospheric pressure plasma treatment with ethanol pretreatment isopropanol and n-butanol and then atmospheric pressure plasma	Polypropylene	Increased interfacial adhesion by 26%	[75]
	Polypropylene	Increased surface roughness	[76]
Plasma treatment 3 min and 200 W	Epoxy	Increased surface energy and adhesion 124.5 and 59.1%	[77]
Low-pressure plasma at 100 W, pressure 0.3 mbar (1, 2, 3, and 4 min)	Phenolic resin	1 min treated shows a smaller angle, 38°±2.2 than untreated fibres 49°± 1.9	[78]

IFSS: Interfacial shear strength

Chemical Methods

Chemical modification is the most commonly used method for surface modification. In this method, the fibre is treated with a chemical agent to introduce functional groups on its surface. Functional groups boost fibre-matrix adhesion, improving composite mechanical characteristics. Commonly used chemical agents for ramie fibre modification include alkali, silane coupling agents, diammonium phosphate (DAP), Ammonium Phosphate (APP), and Poly (Ethylene Oxide) (PEO) [79].

Alkali Treatment

Alkali treatment alters the surface shape and composition of the fibres. This changes the mechanical properties of fibre-reinforced composites, which makes the adhesion between the fibres much better [45]. Cellulose undergoes chemical, physicochemical, and structural modifications during the treatment, and the optimal modifications depend on the concentration of alkali, time, tension, and temperature [80]. Mercerizing cellulose increases amorphous cellulose at the expense of crystalline. Hydrogen bond loss from the network structure may be the biggest alteration. Alkali-cellulose is generated when sodium hydroxide penetrates the crystalline areas of native cellulose (cellulose-I). Following the removal of unreacted NaOH, regenerated cellulose (cellulose-II) is formed [81]. The transformation of crystalline native cellulose into hydrate cellulose (cellulose-II) is known to proceed by the formation of intermediate crystalline soda-cellulose complexes [82]. Alkaline treatment also helps to remove lignin, waxes, and fatty acids from cell walls; in addition, it increases the number of reaction centres on the fibre surface as a result of the hydrogen bonds breaking [83]. The cellulose-I turns into one of a set of soda-cellulose when ramie fibres are mercerized without tension in a caustic soda solution at room temperature. Soda-cellulose decomposes into water-cellulose when the alkali is removed by washing in water at room temperature. Upon drying, the water-cellulose undergoes a transformation into cellulose II, a phenomenon observed in mercerized and regenerated fibres [84]. Under alkaline conditions, hydrogen peroxide can also eliminate surface impurities, hemicellulose, and lignin [85].

Silane Coupling Agent Treatment

Adhesion promoters are chemical coupling agents that work at the interface of organic and inorganic surfaces to improve adhesion between the two materials. Adhesion promoters contain dual functions in their molecular structure. A metallic core atom, such as silicon, zirconium, titanium, aluminium, and others, will offer the adhesion promoter inorganic reactivity, especially if the metal atom is connected to methoxy, ethoxy, or hydroxyl groups. A silicon-based chemical that acts as an adhesion promoter is known as a silane coupling agent [86]. When applied to a surface, the alkoxy groups on the silane coupling agent will react with the surface to form covalent bonds, while the organic functional group will bond with the polymer matrix. This creates a strong chemical bond between the two materials, which enhances the mechanical properties of the resulting composite.

Silane treatment can introduce new functional groups onto the fibre surface besides just affecting wettability. Depending on the chosen silane type, these new functionalities can offer additional benefits. For example, some silanes might improve the fibre's compatibility with specific fillers or additives used in the composite formulation. Therefore, silane treatment modifies the fibre surface from a hydrophilic, less reactive state to a more hydrophobic and chemically reactive one. This improves the bonding between the fibre and the surrounding matrix, leading to better performance in composite materials [87].

Diammonium Phosphate (DAP) or Ammonium Phosphate (APP) Treatment

A large variety of phosphorous compounds has been used to flame-retard cellulose-based fibres. The group of organophosphorus chemicals comprises Pyrovatex CP, diammonium phosphate, phosphoric acid, tributyl phosphate, triallyl phosphate, and triallyl phosphoric triamide [88]. The addition of DAP treatment to the surface of ramie fibres reduced the wettability of ramie fibre-PLA composites. The polar portion of surface energy rose from 2.1 mJ/m² to 5.1 mJ/m² after a single DAP treatment of ramie fibres. The amine and hydroxyl groups in DAP can form a covalent link with the carboxyl groups in the PLA matrix. The presence of the NH group at 3270 cm⁻¹ in the FTIR spectra of ramie fibre-PLA composites is an indication of DAP treatment of ramie fibres [89].

The influence of ammonium polyphosphate (APP) on the flame retardancy and mechanical properties of ramie fibre-reinforced poly(lactic acid) (PLA) biocomposites is also significant. APP is known to be an effective flame retardant due to its ability to release phosphoric acid, polyphosphoric acid, and non-flammable gases when exposed to high temperatures. This acid contributes to the dehydration and charring of the ramie fibres, thereby improving the flame retardancy of the composites. Adding APP to natural fibre-reinforced PLA biocomposites can change their mechanical properties, especially when the temperature is between 50 and 60 °C. The presence of APP can lead to a decrease in storage

modulus, indicating a change in the material's load-bearing capacity. Furthermore, APP loading may disrupt the compatibility between PLA and fibres. However, composites made with APP have shown improved flame retardancy and comparatively better mechanical properties [90].

Functionalization of Cellulosic Fibres with an Aqueous Solution of Poly (Ethylene Oxide) (PEO)

The functionalization of cellulosic fibres with an aqueous solution of PEO, followed by extrusion with PLA and injection moulding, was used to create completely green biocomposites of PLA and cellulose fibres [91]. PEO is a water-soluble polymer that is widely used as a coating agent due to its ability to reduce the surface energy of substrates. When PEO is applied to the surface of cellulosic fibres, it forms a thin coating layer that reduces the interaction between the fibres and water molecules. This decreases fibre swelling in water and enhances tensile strength and elongation. The functionalization process typically involves immersing the cellulosic fibres in an aqueous solution of PEO for a specified time. The concentration of PEO, immersion time, and solution temperature can all affect the degree of functionalization and the resulting properties of the fibres [91].

Biological Methods

Biological methods of cellulosic fibre treatments involve utilizing microorganisms or enzymes derived from them to modify the properties of the fibres. Enzymes like pectinase and cellulase are used to break down non-cellulosic components like pectin and hemicellulose present in the fibre. This process improves fibre purity and increases surface area, leading to reduced wettability and improved adhesion properties [60].

Cellulase Enzyme-assisted Biopolishing

Ramie fibres were polished for five days using *Streptomyces albaduncus* bacteria at pH 7.4 and 2.0 g glucose. Eliminating gum and tiny fibrils from the fibre surface increased softness and brightness. The thermal stability and crystalline structure of ramie fibres were both reduced by bacterial treatment. Modified fibres have a completely different morphology from the original fibres. The surface becomes soft and bright due to extracellular protuberant structures. Because cellulase enzyme treatment changes the morphology of the fibres, it can be employed as a reinforcing material in polymer composites to improve interfacial adhesion between fibres and matrix [92]. Debeli et al. used 10 grams of pectinase enzyme by dissolving it in 500 mL of warm water (45.6 °C). The solution's pH was adjusted between 4 and 5 using acetic acid and a small amount of sodium carbonate solution. The ramie fibres were then added to this activated enzyme solution for 2 hours to degrade pectin, a natural fibre component. The application of pectinase enzyme treatments proved to be a more efficient method for generating hydrophilic ramie samples. The utilization of pectinase enzyme in fibre surface

treatment is safe due to its environmentally friendly characteristics [27]. Specific fungi or bacteria are also employed to selectively eliminate non-cellulose substances such as ash, wax, and extractives from the surface of the fibre by the action of specific enzymes. White rot fungus produces oxidases, which are enzymes that react with lignin components. This reaction reduces the amount of lignin on the surface of the fibres but enhances the hemicellulose content and solubility. As a result, the hydrophobic nature of cellulose fibres is reduced [41].

Physical Methods

Physical methods are also effective in the surface modification of ramie fibre. Corona, cold plasma, surface fibrillation, stretching, thermo-treatment, and electric discharge are physical methods. The surface of natural polymers can be modified without compromising their bulk properties using cold plasma [81]. The surface energy of cellulose fibre is altered during corona treatment. Various surface alterations have been accomplished depending on the type and nature of the employed gases. Surface crosslinks may be added, surface energy could be enhanced or lowered, and reactive-free radicals could be generated [93].

Irradiation Treatment

Plasma treatment introduces functional groups on the fibre's surface by low-pressure plasma. Corona discharge creates active spots on fibre surfaces by exposing it to a high-voltage corona. Plasma contains charged, excited, and neutral atoms, molecules, radicals, and photons [94]. Plasma is a hot, ionized gas. Ionizing energy creates crosslinks and free radicals on materials, improving fibre-polymer matrix adhesion. Ionizing radiation like gamma is thought to have high energy because the Compton Effect deposits energy on the surface. Several studies agree that increasing gamma irradiation dose improves composite material mechanical properties till decay. Irradiation liberates hydroxyl groups from cellulose monomers' carbons 2, 3, and 4 because cellulosic structures make up most of the fibre. Heavy particles (gas molecules and ions) are at ambient temperature in a low-pressure, high-frequency discharge, yet electrons have enough kinetic energy to break covalent bonds and ionize [26]. Free radicals also result from carbon-carbon bond breaking. Ionizing radiation produces ions, peroxides, and free radicals. These features create active locations for fibre-polymer matrix interaction [60]. Plasma alters textile surfaces without changing bulk qualities in a dry, environmentally- and worker-friendly manner [95]. It requires no water or chemicals [96,97].

Cyclic Loading of Ramie Fabrics

Cyclic loading applies recurrent or varying loads, strains, or stress intensities on structural components. Cyclic tensile loading was repeated on wet ramie plain-woven fabrics. Tensile strength improved the

most, up to 38%, in fabrics treated with 70% of the mean fracture force of untreated ones for 10 cycles. Mechanical characteristics of ramie/PLA laminated composites showed that treated fabric-reinforced composites had 35% higher tensile strength, 32% higher Young's modulus, 20% higher flexural strength, and 17% higher flexural modulus [98]. After cyclic loading, fabrics had the highest tensile failure load and lowest failure deformation [35].

PREPARATION METHODS OF BIO-COMPOSITES WITH RAMIE FIBRES

Composite fabrication involves impregnation, lay-up, consolidation, and solidification in various methods [99]. Biocomposites with cellulosic fibres require homogeneous fibre dispersion into the polymer matrix. It maximizes component contact and improves mechanical properties [60]. The most commonly used techniques to produce ramie fibre-reinforced PLA composites are hot compression moulding, vacuum-assisted resin transfer moulding, and injection moulding. Composite qualities depend on process parameters like time, temperature, and pressure. The processing temperature is set based on ramie fibre and polymer matrix thermal properties. Moreover, chemically treated fibres should be dried to remove moisture before composite fabrication. Siakeng et al. dried modified ramie samples at 70 °C for 3 h before carding [27]. A similar pre-treatment was performed by Xu et al., who cut and dried ten layers of 250 × 250 mm ramie fabric at 70 °C for 3 h in an oven and stacked them unidirectionally [72].

Film stacking

In the film stacking process, dried reinforcing fibres are prepared in advance and transformed into mats, fabrics, or unidirectional tapes. A thermoplastic polymer material is prepared in the form of thin films by heating and compressing. The reinforcing fibres and polymer films are arranged in a stacked configuration, with each layer alternating between the two materials. This results in a configuration where the fibres are positioned between the polymer layers [100]. Film stacking reduces impregnation and infiltration time [101]. Xu et al. compacted the ramie fabric stack with vacuum pressure or hot compaction to increase fibre content [72].

Hot compression moulding

Match mould compression moulding requires male and female moulds (Figure 4). The compression mould heats the resin to cure it. Usually, the mould is heated to 120-170 °C. Moulds can be heated using steam, oil, or an electrical cartridge [99]. Dereje et al. moulded ramie fibre/PLA composites at 175 °C and 6-7 MPa for 4 min. The moulded green composite was cold compressed at 3-4 MPa for 2 min at room temperature [27].

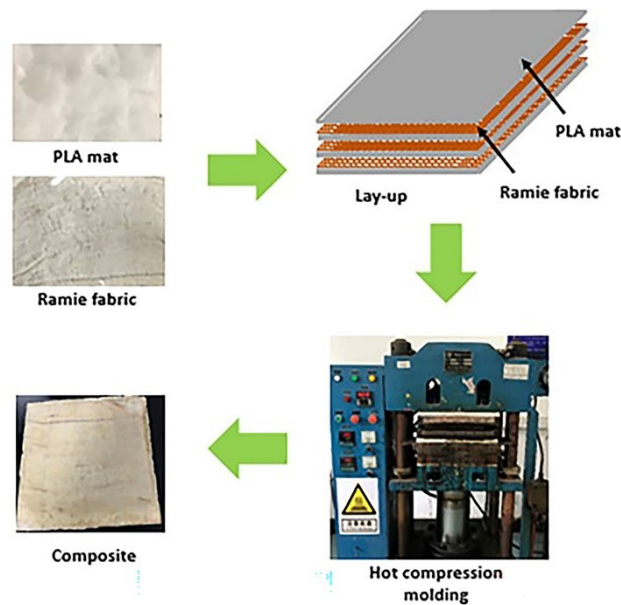


Figure 4. Hot compression moulding process steps

Vacuum-assisted resin transfer moulding (VARTM)

It is a closed-mold process which can make high-performance, large-scale fibre-reinforced polymer products with inexpensive tooling [102]. Air was aspirated after laminating reinforcement on a mould and sealing with plastic film. The reinforcing phase was impregnated with liquid resin and cured [103]. It transfers liquid thermoset resin into a closed mould at low pressure and temperature, producing dimensionally precise composites with great surface quality [104].

Vacuum-assisted resin infusion moulding (VARI)

This composite manufacturing technology produces high-quality, large-scale components. This procedure involves placing dry pre-form fabrics in an open mould with a plastic vacuum bag on top. One-sided moulds have resin sources and vacuum pumps. A vacuum is generated within the mould, and then the liquid resin is poured over the reinforcing fibres. The product is finished by curing and removing the mould. Xu et al. sealed the dry fabric stack with a vacuum bag on one side of the mould. Under vacuum pressure, a vacuum pump injected epoxy resin into the stack and evacuated the gas in a vacuum bag. To fully enter the fabric stack, the resin was injected for 5 min and vacuumed for 20 min. The assembly was oven-cured for 6 h at 70 °C under vacuum [72]. Interest has arisen in the manufacturing of natural fibre composites using the VARI process. As an important driving force for the impregnation of the fibre tows, the capillary effect of natural fibres played an important role in the VARI process [105].

PERFORMANCE OF RAMIE FIBRE AFTER TREATMENT

The objective of fibre pretreatment is to enhance the interfacial adhesion with the polymeric matrix. The composites showed improved physical, chemical, mechanical, thermal, and other properties after treatments. Several analyses can be performed on the fibres as well as on the composite to investigate the effectiveness of the treatments.

Surface Morphology

Scanning electron microscope (SEM) and atomic force microscope (AFM) can examine the fibre/fabric surface and fracture zone of ramie fibre/fabric and its composites. The micrographs show fibre-matrix contact. According to Xinqi et al., silicone oil-modified ramie fibre-reinforced polypropylene composite had superior interfacial bonding when the fracture surface was observed under SEM [65]. In another study, chemical treatment removed lignin and hemicellulose, resulting in increased fibre surface defects and roughness [67]. The ramie-PLA biocomposite fracture surface is shown in Figure 5. Due to the layer produced during treatment, Dereje et al. found that DAP- and alkali-DAP-treated composites have more lustre [27].

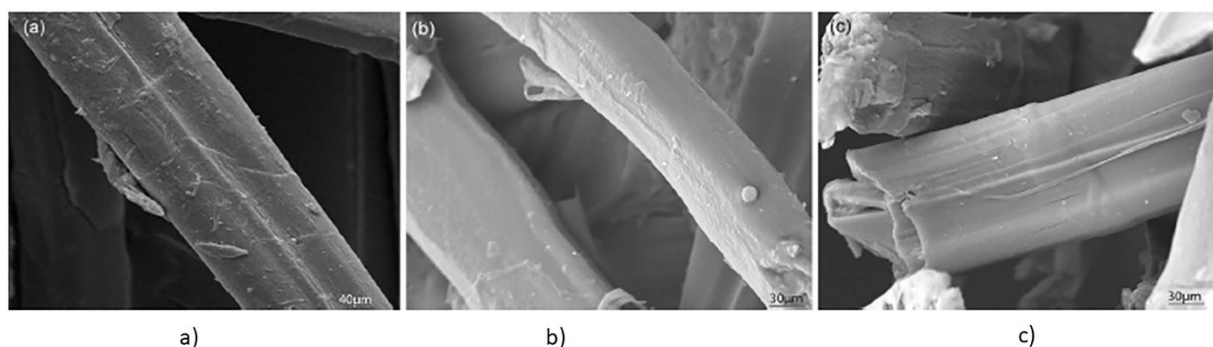


Figure 5. SEM images of ramie fibre: (a) before treatment, (b) coated with DAP, and (c) treated with alkali and DAP [27]

The irradiation treatment also alters the surface morphology of ramie fibres. An investigation was conducted to examine the alterations in surface morphology of biocomposites following irradiation treatment of ramie fibres. Figure 7 demonstrates that a rougher surface was produced after 16 s of plasma treatment on ramie fibre with prior ethanol pretreatment [73]. In similar investigations, Wang et al. reported that scanning electron microscopy shows increased surface roughness [76]. The fibre or fabric surface undergoes an increase in roughness when plasma is used to remove non-cellulose components through the process of kinetic knocking. Figure 6 demonstrates the impact of plasma treatment on the fibre's surface through surface etching. Greater surface roughness enhances the available surface area for polymer matrices to bond with ramie fibres.

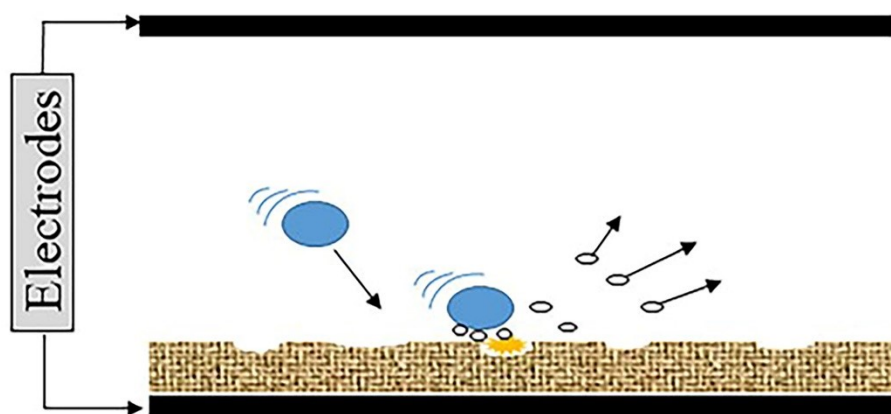


Figure 6. Surface roughening of ramie fibre by physical etching using plasma

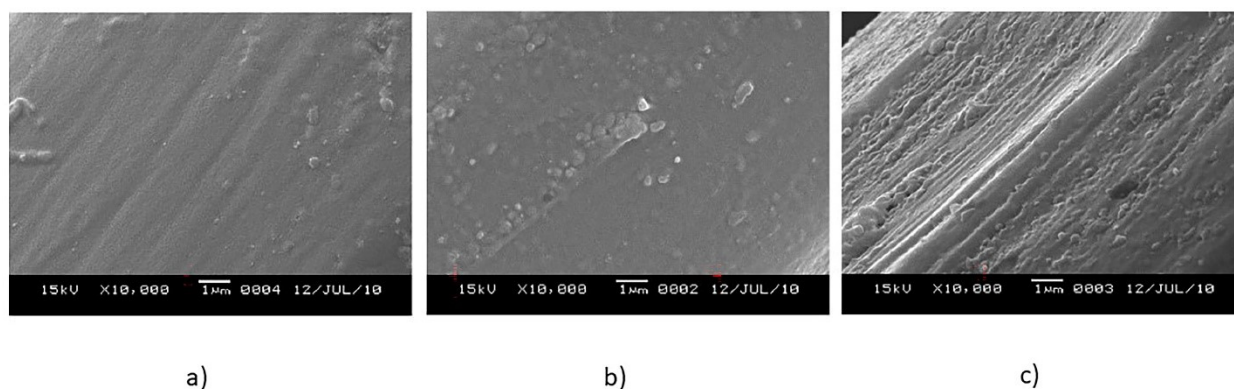


Figure 7. SEM photographs of control and treated ramie fibres: (a) control; (b) plasma treated for 16 s with ethanol pretreatment; (c) plasma treated for 24 s with ethanol pretreatment [73]

Surface Wettability

Surface wettability is a material's ability to reject or attract liquids. Wet surfaces allow liquids to spread, while hydrophobic surfaces bead up and roll off. Wetting or non-wetting is measured by the liquid droplet's contact angle on the surface. Surface energy, the energy needed to break intermolecular bonds, determines a surface's wetting behaviour. A surface with high surface energy can interact harder with liquid molecules, making it more wettable. A surface with low surface energy cannot.

Different researchers investigated the surface wettability of composites reinforced with ramie fibres after fibre surface modification. Hui Li et al. performed alkali treatment (NaOH 10%) as well as combined alkali and silane treatment at room temperature for 24 h. Ramie fibre water contact angle fell from 74.89° (untreated) to 55.25° (alkali-treated) [62]. Dereje et al. found that alkali treatment and DAP reduced ramie fibre hydrophilicity best. The surface energy of untreated ramie fibre dropped from 5.9 ± 1.3 to 2.0 ± 0.8 mJ/m² after treatment. Alkali/silane/DAP-treated ramie fibre/PLA composite has a higher water contact angle ($95.4^\circ \pm 1.3$) than pure ramie fibre/PLA composite ($87.3^\circ \pm 1.9$) [27].

Impurities on the fibre surface were eliminated after alkali treatment, and the diameter of the fibre grew finer (Figure 8). Alkali treatment can break lignin-held fibre bundles into finer fibrils. This greater surface area improves fibre adherence. Equation 1 shows the reaction mechanism during alkali treatment. The contact angles of the ramie fibres rose by 14.26% and 33.12% after the silane only and combined alkali–silane treatments, respectively. The alkali–silane treated fibre has a contact angle of 76.41° [49]. The ramie fibre treated with 0.08% permanganate acetone solution turned out to be rougher so that the specific surface area became larger and the interface contact area between the PLLA matrix and fibre treated with permanganate acetone increased [52]. It can be observed that combined alkali and silane coupling agent treatment results less wettable surface. Figure 9 shows the reaction mechanism during silane treatment.

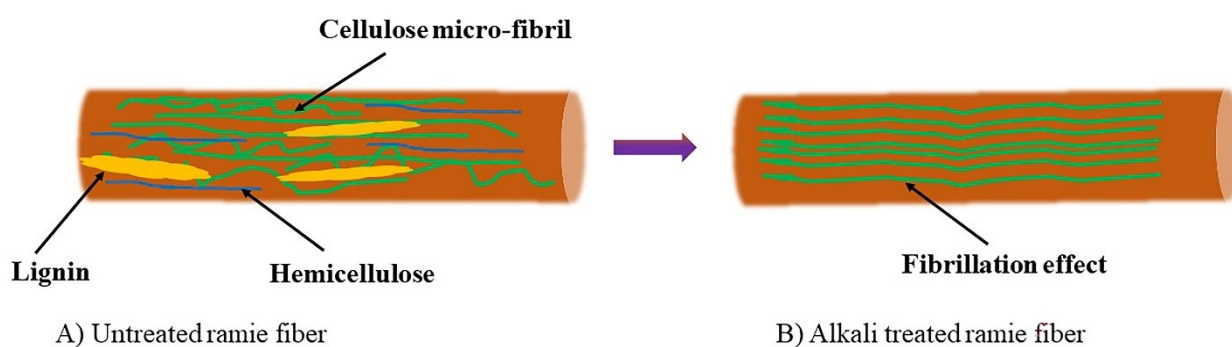
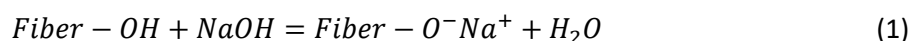


Figure 8. Schematic representation of alkali treatment

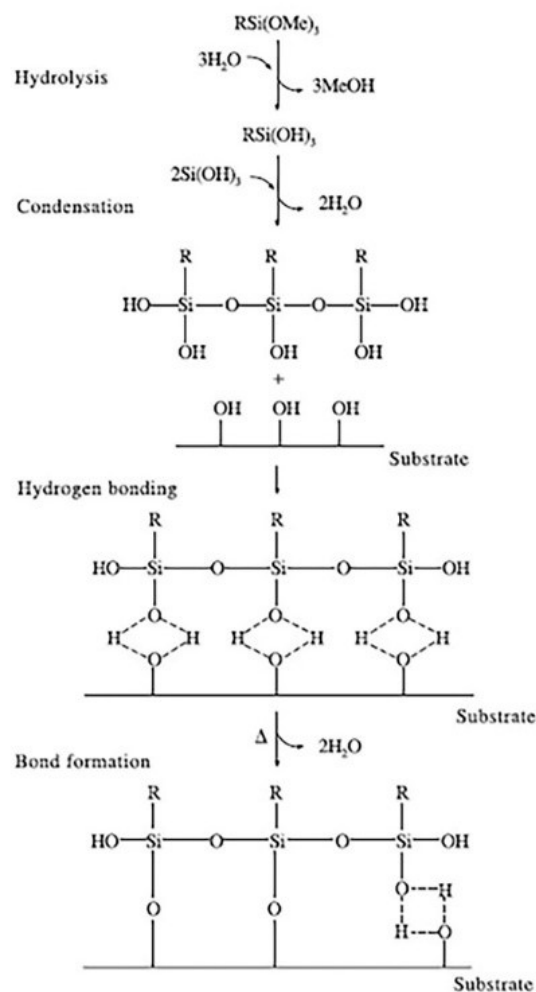


Figure 9. Reaction mechanism between ramie cellulosic fibre and silane [106]

The review also discusses irradiated ramie fibre surface wettability. Zhou et al. found that plasma-only treated ramie fibres had an average advance angle of 48.2° , while ethanol-pretreated fibres had 73.1° . The plasma interaction of cellulose and ethanol makes ramie fibres more hydrophobic [73]. The surface energy of ramie fibres improved by 124.5% while the water contact angle decreased from $83.9 \pm 6.2^\circ$ to $50.5 \pm 4.5^\circ$ after a 3 min 200 W irradiation treatment [77]. The improvement is achieved due to the effective elimination of pectin and impurities; hence, the fine fibre with irregular striations was revealed. The water contact angles of ramie fibres pretreated with isopropanol- and n-butanol followed by plasma treatment had larger contact angles [76]. The plasma-treated ramie fibres' advance contact angles are increased from 66.3° to 106.1° owing to an increase in fibre surface roughness observed under a scanning electron microscope [9]. During low-temperature plasma treatment, the surface energy of ramie fibres increased by 117.0 and 122.9% after 1 min-300 W and 3 min-100 W treatment, respectively [74]. After 1 min plasma treatment fibres show a smaller contact angle, $38^\circ \pm 2.2$ than untreated ramie fibres, whose contact angle was $49^\circ \pm 1.9$ [78]. Prolonged exposure of fibres

to irradiation could affect their tensile strength because it can break down the long polymer chains within the fibre structure. It can be concluded that alcohol pretreatment improves the hydrophobicity of ramie fibres.

Surface Chemical Composition

Surface chemical composition is another property that can be affected by surface modification. According to Choi et al., the FTIR peaks at $1,730\text{ cm}^{-1}$, 750 cm^{-1} , and $1,145\text{--}1,000\text{ cm}^{-1}$ indicate that alkalization reduced hemicellulose and lignin. On the other hand, the hydrophilicity of Ramie fibre was reduced by the application of silane and peroxide treatment, which resulted in the coating of the fibre surface with nonpolar groups [64]. After silane treatment, peaks changed. In silane and CNT-treated fibres, the absorption peaks at 835 cm^{-1} and 852 cm^{-1} indicate cellulose–Si bonding. The small peaks at 934 and 935 cm^{-1} suggest Si–O–C or Si–O–cellulose bonding. CNT grafting onto the ramie fibre surface was also suggested by the low intensity of peaks for –OH stretching and bending vibrations. In all alkali-treated fibres, the peak at 1730 cm^{-1} vanished, indicating the elimination of amorphous materials and perhaps hemicellulose [70]. As the increment of hydrophilicity, the FTIR results were consistent with the wetting measurements [27]. The bond features and peak positions are presented in Table 4. It can be summarized that all chemical treatments change the surface chemical composition of ramie fibre.

Table 4. Bond features and peak positions of control fibre, alkali-treated, silane-treated fibre, and CNT-treated fibre [64,67,70]

Band assignment	Untreated (cm^{-1})	Alkali treated	Combined alkali and Silane treated (triethoxy vinyl)	Combined alkali, Silane and peroxide treated
-OH stretching, vibration	3400	-	3344	-
C-H and CH_2 stretching, the vibration of cellulose/hemicelluloses	2900	Divided into two peaks	2942	Divided into two peaks
C=O stretching, vibration of the acetyl group of hemicellulose	1,730-1,750	Nil	-	-
C=O bonds of the hemicellulose structure	1620	-	-	Peak reduced
the stretching vibration of cellulose polysaccharide CO and O-H groups	1030-1034	-	-	-
Aromatic C=C stretching, lignin vibration	1655	-	1656	-
C-C stretching	1162-1000	-	-	-
Vibrations of waxes' C-O ester, ether, and phenolic groups on fibre surfaces.	1230	-	-	-
C-H bending, b-glucosidic linkage of C1-O-C ₄	1360 and 1320	-	-	-

Band assignment	Untreated (cm ⁻¹)	Alkali treated	Combined alkali and Silane treated (triethoxy vinyl)	Combined alkali, Silane and peroxide treated
Out-of-plane bending vibration of intramolecular H-bonded OH group	668	-	-	-
Lignin components due to C-O, C-C stretching	1,426	Nil	-	-
C-OH bending in hemicelluloses	1,730-1,750	Nil	-	-
Si-O-Si symmetric bonds	-	-	700	-
-Si-C symmetric stretching bond	-	-	765	-
Si-O-C bond cellulose-O-Si vibration	-	-	950-1,150	-
α -cellulose and β -cellulose	838 & 894	Reduced peak at 838		-
Si-cellulose vibration	-	-	835	-
COC asymmetrical stretching, C-C, C-OH, C-H ring, and side group vibrations	-	-	1033, 1055	-
C-O-C asymmetric stretching vibrations at beta-(1-4)-glycosidic linkages	893 - 898	-	1105, 1159	-
CH ₂ rocking vibration	1372	-	1317	-
C-OH in-plane vibration, CH ₂ deformation vibration	-	-	1430	-

Ramie fibre's chemical composition is also affected by plasma treatment. X-ray photoelectron spectroscopy (XPS), Fourier transform infrared spectroscopy (FTIR), and Electron spin resonance (ESR) spectroscopy measured the surface chemical composition of ramie after combined ethanol pretreatment, and the results show that oxygen to carbon (O/C) ratios decreased significantly after plasma treatment for 8 and 16 s. The chemical interaction between ethanol and plasma caused an ethyl group to graft on the fibre surface during short treatment times [73]. The C-C bond increased significantly in the isopropanol-pretreated group, whereas the increase in C=O bonds was most visible in the n-butanol-pretreated group, according to X-ray photoelectron spectroscopy analysis [76].

Mechanical Properties

Tensile Strength

Composite modification is indicated by fibre surface modification-induced mechanical property changes. Hui et al. found that ramie fibre-reinforced vinyl resin composites' tensile strength increased after alkali and silane treatments of the fibre, except for NaIO₄-treated composites. The tensile strength of silane (KH570) treated ramie fibre-reinforced vinyl resin composite increased by 26.46%

[62]. Dilfi KF. et al. found that treating the fibre-matrix interface with alkali, silane, and CNT greatly enhanced the flexural, tensile, and interlaminar shear strength (ILSS) of modified fibre-epoxy composite plates [67]. The two-dimensional structure of potassium permanganate and silane-treated fabric increases PLLA's tensile and flexural strength by 20%. The highest tensile strength was 73 MPa. At 25 °C, the storage modulus of the PLLA/potassium permanganate and silane-treated fibre was 80% higher than that of the film PLLA/potassium permanganate and silane-treated fabric composites. Similarly, it rose by 52% for powdered PLLA/potassium permanganate and silane-treated fabric [61]. Interfacial adhesion between fibres and matrix increases tensile strength and modulus. Alkali treatment removes hemicellulose, lignin, and other impurities, improving fibre hygroscopicity. The silane treatment increased adherence. Alkali treatment removed impurities and hemicelluloses; hence, ramie fibre composites gained improved tensile strength. Combined alkali and silane treatments increased composite mechanical characteristics. Both alkali and silane treatments showed variability in improvement, suggesting the need to adjust chemical treatment conditions and process parameters during composite manufacture.

Ramie fibre's mechanical properties improved significantly after irradiation treatment. The tensile strength of ramie fibre after 3 min of 200 W treatment was 253.8 MPa, which was 30.5% greater than the tensile strength of untreated fibre-reinforced composite [77]. However, Zhou et al. found no statistically significant difference in ramie fibre tensile strength following plasma treatment. This indicates that plasma treatments had no discernible influence on the tensile strength of ramie fibre [73]. Plasma-etched fibres sometimes have lower tensile strength due to cracks and pits on the fibre surface generated by plasma contact. Interfacial characteristics are affected by fibre plasma treatments. After 8, 16, and 24 s of plasma treatment, interfacial shear strength (IFSS) increased by 20%, 19%, and 46%. The hydrophobic surface created by ethanol and cellulose in an atmospheric pressure plasma jet (APPJ) environment may have improved adhesion with polypropylene matrix [73]. The interfacial adhesion of the PP matrix and the ramie increases by 26% in the freshly made ethanol-pretreated + APPJ-treated group, according to an interfacial shear strength test [75]. Fragmentation tests revealed a large interface shear stress of 17.2 MPa, a 54.0% increase above untreated fibre-reinforced composites [77]. Interfacial shear strengths of fibre/polypropylene samples rise by 47 and 34%, respectively, in a microbond pull-out test [76]. To summarize, optimal irradiation removes impurities from the surface of ramie fibres, whereas excessive irradiation might degrade cellulose and weaken the fibres.

Flexural Strength

Matrix cracking, delamination, fibre breaking, and penetration of the impacted surface are the four major failure modes when the fibre-reinforced composites are exposed to impact damage. Alkali-

treated fibre composites resulted in an improvement of 4.89% in flexural strength [67]. According to Hui et al., the flexural strength of silane(KH550) treated ramie fibre-reinforced vinyl resin composites increased by 57.97% [62]. In a similar investigation, silane-treated ramie fibre-reinforced epoxy composites exhibited an increase of 19.34% in flexural strength [67] which further confirmed that in addition to alkali treatment silane coupling agent modification has a good effect. In similar investigations, CNT grafted fibre composites with 0.7 weight % CNT showed a 38.17% increase in flexural strength [67]. Combined alkali, silane treatment, and CNT grafting improved flexural strength [62]. Improved interfacial bonding increases flexural strength. Chemical treatments on the fibre surface improved matrix-fibre adhesion. This increased load transition from matrix to fibre, increasing flexural strength.

Impact Strength

The mechanical properties of ramie fibre-reinforced composite materials modified with 10% silane (KH570) were the best, with tensile strength increasing by 26.45%, flexural strength increasing by 49.29%, and impact strength increasing by 38.28%. The 10% silane (KH550) modification effect came right after the silane modification (KH570). Choi et al. confirmed that peroxide treatment after alkali treatment further enhanced (65%) the impact strength of the composite [64]. The composite with 8% alkali treatment recorded higher tensile strength and flexural strength, and slightly decreased impact strength was demonstrated because of the fibre surface treatment [50]. The potential cause for a reduction in impact strength is the diminished toughness of ramie due to the damaged integrity of the fibre bundle structure. This implies that the negative influence of impaired fibres on the impact performance of composites outweighed the positive enhancement of interfacial adhesion. Additionally, the increased fibre volume ratio (48%) affects the efficiency of load transfer within the matrix, resulting in a decrease in the impact resistance of the ramie-reinforced thermoset PLA composites. In a different investigation, Li et al. found that silicone oil emulsion treatment of ramie improved the impact strength by 17.0% and elongation at the break by 19.6% of ramie fibre-reinforced polypropylene composite [65]. The impact strength of ramie has different degrees of improvement after alkali and silane modifications, and fibre modified by NaOH increased by 73.02% [62].

Flame Retardant Properties

Flame-retardants reduce the risk of fire caused by naturally combustible materials that are essential to maintaining a high level of living. Although bulk flame-retardants have been effective for years, surface treatments that localize the chemistry at the material's surface are gaining popularity [107]. The flame retardant is prone to charring or melting when exposed to fire. Utilizing additives that break down into non-combustible gases decreases the amount of fuel and oxygen in the area surrounding

the flame, as illustrated in Figure 10. The category of flame retardants comprises organic halides and some nitrogen compounds [108]. Yan et al. modified ramie fibre using APP and found that ramie/benzoxazine resin laminate without flame° retardant modification has a 23.2% limiting oxygen index (LOI) and lacks UL94 certification. Although ammonium polyphosphate (APP) lowers the flammability of ramie/benzoxazine resin laminate, it still fails the UL94 test. Tensile and flexural strengths without flame retarding adjustment are 151 MPa and 197 MPa, respectively. However, adding 1% weight APP significantly reduces both tensile and flexural strengths [63]. In a similar study, To protect the ramie/benzoxazine resin laminate from mechanical harm, the APP was covered with melamine resin (Mel-APP). The dynamic mechanical analysis (DMA) revealed a higher glass transition temperature and lower damping peaks, implying a better fibre–matrix interaction as a result of the CNT grafting [67]. The coefficient of thermal expansion was measured by heating the composites from 30 °C to 100 °C at a rate of 5 °C/min with an applied load of 0.05 N. With a temperature increase from 30 to 100 °C, the PLA matrix showed a significant dimensional change [64]. The thermal expansion of the composites reinforced with ramie fibres exhibited a significant reduction. This reduction can be attributed to the fibres' ability to restrict the thermal expansion within the composites. Composites exhibited significant dimensional changes at 70 °C, whereas the tensile strength of the composite decreased. This is attributed to the enlargement of surface area, which resulted in enhanced interlocking between the fibre and PLA matrix. The interlocking between the fibre and PLA matrix hindered the expansion of the PLA matrix.

Li et al. modified ramie using silicone oil. According to thermogravimetric (TGA) analysis, fibre treatment can raise the degradation temperature of ramie fibre-reinforced polypropylene composites. [65]. DAP-modified ramie fibre/PLA composites had a lower thermal decomposition temperature, distinctive breakdown behaviour, and higher residual char formation at the decomposition temperature [27]. According to Chen et al., the composite produced with alkali-treated ramie fabrics had slightly better water resistance and thermal stability [50]. The flame retardant characteristics are enhanced by the treatments; however, they have a negative impact on the mechanical properties.

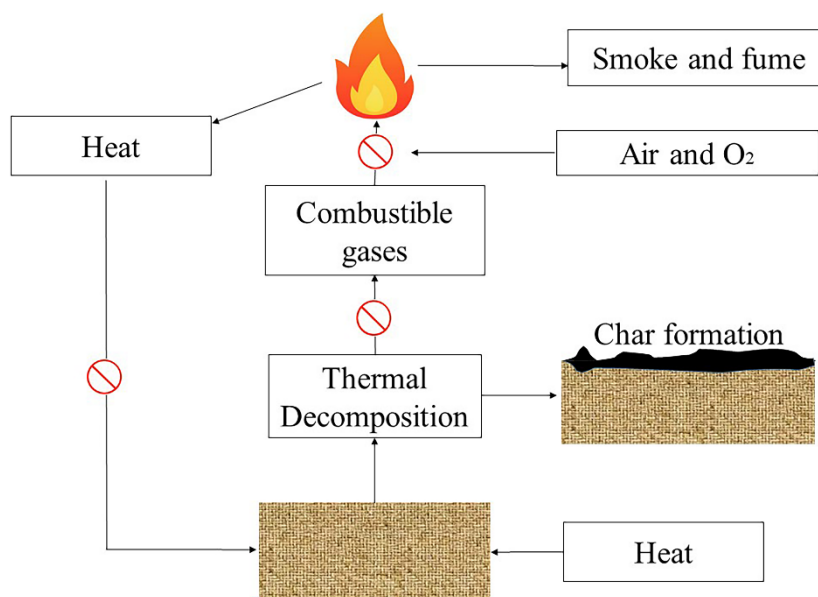


Figure 10. Flame retarding mechanism of cellulosic fibres

FUTURE OUTLOOKS FOR RAMIE FIBRE REINFORCED PLA ACID COMPOSITES

Ramie-polylactic acid biocomposites, made from ramie plant fibre and PLA, are a new sustainable material. Compared to standard synthetic composites, they may have a lower environmental impact, better biodegradability, and more sustainability. Ramie-PLA biocomposites have bright futures. Among them:

Demand: As customers learn about the environmental impact of typical plastics and the benefits of sustainable materials, ramie-PLA biocomposites may become more popular.

Improved performance: Researchers are working to improve biocomposites' mechanical properties, durability, and moisture and UV resistance. The improvement strategy uses chemical, biological, and physical methods. Dipodal and aqueous silane coupling agents can improve natural fibre-polymeric resin adhesion. Biocomposites that outperform synthetic composites may arise from these efforts.

New applications: Automotive, packaging, and building materials use Ramie-PLA biocomposites. Technological advances and a better understanding of biocomposites may lead to new uses in aerospace, sports equipment, and electronics.

Cut costs: Biocomposites are more expensive than synthetic composites, which is a drawback. However, manufacturing optimization and economies of scale should lower the cost of ramie-PLA biocomposites, making them more competitive.

In general, Ramie-PLA biocomposites are promising and rapidly evolving materials with a bright future. These sustainable and eco-friendly materials will likely replace synthetic composites in more applications with continuous research and development.

CONCLUSION

Consideration of the interface between polymer matrices and reinforcing fibres is crucial in producing high-quality biocomposites from natural fibres like ramie. Physicochemical treatments of ramie fibres are essential to enhance their adherence to the matrix. These treatments are diverse and effective in achieving their goals, but their large-scale use requires careful consideration of environmental impact. Alkaline treatments get rid of unnecessary parts of ramie fibres, which expose functional groups in cellulose and make it stick to the matrix better. During alkali treatment, slack and tension both enhance strength uniformity, with tension showing a greater strength gain in mercerized fibre. Future research should focus on treating fibres with tension during chemical processes.

A common way to treat ramie is with alkali (NaOH), and treatments that combine alkali, silane coupling agents, peroxides, and CNT grafting give better mechanical strength. Conventional silane treatments form three silanol linkages, enhancing interfacial adhesion. Adding dipodal silanes creates six silanol linkages that make the material more stable in water, better at bonding to substrates, and stronger mechanically. Ethanol and methanol are used as solvents, with water-based silanes not observed in previous research. Water, having more hydrogen bonding sites than ethanol and methanol, is a more effective polar solvent. The tensile strength of alkali-treated ramie fibre composites increases by eliminating impurities and hemicelluloses. Combined treatments result in better mechanical properties, with variations indicating the need to optimize chemical treatment conditions and process parameters during fabrication.

Gamma irradiation emerges as a promising option for composites with high ramie proportions. Ethanol pretreatment before plasma treatment enhances surface roughness. In the coming years, composites reinforced with hybrid fibres will find wider applications, with sustainable green chemistry changes such as gamma irradiation enhancing the qualities of materials reinforced with plant fibres.

Author Contributions

Conceptualization –Wagaye BT and Guo J; methodology – Nguyen LT and Adamu BF; formal analysis – Gudayu AD; investigation – Wagaye BT; resources – Gudayu AD; writing-original draft preparation – Wagaye BT; writing-review and editing – Guo J and Gudayu AD; visualization – Nguyen LT; supervision – Guo J. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this article.

Funding

The work was financially supported by the China Agriculture Research System for Bast and Leaf Fibre Crops (Grant No. CARS-16).

Acknowledgements

The first author expresses gratitude to the Chinese government scholarship (CSC) for providing financial support.

REFERENCES

- [1] González-López ME, Robledo-Ortíz JR, Manríquez-González R, Silva-Guzmán JA, Pérez-Fonseca AA. Polylactic acid functionalization with maleic anhydride and its use as coupling agent in natural fiber biocomposites: a review. *Composite Interfaces*. 2018; 25(5-7):515-538. <https://doi.org/10.1080/09276440.2018.1439622>
- [2] Siakeng R, Jawaid M, Ariffin H, Sapuan SM, Asim M, Saba N. Natural fiber reinforced polylactic acid composites: A review. *Polymer Composites*. 2018; 40(2):446-463. <https://doi.org/10.1002/pc.24747>
- [3] Filpponen I, Kontturi E, Nummelin S, Rosilo H, Kolehmainen E, Ikkala O, Laine J. Generic method for modular surface modification of cellulosic materials in aqueous medium by sequential "click" reaction and adsorption. *Biomacromolecules*. 2012; 13(3):736-42. <https://doi.org/10.1021/bm201661k>
- [4] Yao C, Tianshu S, Chaunjie F, Xiaodong Z. The effect of surface modification on the properties of sisal fiber and improvement of interfacial adhesion in sisal/starch composites induced by starch nanocrystals. *Composite Interfaces*. 2018; 25(11):981-994. <https://doi.org/10.1080/09276440.2018.1450586>
- [5] Karabulut N, Aktaş M, Balcıoğlu HE. Surface Modification Effects on the Mechanical Properties of Woven Jute Fabric Reinforced Laminated Composites. *Journal of Natural Fibers*. 2018; 16(5):629-643. <https://doi.org/10.1080/15440478.2018.1431995>
- [6] Zhong J, Li H, Yu J, Tan T. Effects of Natural Fiber Surface Modification on Mechanical Properties of Poly(lactic acid) (PLA)/Sweet Sorghum Fiber Composites. *Polymer-Plastics Technology and Engineering*. 2011; 50(15):1583-1589. <https://doi.org/10.1080/03602559.2011.557817>
- [7] Jawaid M, Othman A, Saba N, Shekeil YA, Paridah MT, Abdul Khalil HPS. Effect of Chemical Modifications of Fibers on Tensile Properties of Epoxy Hybrid Composites. *International Journal of Polymer Analysis and Characterization*. 2014; 19(5):391-403. <https://doi.org/10.1080/1023666x.2014.904081>

- [8] Oladele IO, Oladejo MO, Adediran AA, Makinde-Isola BA, Owa AF, Akinlabi ET. Influence of designated properties on the characteristics of dombeya buettneri fiber/graphite hybrid reinforced polypropylene composites. *Sci Rep.* 2020; 10(1):11105. <https://doi.org/10.1038/s41598-020-68033-y>
- [9] Zhang Q, Jiang Y, Yao L, Jiang Q, Qiu Y. Hydrophobic surface modification of ramie fibers by plasma-induced addition polymerization of propylene. *Journal of Adhesion Science and Technology.* 2015; 29(8):691-704. <https://doi.org/10.1080/01694243.2014.997380>
- [10] Atiqah A, Jawaid M, Ishak MR, Sapuan SM. Effect of Alkali and Silane Treatments on Mechanical and Interfacial Bonding Strength of Sugar Palm Fibers with Thermoplastic Polyurethane. *Journal of Natural Fibers.* 2017; 15(2):251-261. <https://doi.org/10.1080/15440478.2017.1325427>
- [11] Goldmann AS, Tischer T, Barner L, Bruns M, Barner-Kowollik C. Mild and modular surface modification of cellulose via hetero Diels-Alder (HDA) cycloaddition. *Biomacromolecules.* 2011; 12(4):1137-45. <https://doi.org/10.1021/bm101461h>
- [12] Fatona A, Berry RM, Brook MA, Moran-Mirabal JM. Versatile Surface Modification of Cellulose Fibers and Cellulose Nanocrystals through Modular Triaziny Chemistry. *Chemistry of Materials.* 2018; 30(7):2424-2435. <https://doi.org/10.1021/acs.chemmater.8b00511>
- [13] Rajeshkumar G. Characterization of Surface Modified Phoenix sp. Fibers for Composite Reinforcement. *Journal of Natural Fibers.* 2020; 18(12):2033-2044. <https://doi.org/10.1080/15440478.2019.1711284>
- [14] Sun Z. Hyperbranched Polymers in Modifying Natural Plant Fibers and Their Applications in Polymer Matrix Composites-A Review. *J Agric Food Chem.* 2019; 67(32):8715-8724. <https://doi.org/10.1021/acs.jafc.9b03436>
- [15] Maichin P, Suwan T, Jitsangiam P, Chindaprasirt P, Fan M. Effect of self-treatment process on properties of natural fiber-reinforced geopolymer composites. *Materials and Manufacturing Processes.* 2020; 35(10):1120-1128. <https://doi.org/10.1080/10426914.2020.1767294>
- [16] Filgueira D, Holmen S, Melbø JK, Moldes D, Echtermeyer AT, Chinga-Carrasco G. Enzymatic-Assisted Modification of Thermomechanical Pulp Fibers To Improve the Interfacial Adhesion with Poly(lactic acid) for 3D Printing. *ACS Sustainable Chemistry & Engineering.* 2017; 5(10):9338-9346. <https://doi.org/10.1021/acssuschemeng.7b02351>
- [17] Zhang Q, Fang X, Sun X, Sun B, Qiu Y. Comparison of the Mechanical Properties between 2D and 3D Orthogonal Woven Ramie Fiber Reinforced Polypropylene Composites. *Polymers and Polymer Composites.* 2014; 22(2):187-192. <https://doi.org/10.1177/096739111402200216>
- [18] Mohanty AK, Vivekanandhan S, Pin JM, Misra M. Composites from renewable and sustainable

- resources: Challenges and innovations. *Science*. 2018; 362(6414):536-542.
<https://doi.org/10.1126/science.aat9072>
- [19] Bouhank S, Nekkaa S, Haddaoui N. Water absorption, biodegradation, thermal and morphological properties of Spartium junceum fiber-reinforced polyvinylchloride composites: effects of fibers content and surface modification. *Journal of Adhesion Science and Technology*. 2016; 30(13):1462-1478. <https://doi.org/10.1080/01694243.2016.1150118>
- [20] Shi S, Yang C, Nie M. Enhanced Interfacial Strength of Natural Fiber/Polypropylene Composite with Mechanical-Interlocking Interface. *ACS Sustainable Chemistry & Engineering*. 2017; 5(11):10413-10420. <https://doi.org/10.1021/acssuschemeng.7b02448>
- [21] Cisneros-López EO, González-López ME, Pérez-Fonseca AA, González-Núñez R, Rodrigue D, Robledo-Ortíz JR. Effect of fiber content and surface treatment on the mechanical properties of natural fiber composites produced by rotomolding. *Composite Interfaces*. 2016; 24(1):35-53. <https://doi.org/10.1080/09276440.2016.1184556>
- [22] Li Y, Jiang L, Xiong C, Peng W. Effect of Different Surface Treatment for Bamboo Fiber on the Crystallization Behavior and Mechanical Property of Bamboo Fiber/Nanohydroxyapatite/Poly(lactic-co-glycolic) Composite. *Industrial & Engineering Chemistry Research*. 2015; 54(48):12017-12024. <https://doi.org/10.1021/acs.iecr.5b02724>
- [23] Liang Y-Y, Xu J-Z, Li Y, Zhong G-J, Wang R, Li Z-M. Promoting Interfacial Transcrystallization in Polylactide/Ramie Fiber Composites by Utilizing Stereocomplex Crystals. *ACS Sustainable Chemistry & Engineering*. 2017; 5(8):7128-7136.
<https://doi.org/10.1021/acssuschemeng.7b01317>
- [24] Jena PK, Mohanty JR, Nayak S. Effect of Surface Modification of Vetiver Fibers on Their Physical and Thermal Properties. *Journal of Natural Fibers*. 2020; 19(1):25-36.
<https://doi.org/10.1080/15440478.2020.1726249>
- [25] Faruk O, Bledzki AK, Fink H-P, Sain M. Progress Report on Natural Fiber Reinforced Composites. *Macromolecular Materials and Engineering*. 2014; 299(1):9-26.
<https://doi.org/10.1002/mame.201300008>
- [26] Liston EM, Martinu L, Wertheimer MR. Plasma surface modification of polymers for improved adhesion: a critical review. *Journal of Adhesion Science and Technology*. 1993; 7(10):1091-1127. <https://doi.org/10.1163/156856193x00600>
- [27] Debeli DK, Guo J, Li Z, Zhu J, Li N. Treatment of ramie fiber with different techniques: the influence of diammonium phosphate on interfacial adhesion properties of ramie fiber-reinforced polylactic acid composite. *Iranian Polymer Journal*. 2017; 26(5):341-354.
<https://doi.org/10.1007/s13726-017-0524-2>

- [28] Vega B, Wondraczek H, Zarth CS, Heikkilä E, Fardim P, Heinze T. Charge-directed fiber surface modification by molecular assemblies of functional polysaccharides. *Langmuir*. 2013; 29(44):13388-95. <https://doi.org/10.1021/la402116j>
- [29] Arbelaiz A, Txueka U, Mezo I, Orue A. Biocomposites Based on Poly(Lactic Acid) Matrix and Reinforced with Lignocellulosic Fibers: The Effect of Fiber Type and Matrix Modification. *Journal of Natural Fibers*. 2020; 19(1):1-14. <https://doi.org/10.1080/15440478.2020.1726247>
- [30] Magniez K, Voda AS, Kafi AA, Fichini A, Guo Q, Fox BL. Overcoming interfacial affinity issues in natural fiber reinforced polylactide biocomposites by surface adsorption of amphiphilic block copolymers. *ACS Appl Mater Interfaces*. 2013; 5(2):276-83. <https://doi.org/10.1021/am302013p>
- [31] Xu H, Xie L, Jiang X, Hakkarainen M, Chen JB, Zhong GJ, Li ZM. Structural basis for unique hierarchical cylindrites induced by ultrahigh shear gradient in single natural fiber reinforced poly(lactic acid) green composites. *Biomacromolecules*. 2014; 15(5):1676-86. <https://doi.org/10.1021/bm500100z>
- [32] Sayeed MMA, Paharia A. Optimisation of the surface treatment of jute fibres for natural fibre reinforced polymer composites using Weibull analysis. *The Journal of The Textile Institute*. 2019; 110(11):1588-1595. <https://doi.org/10.1080/00405000.2019.1610998>
- [33] Ovlaque P, Foruzanmehr M, Elkoun S, Robert M. Milkweed floss fiber/PLA composites: effect of alkaline and epoxy-silanol surface modifications on their mechanical properties. *Composite Interfaces*. 2019; 27(5):495-513. <https://doi.org/10.1080/09276440.2019.1655316>
- [34] Benyahia A, Merrouche A. Effect of Chemical Surface Modifications on the Properties of Alfa Fiber-Polyester Composites. *Polymer-Plastics Technology and Engineering*. 2014; 53(4):403-410. <https://doi.org/10.1080/03602559.2013.844828>
- [35] Yang J, Zhu L, Yang Z, Yao L, Qiu Y. Improving mechanical properties of ramie/poly (lactic acid) composites by synergistic effect of fabric cyclic loading and alkali treatment. *Journal of Industrial Textiles*. 2016; 47(3):390-407. <https://doi.org/10.1177/1528083716648763>
- [36] Hatch KL. *Textile Science*. Saint Paul, Minnesota: West Publishing Company College & School Division; 1993. 500 p.
- [37] Ramamoorthy SK, Skrifvars M, Persson A. A Review of Natural Fibers Used in Biocomposites: Plant, Animal and Regenerated Cellulose Fibers. *Polymer Reviews*. 2015; 55(1):107-162. <https://doi.org/10.1080/15583724.2014.971124>
- [38] Du Y, Yan N, Kortshot MT. The use of ramie fibers as reinforcements in composites. In: Faruk O, Sain M, editors. *Biofiber Reinforcements in Composite Materials*. Cambridge, UK: Woodhead Publishing; 2015. p. 104-137. <https://doi.org/10.1533/9781782421276.1.104>

- [39] Rehman M, Gang D, Liu Q, Chen Y, Wang B, Peng D, Liu L. Ramie, a multipurpose crop: potential applications, constraints and improvement strategies. *Industrial Crops and Products*. 2019; 137:300-307. <https://doi.org/10.1016/j.indcrop.2019.05.029>
- [40] Nayak SY, Sultan MTH, Shenoy SB, Kini CR, Samant R, Shah AUM, Amuthakkannan P. Potential of Natural Fibers in Composites for Ballistic Applications – A Review. *Journal of Natural Fibers*. 2020; 19(5):1648-1658. <https://doi.org/10.1080/15440478.2020.1787919>
- [41] Mohit H, Arul Mozhi Selvan V. A comprehensive review on surface modification, structure interface and bonding mechanism of plant cellulose fiber reinforced polymer based composites. *Composite Interfaces*. 2018; 25(5-7):629-667. <https://doi.org/10.1080/09276440.2018.1444832>
- [42] Woods HJ. The Structure of Textile Fibres. Iii.—Fibre Properties and Molecular Arrangement. *Journal of the Textile Institute Proceedings*. 1949; 40(4):P363-P369. <https://doi.org/10.1080/19447014908664662>
- [43] Yu T, Ren J, Li S, Yuan H, Li Y. Effect of fiber surface-treatments on the properties of poly(lactic acid)/ramie composites. *Composites Part A: Applied Science and Manufacturing*. 2010; 41(4):499-505. <https://doi.org/10.1016/j.compositesa.2009.12.006>
- [44] Bello CBdC, Cecchi A. Experiments on natural fibers: durability and mechanical properties. *Advances in Materials and Processing Technologies*. 2017; 3(4):632-639. <https://doi.org/10.1080/2374068X.2017.1364880>
- [45] Fuqua MA, Huo S, Ulven CA. Natural Fiber Reinforced Composites. *Polymer Reviews*. 2012; 52(3):259-320. <https://doi.org/10.1080/15583724.2012.705409>
- [46] Nodder CR, Kinkad RW. The Behaviour of Bast Fibres under the Influence of Caustic Alkalies. *Journal of the Textile Institute Transactions*. 1923; 14(6):T133-T156. <https://doi.org/10.1080/19447022308661243>
- [47] Misra T, Shukla J, Khan MN, Bisoyi DK, Patel T. Fine Structure Analysis of Ramie Fibres by Small Angle X-Ray Scattering Technique. *Polymer Journal*. 1997; 29(12):1007-1011. <https://doi.org/10.1295/polymj.29.1007>
- [48] Marsyahyo E, Jamasri, Heru Santoso Budi R, Soekrisno. Preliminary Investigation on Bulletproof Panels Made from Ramie Fiber Reinforced Composites for NIJ Level II, IIA, and IV. *Journal of Industrial Textiles*. 2009; 39(1):13-26. <https://doi.org/10.1177/1528083708098913>
- [49] Wang C, Ren Z, Li S, Yi X. Effect of Ramie Fabric Chemical Treatments on the Physical Properties of Thermoset Polylactic Acid (PLA) Composites. *Aerospace*. 2018; 5(3). <https://doi.org/10.3390/aerospace5030093>

- [50] Chen X, Zhang N, Gu S, Li J, Ren J. Preparation and properties of ramie fabric- reinforced thermoset poly lactic acid composites. *Journal of Reinforced Plastics and Composites*. 2014; 33(10):953-963. <https://doi.org/10.1177/0731684413520264>
- [51] Buschle-Diller G. Substrates and their structure. In: Cavaco-Paulo A, Gübitz GM, editors. *Textile processing with enzymes*. Boca Raton, USA: Woodhead Publishing Limited; 2003. p. 42-86. <https://doi.org/10.1533/9781855738669.42>
- [52] Chen D, Li J, Ren J. Influence of fiber surface-treatment on interfacial property of poly(l-lactic acid)/ramie fabric biocomposites under UV-irradiation hydrothermal aging. *Materials Chemistry and Physics*. 2011; 126(3):524-531. <https://doi.org/10.1016/j.matchemphys.2011.01.035>
- [53] Lalit R, Mayank P, Ankur K. Natural Fibers and Biopolymers Characterization: A Future Potential Composite Material. *Strojnícky casopis – Journal of Mechanical Engineering*. 2018; 68(1):33-50. <https://doi.org/10.2478/scjme-2018-0004>
- [54] Petroudy SRD. Physical and mechanical properties of natural fibers. In: Fan M, Fu F, editors. *Advanced High Strength Natural Fibre Composites in Construction*. Cambridge, UK: Woodhead Publishing; 2017. p. 59-83. <https://doi.org/10.1016/B978-0-08-100411-1.00003-0>
- [55] Kumar R, Ul Haq MI, Raina A, Anand A. Industrial applications of natural fibre-reinforced polymer composites – challenges and opportunities. *International Journal of Sustainable Engineering*. 2018; 12(3):212-220. <https://doi.org/10.1080/19397038.2018.1538267>
- [56] Sreenivasan S, Sulaiman S, Baharudin BHT, Ariffin MKA, Abdan K. Recent developments of kenaf fibre reinforced thermoset composites: review. *Materials Research Innovations*. 2013; 17(sup2):s2-s11. <https://doi.org/10.1179/1432891713z.000000000312>
- [57] Akil HM, Omar MF, Mazuki AAM, Safiee S, Ishak ZAM, Abu Bakar A. Kenaf fiber reinforced composites: A review. *Materials & Design*. 2011; 32(8-9):4107-4121. <https://doi.org/10.1016/j.matdes.2011.04.008>
- [58] Ghoushji MJ, Eshkoo RA, Zulkifli R, Sulong AB, Abdullah S, Azhari CH. Energy Absorption Capability of Axially Compressed Woven Natural Ramie/Green Epoxy Square Composite Tubes. *Journal of Reinforced Plastics and Composites*. 2017; 36(14):1028-1037. <https://doi.org/10.1177/0731684417700482>
- [59] Puglia D, Biagiotti J, Kenny JM. A Review on Natural Fibre-Based Composites—Part II. *Journal of Natural Fibers*. 2005; 1(3):23-65. https://doi.org/10.1300/J395v01n03_03
- [60] Burrola-Núñez H, Herrera-Franco PJ, Rodríguez-Félix DE, Soto-Valdez H, Madera-Santana TJ. Surface modification and performance of jute fibers as reinforcement on polymer matrix: an overview. *Journal of Natural Fibers*. 2018; 16(7):944-960. <https://doi.org/10.1080/15440478.2018.1441093>

- [61] Chen D, Li J, Ren J. Biocomposites based on ramie fibers and poly(L-lactic acid) (PLLA): morphology and properties. *Polymers for Advanced Technologies*. 2010; 23(2):198-207. <https://doi.org/10.1002/pat.1852>
- [62] Li H, Zhang YF, Liu YQ, Zhao GZ, Du RK. Study on Preparation and Properties of Ramie Fiber Reinforced Composites. *Advanced Materials Research*. 2013; 712-715(2):203-207. <https://doi.org/10.4028/www.scientific.net/AMR.712-715.203>
- [63] Yan H, Wang H, Fang Z. Flame-Retarding Modification for Ramie/Benzoxazine Laminates and the Mechanism Study. *Industrial & Engineering Chemistry Research*. 2014; 53(51):19961-19969. <https://doi.org/10.1021/ie504070t>
- [64] Choi HY, Lee JS. Effects of surface treatment of ramie fibers in a ramie/poly(lactic acid) composite. *Fibers and Polymers*. 2012; 13(2):217-223. <https://doi.org/10.1007/s12221-012-0217-6>
- [65] Li X, He L, Zhou H, Li W, Zha W. Influence of silicone oil modification on properties of ramie fiber reinforced polypropylene composites. *Carbohydrate Polymers*. 2012; 87(3):2000-2004. <https://doi.org/10.1016/j.carbpol.2011.10.023>
- [66] Lee TS, Choi HY, Choi HN, Lee K-Y, Kim S-H, Lee SG, Yong DK. Effect of surface treatment of ramie fiber on the interfacial adhesion of ramie/acetylated epoxidized soybean oil (AESO) green composite. *Journal of Adhesion Science and Technology*. 2013; 27(12):1335-1347. <https://doi.org/10.1080/01694243.2012.697326>
- [67] Dilfi K. F A, Che Z, Xian G. Grafting ramie fiber with carbon nanotube and its effect on the mechanical and interfacial properties of ramie/epoxy composites. *Journal of Natural Fibers*. 2018; 16(3):388-403. <https://doi.org/10.1080/15440478.2017.1423259>
- [68] Yu T, Jiang N, Li Y. Study on short ramie fiber/poly(lactic acid) composites compatibilized by maleic anhydride. *Composites Part A: Applied Science and Manufacturing*. 2014; 64:139-146. <https://doi.org/10.1016/j.compositesa.2014.05.008>
- [69] Yang Q, Lue A, Zhang L. Reinforcement of ramie fibers on regenerated cellulose films. *Composites Science and Technology*. 2010; 70(16):2319-2324. <https://doi.org/10.1016/j.compscitech.2010.09.012>
- [70] Lavoratti A, Romanzini D, Amico SC, Zattera AJ. Influence of Fibre Treatment on the Characteristics of Buriti and Ramie Polyester Composites. *Polymers and Polymer Composites*. 2017; 25(4):247-256. <https://doi.org/10.1177/096739111702500401>
- [71] Ma Q, Yang Z, Gu Y, Li M, Wang S, Zhang Z. Permeabilities along fiber direction of ramie bundles and through-thickness of ramie fabric stack for liquid composite molding. *Journal of Reinforced Plastics and Composites*. 2016; 36(1):40-52. <https://doi.org/10.1177/0731684416670530>

- [72] Xu C, Gu Y, Yang Z, Li M, Li Y, Zhang Z. Mechanical properties of surface-treated ramie fiber fabric/epoxy resin composite fabricated by vacuum-assisted resin infusion molding with hot compaction. *Journal of Composite Materials*. 2015; 50(9):1189-1198.
<https://doi.org/10.1177/0021998315590259>
- [73] Zhou Z, Liu X, Hu B, Wang J, Xin D, Wang Z, Qiu Y. Hydrophobic surface modification of ramie fibers with ethanol pretreatment and atmospheric pressure plasma treatment. *Surface and Coatings Technology*. 2011; 205(17-18):4205-4210.
<https://doi.org/10.1016/j.surfcoat.2011.03.022>
- [74] Liu X, Cheng L. Influence of low-temperature plasma treatment on properties of ramie fibers. *The Journal of The Textile Institute*. 2017; 109(2):172-177.
<https://doi.org/10.1080/00405000.2017.1335368>
- [75] Li Y, Sun J, Cheng P, Jiang Y, Zhou Z, Zhang Q, Qiu Y. Aging of hydrophobized surfaces of ramie fibers induced by atmospheric pressure plasma treatment with ethanol pretreatment. *Journal of Adhesion Science and Technology*. 2013; 27(22):2387-2397.
<https://doi.org/10.1080/01694243.2013.777321>
- [76] Wang J, Zhou Z, Huang X, Zhang L, Hu B, Moyo S, Sun J, Qiu Y. Effect of alcohol pretreatment in conjunction with atmospheric pressure plasmas on hydrophobizing ramie fiber surfaces. *Journal of Adhesion Science and Technology*. 2013; 27(11):1278-1288.
<https://doi.org/10.1080/01694243.2012.738323>
- [77] Liu X, Cheng L. Influence of plasma treatment on properties of ramie fiber and the reinforced composites. *Journal of Adhesion Science and Technology*. 2016; 31(15):1723-1734.
<https://doi.org/10.1080/01694243.2016.1275095>
- [78] Hamad SF, Stehling N, Hayes SA, Foreman JP, Rodenburg C. Exploiting Plasma Exposed, Natural Surface Nanostructures in Ramie Fibers for Polymer Composite Applications. *Materials (Basel)*. 2019; 12(10). <https://doi.org/10.3390/ma12101631>
- [79] Mohanty AK, Misra M, Drzal LT. Surface modifications of natural fibers and performance of the resulting biocomposites: An overview. *Composite Interfaces*. 2001; 8(5):313-343.
<https://doi.org/10.1163/156855401753255422>
- [80] Karmakar SR. *Chemical Technology in the Pre-treatment of Textiles*. Amsterdam, The Netherlands: ELSEVIER; 1999. 498 p.
- [81] Kozłowski R, Władysław-Przybylak M, Helwig M, Kurzydłowski K. Composites Based on Lignocellulosic Raw Materials. *Molecular Crystals and Liquid Crystals*. 2004; 418(1):131-151.
<https://doi.org/10.1080/15421400490479217>
- [82] Mukherjee RR, Woods HJ. Mercerization of jute. *Nature*. 1950; 165(4203):818-9.
<https://doi.org/10.1038/165818b0>

- [83] Sydow Z, Bieńczyk K. The overview on the use of natural fibers reinforced composites for food packaging. *Journal of Natural Fibers*. 2018; 16(8):1189-1200. <https://doi.org/10.1080/15440478.2018.1455621>
- [84] Mukherjee RR, Woods HJ. 17—An X-Ray Study of the Transition from Water-Cellulose to Cellulose II in Ramie, Jute and Sisal. *Journal of the Textile Institute Transactions*. 1951; 42(7-8):T309-T313. <https://doi.org/10.1080/19447027.1951.10750276>
- [85] Then YY, Ibrahim, N. A., Zainuddin, N., Chieng, B. W., Ariffin, H., and Wan Yunus, W. M. Z. Influence of alkaline-peroxide treatment of fiber on the mechanical properties of oil palm mesocarp fiber/poly(butylene succinate) biocomposite. *BioResources*. 2015; 10(1):1730-1746.
- [86] Lee Y-I, Choa Y-H. Adhesion enhancement of ink-jet printed conductive copper patterns on a flexible substrate. *Journal of Materials Chemistry*. 2012; 22(25):12517-12522. <https://doi.org/10.1039/C2JM31381B>
- [87] Abdelmouleh M, Boufi S, Belgacem M, Duarte A, Salah AB, Gandini A. Modification of cellulosic fibres with functionalised silanes: development of surface properties. *International Journal of Adhesion and Adhesives*. 2004; 24(1):43-54. [https://doi.org/10.1016/S0143-7496\(03\)00099-X](https://doi.org/10.1016/S0143-7496(03)00099-X)
- [88] Ao X, Vázquez-López A, Mocerino D, González C, Wang D-Y. Flame retardancy and fire mechanical properties for natural fiber/polymer composite: A review. *Composites Part B: Engineering*. 2024; 268:111069. <https://doi.org/10.1016/j.compositesb.2023.111069>
- [89] Debeli DK, Zhang Z, Jiao F, Guo J. Diammonium phosphate-modified ramie fiber reinforced polylactic acid composite and its performances on interfacial, thermal, and mechanical properties. *Journal of Natural Fibers*. 2018; 16(3):342-356. <https://doi.org/10.1080/15440478.2017.1423255>
- [90] Shumao L, Jie R, Hua Y, Tao Y, Weizhong Y. Influence of ammonium polyphosphate on the flame retardancy and mechanical properties of ramie fiber-reinforced poly(lactic acid) biocomposites. *Polymer International*. 2010; 59(2):242-248. <https://doi.org/10.1002/pi.2715>
- [91] Singh AA, Genovese ME, Mancini G, Marini L, Athanassiou A. Green Processing Route for Polylactic Acid–Cellulose Fiber Biocomposites. *ACS Sustainable Chemistry & Engineering*. 2020; 8(10):4128-4136. <https://doi.org/10.1021/acssuschemeng.9b06760>
- [92] Kalia S, Sheoran R. Modification of Ramie Fibers Using Microwave-Assisted Grafting and Cellulase Enzyme–Assisted Biopolishing: A Comparative Study of Morphology, Thermal Stability, and Crystallinity. *International Journal of Polymer Analysis and Characterization*. 2011; 16(5):307-318. <https://doi.org/10.1080/1023666x.2011.587946>
- [93] Gassan J, Gutowski VS. Effects of corona discharge and UV treatment on the properties of jute-fibre epoxy composites. *Composites Science and Technology*. 2000; 60(15):2857-2863. [https://doi.org/10.1016/S0266-3538\(00\)00168-8](https://doi.org/10.1016/S0266-3538(00)00168-8)

- [94] Nadi A, Boukhriss A, Bentis A, Jabrane E, Gmouh S. Evolution in the surface modification of textiles: a review. *Textile Progress*. 2019; 50(2):67-108. <https://doi.org/10.1080/00405167.2018.1533659>
- [95] Zille A, Oliveira FR, Souto AP. Plasma Treatment in Textile Industry. *Plasma Processes and Polymers*. 2015; 12(2):98-131. <https://doi.org/10.1002/ppap.201400052>
- [96] Jelil RA. A review of low-temperature plasma treatment of textile materials. *Journal of Materials Science*. 2015; 50(18):5913-5943. <https://doi.org/10.1007/s10853-015-9152-4>
- [97] Mather RR. Surface modification of textiles by plasma treatments. In: Wei Q, editors. *Surface Modification of Textiles*. Edinburgh, UK: Woodhead Publishing; 2009. p. 296-317. <https://doi.org/10.1533/9781845696689.296>
- [98] Zhou N, Yao L, Liang Y, Yu B, Ye M, Shan Z, Qiu Y. Improvement of mechanical properties of ramie/poly (lactic acid) (PLA) laminated composites using a cyclic load pre-treatment method. *Industrial Crops and Products*. 2013; 45:94-99. <https://doi.org/10.1016/j.indcrop.2012.12.014>
- [99] Mazumdar SK. *Composites Manufacturing Materials, Product, and Process Engineering*. Boca Raton, USA: CRC Press LLC; 2002. 416 p.
- [100] Ouagne P, Bizet L, Baley C, Bréard J. Analysis of the Film-stacking Processing Parameters for PLLA/Flax Fiber Biocomposites. *Journal of Composite Materials*. 2009; 44(10):1201-1215. <https://doi.org/10.1177/0021998309349019>
- [101] Jespersen ST, Wakeman MD, Michaud V, Cramer D, Månson JAE. Film stacking impregnation model for a novel net shape thermoplastic composite preforming process. *Composites Science and Technology*. 2008; 68(7-8):1822-1830. <https://doi.org/10.1016/j.compscitech.2008.01.019>
- [102] Hsiao K-T, Heider D. Vacuum assisted resin transfer molding (VARTM) in polymer matrix composites. In: Advani SG, Hsiao K-T, editors. *Manufacturing Techniques for Polymer Matrix Composites (PMCs)*. Cambridge, UK: Woodhead Publishing; 2012. p. 310-347. <https://doi.org/10.1533/9780857096258.3.310>
- [103] Ishikawa H, Takagi H, Nakagaito AN, Yasuzawa M, Genta H, Saito H. Effect of surface treatments on the mechanical properties of natural fiber textile composites made by VaRTM method. *Composite Interfaces*. 2014; 21(4):329-336. <https://doi.org/10.1080/15685543.2013.876322>
- [104] Crosky A, Soatthiyanon N, Ruys D, Meatherall S, Potter S. Thermoset matrix natural fibre-reinforced composites. In: Hodzic A, Shanks R, editors. *Natural Fibre Composites: Materials, Processes and Applications*. Cambridge, UK: Woodhead Publishing; 2014. p. 233-270. <https://doi.org/10.1533/9780857099228.2.233>

- [105] Xia C, Shi SQ, Cai L. Vacuum-assisted resin infusion (VARI) and hot pressing for CaCO_3 nanoparticle treated kenaf fiber reinforced composites. *Composites Part B: Engineering*. 2015; 78:138-143. <https://doi.org/10.1016/j.compositesb.2015.03.039>
- [106] Laurent S, Forge D, Port M, Roch A, Robic C, Vander Elst L, Muller RN. Magnetic Iron Oxide Nanoparticles: Synthesis, Stabilization, Vectorization, Physicochemical Characterizations, and Biological Applications. *Chemical Reviews*. 2008; 108(6):2064-2110. <https://doi.org/10.1021/cr068445e>
- [107] Lazar ST, Kolibaba TJ, Grunlan JC. Flame-retardant surface treatments. *Nature Reviews Materials*. 2020; 5(4):259-275. <https://doi.org/10.1038/s41578-019-0164-6>
- [108] Sharma NK, Verma C, Chariar VM, Prasad R. Eco-friendly flame-retardant treatments for cellulosic green building materials. *Indoor and Built Environment*. 2015; 24(3):422-432. <https://doi.org/10.1177/1420326X13516655>