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Mechanical and durability properties of sustainable composites derived from recycled polyethylene terephthalate and enhanced with natural fibers: a comprehensive review

A. Kumar ✉, **R. Bedi**

National Institute of Technology Jalandhar, Jalandhar (Punjab), India

✉ ajayk.me.19@nitj.ac.in

ABSTRACT

Pursuing sustainable and environment-friendly materials has spurred a paradigm shift in composite materials, with an increasing focus on composite materials with natural fibers as reinforcement. An in-depth study of composites crafted from natural fibers and recycled polyethylene terephthalate (rPET) as matrix is presented in this comprehensive review. The cellulosic fibers offer an eco-friendly alternative to synthetic reinforcements with attributes such as high tensile strength, low density, easy availability in abundance at low cost, and biodegradability. When incorporated into composites, they improve the mechanical behavior, and there is less material consumption, making it preferable for lightweight applications. Conversely, using rPET represents an innovative step in addressing the environmental challenges of using traditional petroleum-based plastics. The rPET matrix, derived from the chemical recycling of used PET bottles, is a promising material for developing environmentally conscious composites. The discussion includes mechanical properties and extends to the durability behavior of these composites, evaluating their resistance to moisture, UV radiation, and temperature variations: durability, resistance to deterioration, and performance of the composite materials under actual situations. The review concludes by exploring current challenges and limitations, highlighting areas for future research and development.

KEYWORDS

sustainability • NFRCs • rPET • mechanical performance • surface modification • durability behavior

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Introduction

This article aims to thoroughly analyze the durability and mechanical performance of composite materials, specifically those incorporating natural fibers with rPET (recycled polyethylene terephthalate) as the matrix. The integration of natural fibers with rPET holds great promise in enhancing the overall properties of composite materials while addressing concerns related to ecological impact and resource depletion. Using natural fibers as reinforcements in polymer composites has gained widespread focus due to their renewable nature, low environmental footprint, and favorable mechanical properties. These plant-based fibers offer unique combinations of strength, stiffness, and lightness, making them attractive candidates for reinforcing polymer matrices. This review explores the diverse range of natural fibers, emphasizing their intrinsic attributes that add to the mechanical performance of polymeric composites. In parallel, the focus on rPET as a matrix material adds another layer of sustainability to the composite system. rPET is a versatile and recyclable matrix option chemically derived from virgin PET bottles. The compatibility between natural fibers and rPET offers a synergistic effect, leveraging the strengths of both components and potentially mitigating the environmental issues interrelated with traditional crude oil-based matrices. The mechanical performance of composite materials is a critical aspect influencing their suitability for various applications. This review delves into the mechanical performance of rPET-based composites utilizing fibers from natural resources. Durability is another pivotal factor determining the practicality and longevity of composites. Natural fiber-reinforced rPET composites are scrutinized for their resistance to aging or degradation in different conditions such as water, moisture, UV radiation, temperature variations, etc. The assessment will extend beyond short-term effects to address the long-term stability and resistance to degradation, offering insights into the durability of these composites in real-world conditions. The aim is to identify existing challenges, pose avenues for future research, and present an overview of the recent developments in green composites. Despite the remarkable progress made in the field, certain obstacles still need to be addressed, including issues related to fiber-matrix interactions, processing techniques, and exploring novel natural fibers. The concluding section provides a roadmap for future research, urging a collective effort to overcome these challenges and further propel the adoption of natural fiber-reinforced rPET composites in engineering and manufacturing applications. In essence, this review sets the stage for a deeper understanding of the mechanical properties and durability aspects of natural fiber-reinforced rPET composites, aiming to contribute to the broader discourse on sustainability and drive innovation

Fig. 1. Types of plant-based natural fibers. Reprinted with permission from [1], © Elsevier 2022

in pursuing a more ecologically responsible future. Traditional composite materials, often reliant on synthetic reinforcements, face scrutiny due to their environmental impact and limited sustainability. Plant or animal-sourced fibers offer a promising solution as reinforcement materials in composites. The motivation for exploring natural fiber-reinforced composites stems from the desire to reduce reliance on non-renewable resources and mitigate environmental concerns. Recycled PET, chemically derived from used PET bottles, emerges as a significant matrix material in this review. Its unique properties, including versatility, recyclability, and compatibility with natural fibers, contribute to composites' overall sustainability and performance.

Fibers from natural resources

Natural fibers come directly from plants, animals, or minerals (Fig. 1). These fibers can be turned into thread, filaments, or rope and are used for making composite materials. Many researchers have shown interest in using natural fibers, which are available in abundance. It has also been economically investigated that integrating farm waste into composite manufacturing is advantageous for applications where products are subjected to environmental degradation [2–4].

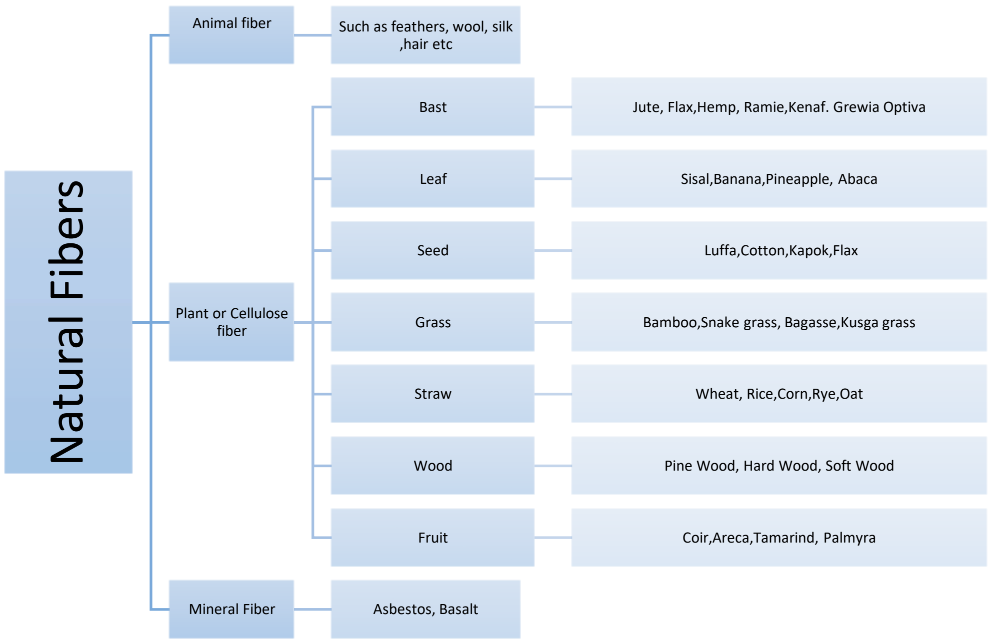

Fig. 2. Categorization of fibers from natural resources. Based on [2]

Classification of natural fibers

The fibers from natural resources can be grouped into three broad categories (Fig. 2):
1. animal fiber: animal fibers include wool, feathers, hair/fur, and bird feathers;
2. plant or cellulose fiber: flax, cotton, kenaf, sisal, jute, and hemp are examples of plant fiber;
3. mineral fiber: asbestos, ceramic fibers, and basalt fibers are naturally occurring mineral fibers.

Composition of natural fibers

Plant fibers primarily comprise cellulose, hemicellulose, pectin, lignin, natural waxes, and other compounds soluble in water. Even though these fibers belong to the same family or kind, their chemical composition varies based on the fiber's source, affecting its mechanical behavior and other properties. The variation in properties is also noticed for the same natural fibers (Fig. 3) depending upon the type of soil they were cultivated, the amount of water and sun received by them, the amount of fertilizer or other chemicals used during the growing stage, the seasonal variation during growth and harvesting for a particular year, i.e., some of the reasons for variability in the properties of same natural fibers are beyond human control as they are cultivated in fields but not produced in factories.

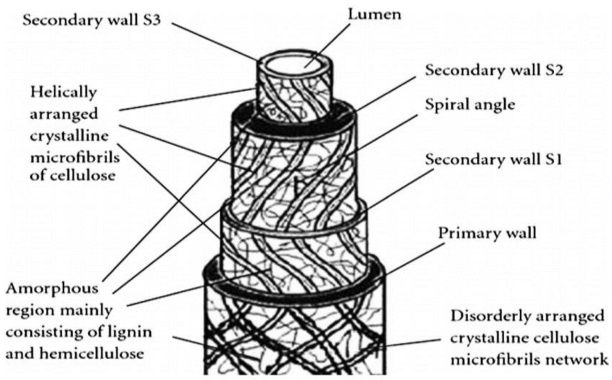

Fig. 3. Natural fiber cell wall's structural organization. Reprinted with permission from [3], © Elsevier 2001

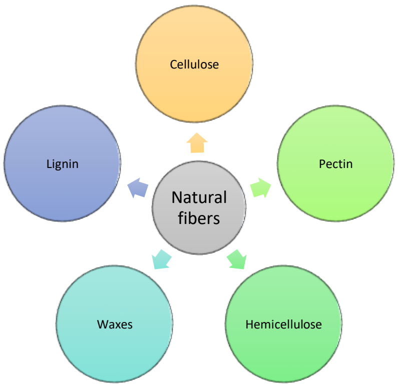

Fig. 4. Constituents of plant-based fibers. Based on [9]

Plant fibers are often known as lingo cellulosic structures containing cellulose, hemicellulose, and lignin [4]. Cellulose is an unbranched macromolecule present in all fiber cell chemical compositions. The amount of cellulose in a plant fiber can substantially impact its functionality and performance in many applications, hemicellulose is a multi-branched polysaccharide polymer composed of sugars which binds cellulose and lignin and in some way also improves the stiffness as well as strength of plant fibers [5]. In the textile and paper industry, bast and jute are favored due to their outstanding cellulose content. Lignin in the plant cell is the second-most found polymer after cellulose. It is a unique biomacromolecule that is highly branched and doesn't have a clear fundamental structure. Plant tissue and individual fibers (Fig. 4) get their stiffness and strength from the lignin (Table 1). It also protects the carbohydrates and sugars from chemical and microbial attack [6–8].

Table 1. Chemical composition of plant-based natural fibers (wt. %) [10]

Fiber	Cellulose	Hemicellulose	Lignin	Pectin	Waxes
Hemp	70–75	17.5–22.5	3.7–5.7	0.9–1.0	0.7–0.8
Jute	60–71	13.5–20.5	12–14	0.2–0.3	0.4–0.5
Ramie	68–76	13–16.5	0.6–0.8	1.9–2.0	0.2–0.3
Nettle	86	4.0	5.4	0.6	3.1
Sisal	67–78	10–14	8–11	10	10–22
Pineapple	80–83	15–20	8–12	1–3	8–15
Cotton	85–90	5.7	0.7–1.6	1.0	20–30
Coir	36–43	0.15–0.25	41–45	5.2–16.0	30–49

Merits and demerits of fibers from nature

One of the primary motivations for incorporating natural fibers in composite materials is their renewable nature and eco-friendly characteristics. Unlike synthetic fibers, natural fibers are sourced from readily available raw materials, contributing to sustainability. Cultivating these fibers requires less energy and results in lower carbon footprints than synthetic reinforcements. The biodegradability of natural fibers further enhances their environmental appeal, reducing the long-term impact on ecosystems.

Merits

Natural fiber's intrinsic properties, such as low density, low cost, easy availability, high toughness, strength, stiffness, outstanding acoustic and thermal insulating capabilities, and high electrical resistance combined with minimum health impact and comparable mechanical performance, make them suitable candidates for composite manufacturing. They also have a low adverse effect on cutting tools and machines [11]. Traditionally, rice and wheat straw fibers were used to strengthen mud bricks, which are still used in various applications (Fig. 5).

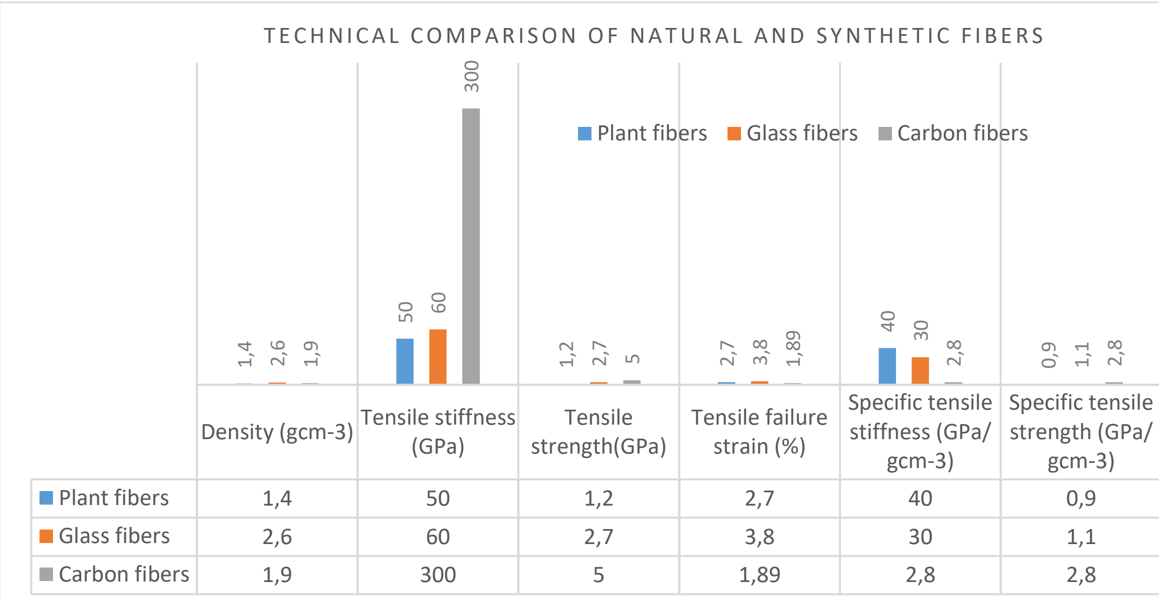

Fig. 5. Comparison (average values) of standard natural and synthetic fibers. Based on [12]

Demerits

Despite their numerous benefits, natural fibers have drawbacks, such as poor compatibility and the matrix's non-uniform wetting of the fibers, high moisture absorption, and deterioration due to heat during manufacturing. These make them unsuitable for various composite applications [13]. One of the problems experienced due to poor wetting of the hydrophilic natural fibers by hydrophobic organic polymer matrix is weak adhesion at the reinforcement-matrix interface [14]. Therefore, the natural fibers need surface treatment before the composite is made from them. Several treatments and techniques are available to modify the surface of natural fibers to form a strong bond between the fibers and matrix. For example, mercerization, also called alkali treatment, is a chemical process used to clean and alter the surface of plant-based fibers to increase interfacial adhesion with the matrix [15]. Some of the merits and demerits of natural fibers are listed below (Fig. 6).

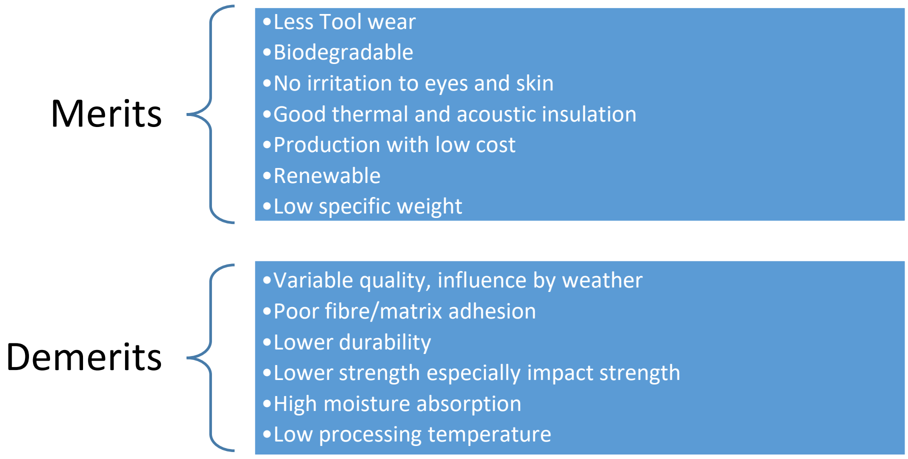

Fig. 6. Merits and demerits of natural fibers. Based on [16]

Comparison of natural and synthetic fibers

The comparative analysis of natural and synthetic fibers (Fig. 7) reveals significant differences in environmental impact, mechanical properties, and applications. Natural fibers, such as jute and flax, are increasingly favored for their eco-friendliness and lower greenhouse gas emissions, while synthetic fibers like polyester are associated with higher environmental costs.

Despite the advantages of natural fibers, challenges still need to be addressed, such as variability in quality and performance compared to synthetic alternatives. Balancing these factors is crucial for their use in composites.

The interfacial adhesion between natural fibers and polymer matrixes (Fig. 8) is crucial for enhancing composites' mechanical strength and overall characteristics. This adhesion facilitates effective stress transfer, which is essential for the all-around performance of the composite material. Interdiffusion, electrostatic adhesion, and mechanical interlocking contribute to adequate bonding [25]. Coating fibers with acrylic resins can significantly enhance interfacial shear strength, improving load transfer [26]. Chemical treatments and surface roughening of fibers have increased compatibility with polymer matrices [27,28]. However, natural fibers often exhibit poor moisture resistance

Table 2. Comparison of natural and synthetic fibers

	Natural fibers	Synthetic fibers
Environmental impact	These generally exhibit lower environmental impacts across various categories, including fossil resource scarcity and global warming potential.	Synthetic fibers contribute significantly to Eco-toxicity and require extensive energy for production [17].
Mechanical properties	Natural fiber-reinforced composites demonstrate favorable mechanical properties, such as high flexibility and lower weight, making them suitable for non-structural applications [18].	These provide superior strength and durability but at a higher environmental cost [19].
Durability aspects	These are hydrophilic, leading to higher moisture absorption, which can compromise durability, but at the same time, natural fibers are biodegradable and more sustainable, making them preferable in eco-conscious applications despite their lower durability [20,21].	Synthetic fibers maintain structural integrity under varying environmental conditions [22].
Applications	Natural fibers are increasingly used in sustainable construction and automotive sectors.	Synthetic fibers dominate in high-performance applications due to their enhanced properties [23].

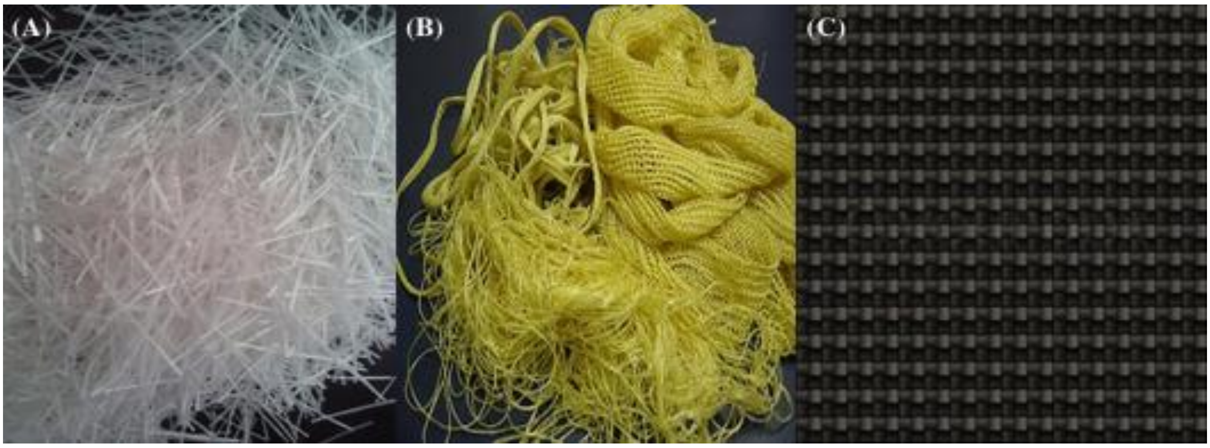

Fig. 7. Displaying synthetic fibers (A) glass, (B) kevlar, and (C) carbon fibers. Reprinted with permission from [24], © Elsevier 2017

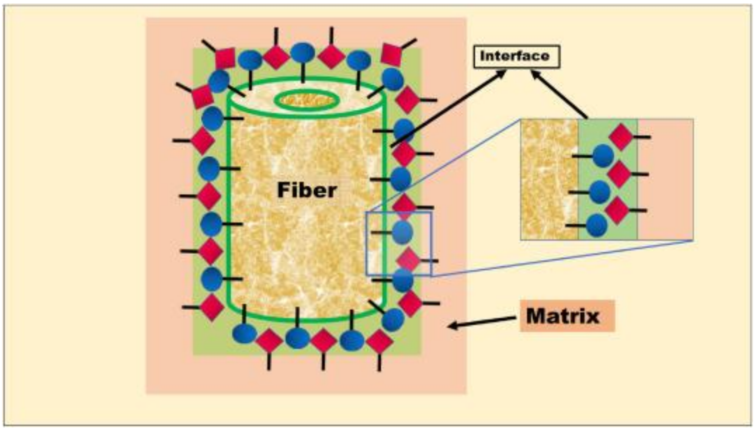

Fig. 8. Interfacial chemical bonding mechanisms between fiber and matrix. Reprinted with permission from [29], © Elsevier 2022

and variability, leading to weak fiber-matrix adhesion [28]. Addressing these challenges through strategic modifications can lead to composites with superior mechanical performance.

While enhancing interfacial adhesion is vital for improving composite strength, it is also essential to consider the balance between fiber properties and matrix characteristics to achieve optimal performance in various applications.

Surface modification of natural fibers

Naturally found fibers are polar and hydrophilic [30], whereas the polymer matrix is hydrophobic, which results in poor interface bonding between the two. Due to this weak adhesion at the interface of cellulosic fiber and the polymer matrix, the overall performance of the fabricated composites is compromised [31,32]. Plant-based fiber surfaces can be surface-treated to improve their hydrophilic nature [33]. Treated natural fibers improve bonding at the fiber interface and the matrix, enhancing stress transferability [34]. The adherence of plant fibers to the inorganic matrix can be enhanced by (i) altering the top layer of the fibers, (ii) altering the polymer matrix, or (iii) altering both. Surface modification with physical methods such as plasma, ultrasound, ultraviolet light, and treatment with chemicals like silane, alkali, acetylation, benzoylation, and sodium chloride helps to improve the bonding of plant-based fibers with polymer matrix [35,36].

Surface modification by physical methods

Physical modification procedures like plasma treatment attempt to roughen the plant-based fibers' surface and remove surface contaminants such as oils, waxes, and pectin to increase the mechanical bonding of fiber with the matrix [12]. The Ultraviolet treatment improves the polarity of the fiber surface, resulting in enhanced wettability by the matrix and overall strength of the composite [37,38]. The fiber-beating increases fiber surface area, defibrillation, and mechanical interlocking, improving the strength of natural fibers [39].

Corona and plasma treatments

The corona treatment raises the surface energy and oxides on the plant fibers' surface, making it blend more efficiently with the polymer matrix [40]. Plasma treatment alters the surface of plant-based fibers by eliminating poorly connected surface layers and generating new functional groups [41].

Surface modification by chemical methods

The chemical treatment removes unwanted constituents in plant fibers, such as hemicellulose, waxes, lignin, and pectin, improving matrix adhesiveness's overall physical and chemical characteristics [42]. A study by Maya Jacob John and Rajesh D. Anandjiwala reports that lignocellulosic fibers are naturally polar and water-loving, but most thermoplastics aren't polar; mixing them is hard. Another problem with using natural fibers to make durable composites is that they aren't very resistant to bacteria and mounds, making them prone to rotting and causing significant issues when shipping, storing, and making composites. Chemical treatments can fix most of these problems associated with natural fibers, making them suitable for composites with polymer matrix [43].

NaOH treatment

Treatment with alkali, such as mercerization, is one of the easiest, most cost-efficient, and most effective methods for enhancing plant-based fiber adherence to the polymer matrix. This approach uses sodium hydroxide (NaOH) to alter the cellulose composition of natural fibers [44]. The fibers' surfaces become clean and homogeneous. An optimal alkali concentration should be attained, as substantial concentrations can weaken and destroy the fibers. The diameter of the fibers is decreased at an optimum alkali concentration, resulting in improved adhesion of fibers with matrix due to enhanced fiber surface area and improved aspect ratio [45].

The equation below shows the reaction of plant-based fiber with NaOH:

$$\text{Fiber} - \text{OH} + \text{NaOH} = \text{Fiber} - \text{O} - \text{Na} + \text{H}_2\text{O}.$$

The surface roughness of sisal fibers was significantly increased after alkali treatment, resulting in fibers with lower diameters, most likely due to eliminating noncellulosic chemicals. Furthermore, sisal fibers treated with NaOH solution produced somewhat greater contact angles with water drops, implying enhancement in wettability with hydrophobic polymer matrices [46,47]. Studies have shown that alkali treatment increases the proportion of amorphous cellulose at the expense of crystalline cellulose and removes hydrogen bonding in the network structure [48,49]. Huda et al. [50] discovered that the NaOH-treated pineapple leaf fibers reinforced PLA composites had better mechanical qualities than the untreated leaf composites. Edeerozey et al. [51] found that 6 % NaOH was optimal for superior mechanical attributes compared to stock kenaf fibers. Fiore et al. [52] calculated the effect of NaOH treatment on the mechanical performance of kenaf fiber-reinforced epoxy composites, stating that the treated composite had better mechanical properties, according to the study. Nurul Munirah Abdullah and Ishak Ahmad [53] prepared coconut fiber/polyester composite using alkali, silane, and silane on alkalized fibers to increase the adhesion between coconut fiber and polyester resin. The tensile characteristics of coconut fiber were significantly improved by surface treatment.

Additionally, it was noted that composites made using treated fibers had less water absorption capacity than untreated fiber composites. Overall, the current literature [54] suggests that treating plant-based fibers with alkali concentrations of 5 to 8 % improves the mechanical behavir of the composites.

Silane treatment

In this method, silane coupling agents cover micropores on the fiber surface. Silane (SiH_4) is an inorganic compound in nature is made up of both hydrophilic and hydrophobic compounds connected to the silicon molecule, which can be thickened by adding a coagulant agent to make silanol (SiOH) groups (Fig. 9) and subsequently siloxane bridges [55].

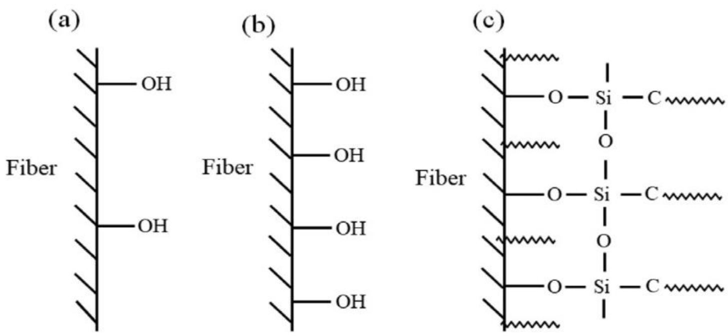

Fig. 9. Schematic showing how chemicals interact with the fiber when it is (a) untreated, (b) treated with NaOH, and (c) treated with Silane. Reprinted with permission from [56], © Elsevier 2020

In a research conducted by Y. Liu et al. [57], fibers from corn waste were treated with silane solution. Silane's effects on the chemical, morphological, and mechanical properties of Corn Stalk Fibre (CSF) and CSF-reinforced polymer composites were studied. Pull-out tests confirmed the improved interfacial adhesion between silane-treated sisal fiber and matrix; silane-treated fibers had higher interfacial shear strength than untreated fibers.

Peroxide treatment

Researchers from diverse fields have investigated treating natural fibers with peroxide and the mechanical characteristics of polymer composites reinforced with these peroxide-treated fibers (Fig. 10). Free radicals are created when the peroxide breaks down: RO + Cellulose-H = R-OH + Cellulose.

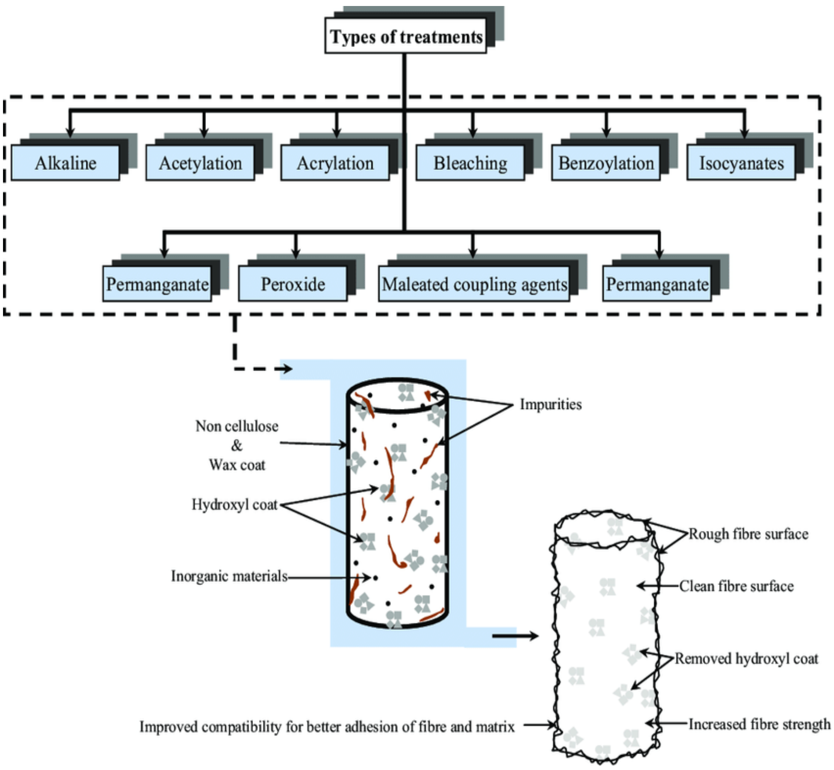

Fig. 10. Changes in the surface of cellulosic fiber after various treatments. Reprinted with permission from [58], © Elsevier 2015

Benzoylation treatment

Benzoyl chloride is also used in modifying the surface of natural fibers. This includes benzoyl ($C_6H_5C = O$), which is responsible for lowering the hydrophilicity of the plant fiber and improving bonding with the polymer matrix. Joseph et al. [59] used NaOH and benzoyl chlorite (C_6H_5COCl) solution for surface modification of sisal fibers and reported improved hydrophobicity. Mixing two or more chemicals for surface treatment, such as alkali with silane, improves mechanical properties better than just one treatment. Chemical agents like maleic anhydride grafted polypropylene (MAGP) and zein also improve flexural strength [60]. A study [61] investigated the influence of different surface chemical treatments (Fig. 11) on the mechanical characteristics and durability of jute fiber-reinforced epoxy composites (JFRECs). It was discovered that the combined chemical treatment is far better than individual treatments. Water absorption rates for composites with chemically treated fibers were lower than those for untreated fiber composites.

Other miscellaneous treatments

In composites reinforced with natural fibers, various chemical treatments are used to modify the surface of the fibers, but on the other hand, inadequate treatment with chemicals could damage the structure of the natural fibers, thereby considerably decreasing their mechanical properties [62]. R. Sepe et al. [63] reported that both tensile and flexural strengths of composites

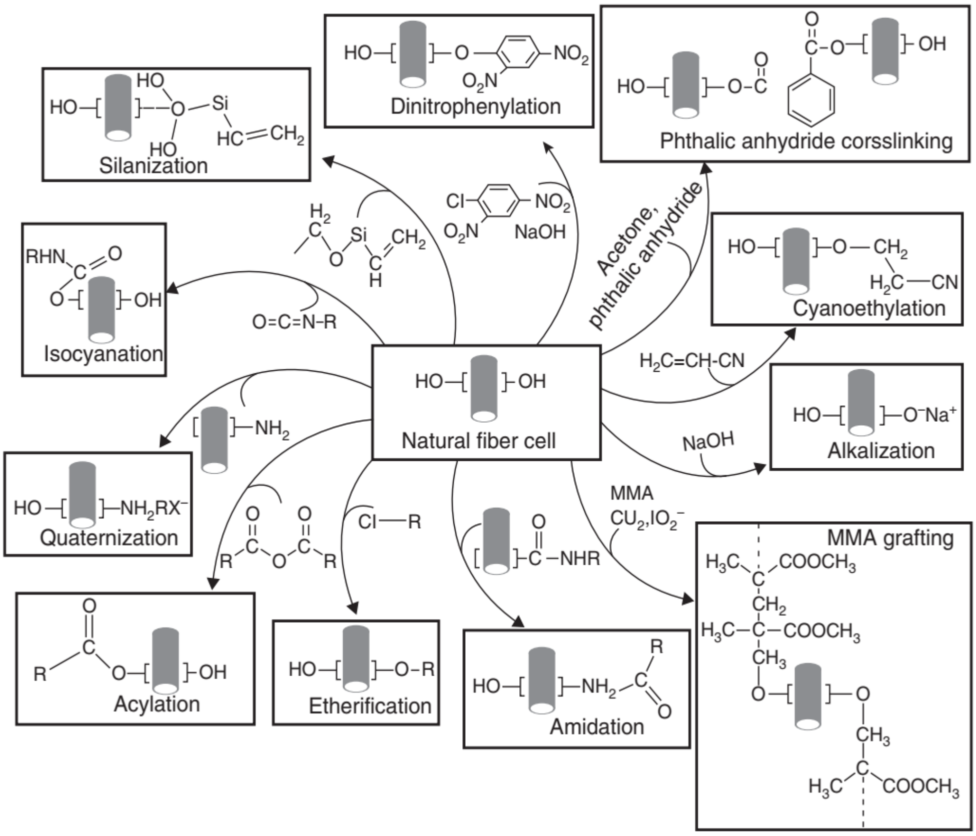

Fig. 11. Basic chemistry of surface modification treatments of plant fibers. Reprinted with permission from [10], © Elsevier 2015

reinforced with hemp fibers improved when these fibers were treated with (3Glycidyloxypropyl) trimethoxysilane. The effect of an eco-friendly chemical treatment consisting of a 10 % solution of sodium bicarbonate on the resistance to aging in some marine conditions of epoxy-based composites reinforced with flax and jute fibers was studied by V. Fiore et al. [64]. The samples prepared were put in a salty and foggy spray environment for around 60 days. The experimental campaign showed that the sodium bicarbonate treatment makes plant-based fibers and epoxy matrix stick together in a better way, which helps treated laminates retain their flexural properties better than untreated laminates during salt fog exposure. M. Bayart et al. [65] investigated the adhesion at the interface of surface-treated flax fibers and polylactic acid (PLA) utilizing lignin and tannin, which are eco-friendly polyphenolic chemicals generated from the waste of paper industries. The findings demonstrated that the interfacial strength of composites was significantly increased by using a coating of solutions containing 1 % (wt/V) tannin/lignin. Tannin demonstrated improved compatibility with PLA and flax fibers, increasing the composites' ultimate flexural strength and interlaminar shear strength by 17 % and 29 %, respectively. Research work was carried out by A. L. N. Inácio et al. [66] to test the mechanical and thermal aspects of composite materials with bamboo fiber-reinforced recycled talc-filled PP/EPDM that has been grafted with maleic anhydride (PP-g-MAH). The composite was held at 90 °C for seven days in a chamber with hot air moving through it. Improvements in tensile strength, flexural strength, and fatigue life were noticed (Fig. 12).

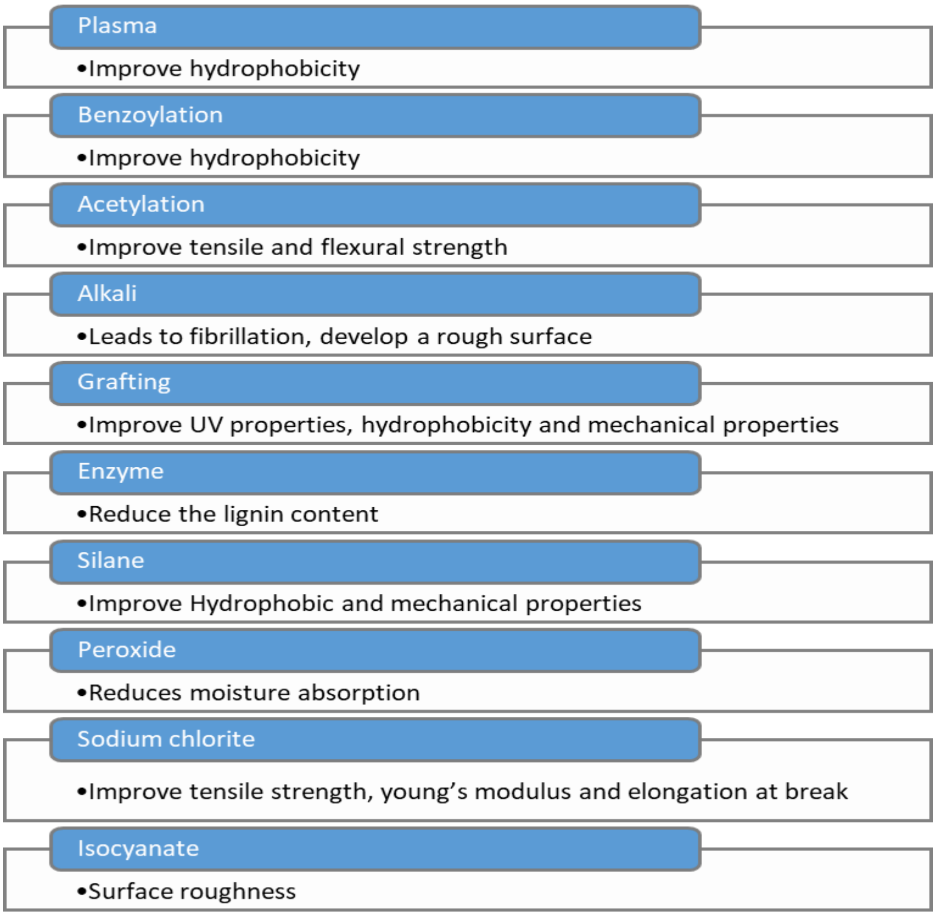

Fig. 12. Effect of various surface treatments on natural fibers. Based on [67]

S. Satapathy and R.V.S. Kothapalli [68] fabricated composites based on recycled high-density polyethylene (RHDPE) / fly ash cenospheres (FACS)/banana fiber (BF) to enhance the commercial value of these waste products. Maleic anhydride-grafted HDPE (MA-g-HDPE) was also used as a compatibilizer to promote the dispersion of fibers within the polymer matrix and the compatibility of the matrix with fillers. The addition of 7.5 wt. % FACS, 30 wt. % BF, and three wt. % MAg-HDPE to the RHDPE matrix improved the tensile strength by 17 %, the tensile modulus by 188 %, the flexural strength by 38 %, the flexural modulus by 159 %, and the hardness by 37 %. The surface of raw natural fibers can be modified by various treatments to improve the bonding of these fibers with the polymer matrix.

Recycled PET as matrix material

The matrix material in composite structures is pivotal in calculating the composite's overall performance, durability, and sustainability. Polyethylene terephthalate (PET) (Fig. 13) is a popular technical polymer. Because of its multiple vital characteristics it is used in household items, textiles [69], and single-use bottles such as water, cold drinks, juices, etc. PET or polyethylene terephthalate is a functional polymer with many applications, and the market for PET goods has been gradually rising in recent years. This has resulted in a considerable amount of non-biodegradable PET waste [70], which poses significant threat to the environment due to pollution and depletion of resources. PET waste does not decay and can take hundreds of years in the environment. Single-use plastic in the form of PET significantly contributes to garbage in landfills. Moreover, PET is derived from fossil fuels, especially crude oil, and as such, extensive use of PET means more resource depletion and negative environmental consequences. Therefore, the safe method of mitigating the harmful impacts of PET waste is to encourage and improve recycling efforts. Various recycling systems have emerged to address the pollution caused by PET [71]. PET waste can be recycled mechanically and chemically (Fig. 14), with glycolysis being the most successful [72].

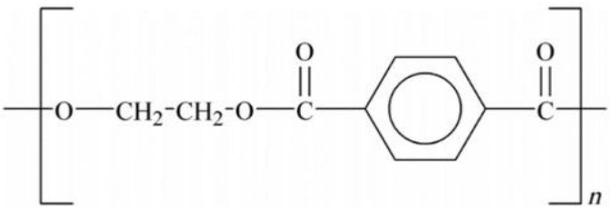

Fig. 13. Chemical structure of PET. Based on [73]

Consequently, we need to increase waste collection activities and establish efficient recycling processes, promoting a circular plastics economy and decreasing the amount of plastic garbage entering the environment. One solution that holds promise is chemical recycling, which could turn PET waste back into valuable raw materials and reduce the negative environmental impacts associated with the disposal of plastic waste. The recycled resin (rPET), thus derived from chemical recycling, has properties akin to those of virgin PET and can be used as a matrix for manufacturing composite materials. Recycled PET exhibits excellent processability, allowing for ease of manufacturing and compatibility with various reinforcement materials. Understanding the molecular structure, processing methods, and fundamental characteristics of rPET is crucial for comprehending its role within the composite system.

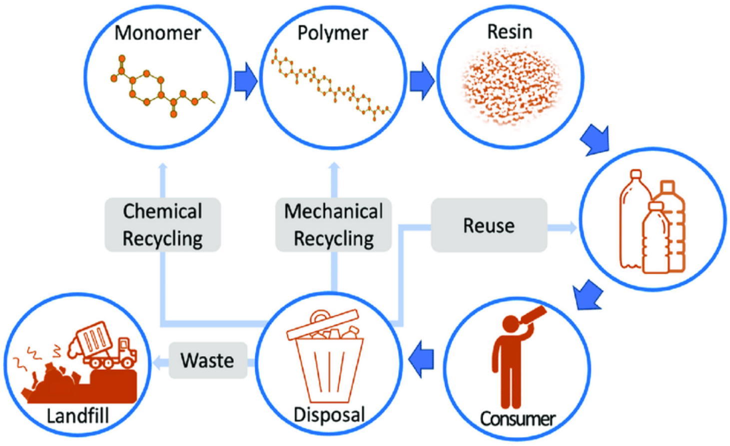

Fig. 14. Life cycle of a typical PET product [74]. Reproduced with the kind permission from https://www.mdpi.com/openaccess#Permissions

Advantages of rPET as matrix material

Recycled PET brings several advantages (Fig. 15), making it a sought-after matrix material for natural fiber-reinforced composites:
1. versatility: rPET can be tailored to meet specific application requirements by adjusting its chemical composition and processing parameters. This versatility allows for the formulation of composites with a wide range of mechanical and thermal properties;
2. process ability: The ease of processing rPET facilitates various manufacturing techniques, including injection molding, vacuum bagging, infusion, compression molding, and extrusion. This processability contributes to the efficiency and scalability of composite production;
3. mechanical performance: rPET offers commendable mechanical properties, including tensile strength, toughness, and resistance to wear. These characteristics enhance the overall mechanical performance of composites, especially when combined with natural fiber reinforcements.

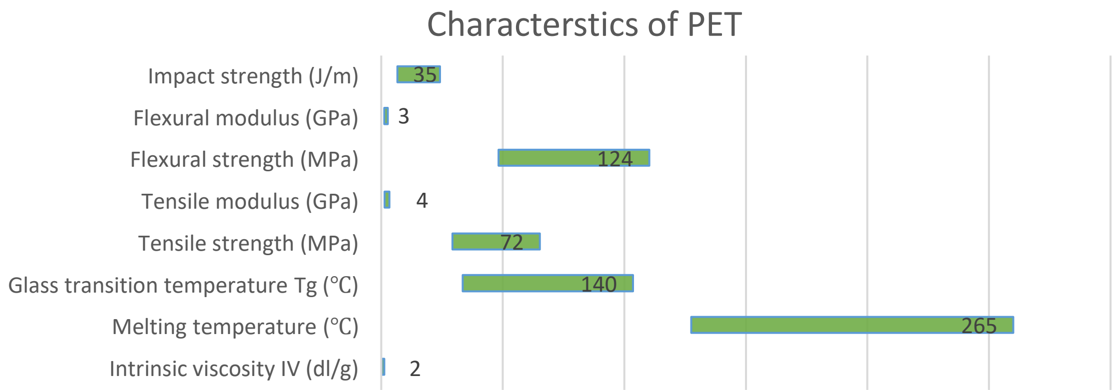

Fig. 15. Properties of PET (polyethylene terephthalate). Based on [75]

Several polymer matrix composites utilize recycled polyethylene terephthalate (RPET) as the matrix material. This also implements the EU's mandate, which requires all plastic packaging to be recyclable or reused by 2030 [76].

Mechanical and durability properties of natural fiber reinforced composites based on recycled PET

Composites based on recycled PET and reinforced with wood flour and polymeric cobalt were aged by Przemysław P. Aczkowski et al. [77] and found that the mechanical performance of the composites decreased drastically as the amount of wood increased. Composites lost their shine and became more brittle as they aged. In a work by J. Mikes [78], the reinforcing impact of basalt fibers and talc powder on recycled PET was examined using the rheometric system, tensile tests, and DSC, comparing the processing and usability characteristics of reinforced composites to those of an unreinforced matrix showed improvement.

A. Dehghani et al. [79] developed a composite based on recycled polyethylene terephthalate (rPET) reinforced with surface-treated 5 % NaOH at 100 °C for one h, date palm leaf fiber (DPLF). Dynamic mechanical analysis (DMA) showed that adding DPLF to the rPET matrix made the composites more robust, the crystallization temperature rose, and the composites were more crystalline. E. Corradini et al. [80] examined composites made from recycled PET and bagasse fiber from sugarcane, the composites' interfacial behavior was studied, it was reported that the bagasse sugarcane fiber reinforcement decreased the tensile strength and elongation of PET but had no effect on the tensile modulus. A study by M.E. Cinar and F. Kar [81] sought to create composite materials using marble dust and PET (polyethylene terephthalate) bottles. In a screwed extruder, marble dust is combined with PET waste particles to create a composite material. Vickers hardness values increased together with the rise in the marble ratio in composite material structure. After a particular ratio (25 % marble dust), it was seen that particle size has significant impact on hardness. Composites were fabricated from unsaturated polyester resin (UPR), which has been synthesized from recycled polyethylene terephthalate (PET) and empty fruit bunch fibers (EFB), as reported by C. Tan et al. [82]. The effects of treating EFB's surface with silane, sodium hydroxide solution (NaOH) and maleic anhydride (MA) were investigated. The testing findings revealed that the chemically modified EFB had more excellent tensile and impact strength than untreated EFB. The silane treatment had the best results, followed by the MA and NaOH treatments, which raised the tensile strength by approximately 21, 18, and 13 %, respectively. It was also reported that treating EFB fibers with these chemicals increased their interfacial bonding with the polymer matrix compared to neat UPR/EFB composites. Sara Madadi Ardekani et al. [83] studied the mechanical and chemical characteristics of recycled PET (rPET) composites reinforced with recycled newspapers to ascertain the impact of NPF. The impact strength of rPET was dramatically increased by adding SEBS-g-MA (10 phr). With adding more fiber, the Young's and flexural moduli improved, and at 5 wt. % fiber loading, the tensile and flexural strengths displayed the most significant values.

In a study by R. Passos et al. [84], different plasticizer mixes were employed to reduce the Tm of recycled PET, allowing the materials to be processed at a temperature much below the melting point of the neat polymer. Without a plasticizer, this temperature is high enough to cause considerable breakdown of the lignocellulosic sisal fiber. Furthermore, the inclusion of plasticizers reduced the melt viscosity of PET, which resulted in increased wetting of the fibers by the polymer. Additionally, the content of the fiber, surface treatment of the fiber, compatibilizers, recycled plastic, etc., affect the qualities of natural fiber-reinforced rPET composite as a whole [85]. The development of a composite filled with wood flour (WF) based on a PP/r-PET blend was reported by Dairi et al. [86] morphological, mechanical, and physical properties were studied and reported that higher WF loading decreased mechanical properties in PP/r-PET/WF composites except for flexural and tensile modulus due to WF clumping and weak interfacial adhesion. However, with MAPP, these composites showed improved tensile and flexural strength and modulus, as MAPP promoted even WF distribution and better adhesion. Additionally, while higher WF content increased water absorption, adding MAPP reduced it by improving PP-WF adhesion and reducing voids. Uncoated and epoxy-coated kenaf fiber reinforced recycled polyethylene terephthalate (rPET) composites at weight percentages of 5, 10, and 15 % by M.M. Owen et al. [87]. DSC and TGA were utilized to analyze the composites' thermal phase and decomposition. Coated kenaf composites were thermally stable, showing no fiber degradation. Mechanical characteristics of coated kenaf composites were better than untreated and improved with fiber content. In a work by P. Chaiwutthinan et al. [88], WPCs were made by combining rPET and PBAT with 5–30 % wood flour (WF). The results showed improvement in impact strength and elongation compared to neat rPET at 5–15 wt. % WF of WPCs. The 20–30 wt. % WF composites showed greater tensile strength than the clean blend. In research by S.T. Mosavi-mirkolaei et al. [89], recycled HDPE/LDPE blends were strengthened with PET microfibrils to make composites. The results reported improvement in the crystallinity of the plastics (HDPE and LDPE) in these wood plastic composites with the addition of PET microfibrils, E-GMA, and WF. The mechanical properties of the WPCs with PET and E-GMA also improved.

Y. Martinez Lopez et al. [90] aimed to fabricate a wood-plastic composite board sawdust, high-density polyethylene, thermoplastic polyethylene terephthalate, polypropylene, and calcium carbonate were raw materials. The results showed better physical and mechanical properties. Ahmad et al. [91] prepared sawdust/UPR composite and investigations into the effects of treatment with alkali, filler content, and filler size on mechanical properties and water absorption of the composites revealed that mechanical strength of composites increased with increasing filler contents, the particle size of the sawdust significantly influenced the mechanical parameters with smaller sawdust providing higher strength and modulus which may be attributed to increased surface area for filler-matrix interaction.

Recycled high-density polyethylene (HDPE) and recycled polyethylene terephthalate (rPET) reinforced with rice husk (RH) composites were fabricated by R. S. Chen et al. [92]. Dimensional stability, swelling, and mechanical behavior of these composites were evaluated as functions of bio-filler content both water absorption and swelling indicated a linear rise with RH concentration. The composites' thickness experienced the most swelling, followed by their width and length. Compatibilized recycled polymer blend rPB-based composites showed less water absorption and dimensional instability than neat rPB-based composites. I. Ahmad and T.M. Mei [93] prepared a UPR composite reinforced with sawdust from rubber wood with 10 % NaOH treatment. Researchers looked at how the surface treatment and amount of filler affected the composite's mechanical properties and how much water it could hold. The results show that the tensile modulus increased as the filler amount increased. The alkali treatment made the rubber wood sawdust stick better to the UPR matrix and improved the mechanical properties. The flooring tiles based on recycled PET reinforced with natural fibers meet the required compression strength. They may be utilized as possible candidate materials as they are light in weight, economical, and environmentally friendly [94]. I. Ahmad and A. Ramli [95] produced UPR composite based on recycled PET reinforced with sawdust. The results show that as the amount of sawdust filler treated with alkali went up, the tensile strength and bending strength came down as the filler is hydrophilic; adding it makes the composites absorb more water.

Applications, challenges and limitations

The use of plant and animal-sourced natural fibers as reinforcing materials in polymer composites has been investigated around the globe. Natural fibers are becoming increasingly popular for cost-effective buildings and transportation [96]. These polymer composites, which have natural fibers as reinforcement, have the potential to be used in various applications (Fig. 16, 17) and are viable alternatives to synthetic fiber-reinforced polymer composites. As the general public becomes more concerned about the environmental difficulties caused by non-degradable and non-recyclable materials, the industry prefers the shift to natural fibers [97]. To do this, the US and Europe have given specific rules about vehicles that have completed their useful lives. The European Commission put into place the "European Guidelines 2000/53/EG", which said that by 2005, 85 % of an automobile's weight should be recyclable. By 2015, this number rose to 95 %. This kind of law is a big reason NFRCs are becoming more popular.

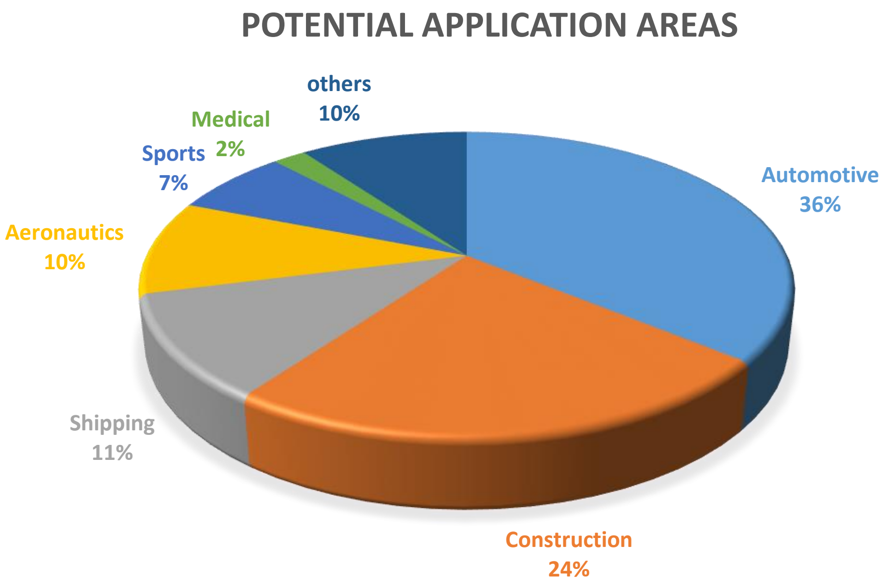

Fig. 16. Application potential of NFRCs. Based on [104]

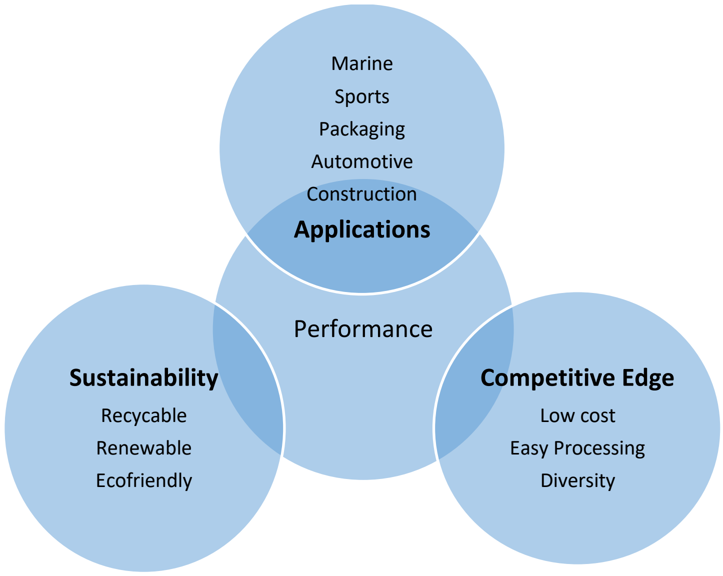

Fig. 17. The performance of commercial success for NFPCs. Based on [105]

Polymers based on natural fibers have been used in many car models over the past 20 years, initially in Europe and then in North America. These include boot liners, sun visors, oil and air filters, rear seat backs, exterior underfloor paneling, internal engine covers, door panels, trays, utility racks, and console panels [98]. At the same time, we can reduce our reliance on crude oil by building more fuel-efficient cars, not just by putting efficient engines in them but also by making them lighter so that they need less fuel [99]. Strength and safety must not be compromised to reduce vehicle weight. High-strength materials are usually heavier. It seems more complicated to achieve good strength with lightweight materials than conventional ones. The only practical approach is to use plant-based materials, which might replace steel. The automotive industry is using plant-based materials to make car components [100].

The avenues of application of natural fiber-reinforced composites are growing at a pace in a large number of engineering and multidisciplinary areas [101] owing to their intrinsic properties such as low specific weight, comparatively high strength, relatively low production cost, resistance to corrosion, and fatigue NFRCs are widely used in automotive applications, structural components, packaging, building, electronic and electrical industries, aircraft, sports and recreational equipment, boats, machinery, office supplies, etc. [102].

Compared with composites reinforced with synthetic fibers such as glass fibers, it was found that composites with natural fibers were better for industrial use. Also, because natural fibers are now seeing a place in different engineering and construction industries, it helps rural areas grow their economies [103].

NFPCs have also been employed as a replacement for synthetic fiber (Fig. 18) in the aircraft sector for interior paneling, goods for funeral and packaging items, railings of ships and boats, packaging cases for laptops, mobile phones, and other lightweight electronic items and toys [6]. Bamboo culms are interconnected to form a multi-culm composite part to withstand heavy loads and provide sustainable green building resources [105]. A study carried out by Saxena et al. [106] revealed that Indian railways make floor and roof panels, berths, walls, and modular toilets in the coaches using natural fibers, which is essential for getting high speeds, low power consumption, less weight, less inertia, decreased wear of the tracks and other things. Swamy et al. [107] examined fiber usage from the areca plant as reinforcement in polymer composites. They discovered it to be an excellent acoustic material for structural applications. Maheswari et al. [108] suggested that composites made from high-density polyethylene (HDPE) reinforced with Borassus can be utilized in various secondary structural applications and as suitable packaging materials.

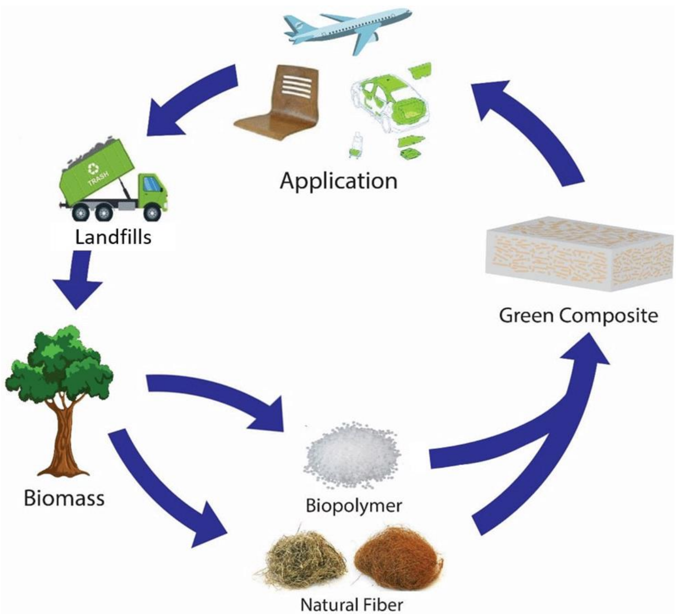

Fig. 18. Life cycle of green composites [109]. Reproduced with the kind permission from https://materialsopenresearch.org/about/policies/#licenses

Furthermore, some researchers fabricated cellular beams and plates reinforced with fibers from jute, hemp, and flax plants based on unsaturated polyester matrix as load-bearing structural members, demonstrating that NFRCs can be used as load-bearing members in structural applications [110]. Ghavami [111] used bamboo fiber to produce structural concrete parts. Rethinam and Sivaraman [112] examined the acoustic qualities of areca plant fiber and found it better than plywood, making it more suitable for furniture applications. Board made out of banana fiber is a potential substitute for plywood and medium-density boards made of wood [113]. RH-filled composites will likely be used in building and construction as alternatives to other materials because they are light and inexpensive [114]. Zawawi et al. [115] conducted a microwave property comparison study on PLA composites reinforced with plant-based fibers to assess their potential as alternatives to typical printed circuit boards (PCBs) for electronic industries. They reported that kenaf/PLA and RH/PLA composites can be used for microwave applications. Wind and solar energy are the most popular renewable energy sources that are being looked at as alternatives to oil-based energy sources. The cost of making the molds for the parabolic trough collector (PTC) makes it expensive. In these situations, adding plant-based fibers is an excellent way to make up for the overall cost of the composite materials. It was recently shown that the price of trough collectors can be reduced by adding natural fibers [116]. Carbon and glass fiber hybrids are the most used turbine blade fillers. Price and weight are the key disadvantages of carbon and glass fibers. Utilizing comparably less expensive fillers that are significantly lighter than carbon fibers can increase the efficiency of wind turbines. A recent study indicated that it is possible to substitute 20–30 % of carbon fibers in turbine blade composites (Fig. 19) with basalt fibers without adversely affecting the composite's characteristics [117].

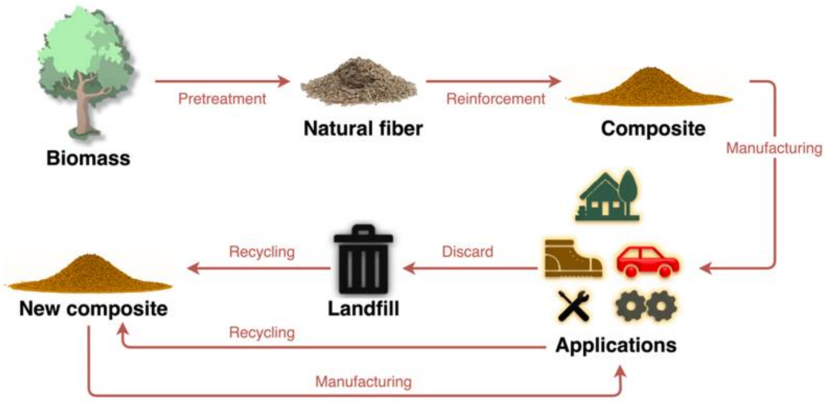

Fig. 19. Recycling natural fiber-reinforced composites toward a circular economy. Reprinted with permission from [118], © Elsevier 2022

One of the primary challenges in natural fiber-reinforced composites lies in achieving optimal interactions at the fiber-matrix interface [119]. The composite's effectiveness heavily depends on the strength and compatibility of this interface. More studies that explore methods to enhance interfacial adhesion, such as surface treatments, coupling agents, and modifications to the matrix formulation, are needed. Understanding the challenges of achieving a solid interface is essential for overcoming mechanical performance limitations and ensuring these composites' longevity. The processing techniques employed in fabricating natural fiber-reinforced rPET composites are crucial in determining their mechanical properties and overall performance. Challenges arise in achieving uniform fiber dispersion, minimizing defects, and optimizing processing parameters to ensure the desired material characteristics. While traditional natural fibers like jute, flax, hemp, and kenaf exhibit favorable properties, their availability, processing, or mechanical performance limitations pose challenges. Exploration of novel natural fibers is needed to address these limitations. Research efforts focusing on alternative natural fibers with unique characteristics are required, shedding light on the potential for diversifying fiber options and overcoming inherent challenges associated with existing choices. The environmental impact of natural fiber-reinforced rPET composites, while generally having a lower carbon footprint than traditional materials, introduces challenges to sustainability and end-of-life considerations. Addressing the environmental impact of composite materials is integral to ensuring their alignment with broader sustainability goals and minimizing their ecological footprint. By understanding the intricacies of interactions at the fiber-matrix interface, refining processing techniques, exploring novel natural fibers, and considering environmental factors, researchers and practitioners can pave the way for overcoming these challenges.

Conclusions

In exploring the mechanical performance and durability behavior of natural fiber-reinforced rPET composites, this review has uncovered valuable insights into the promising realm of sustainable materials. The interfacial adhesion, compatibility, and interaction of the fiber with the matrix play a crucial role in NFRC's mechanical behavior. Various issues are associated with using natural fibers as reinforcement in polymer composites. As such, more research is required, focusing on parameters like stiffness in composites, moisture effects, fatigue and creep life, physical degradation, and less long-term stability. These composites have outstanding structural qualities, and more research is needed to analyze the performance of natural fiber composite materials in impact loading. Compared to virgin PET, rPET degrades during recycling, resulting in inferior mechanical characteristics. Mixing rPET with virgin PET, polyamide, polycarbonate, and other materials may address the mechanical performance loss of rPET. Combining rPET with additives like chain extender, polypropylene, polyethylene, polyamide, and polycarbonate enhances the blend's properties, reduces environmental waste, and slows the spread of contaminants. Hybridization using several natural (and synthetic) fiber reinforcements has emerged as a viable composite manufacturing avenue. Furthermore, it can enhance mechanical behavior and durability. External and load-bearing applications for NFRCs have significantly grown, but more research is needed to broaden their use range, including improvements in the hydrothermal environment and durability. Future research and development must focus on developing eco-friendly materials with adequate qualities to replace synthetic polymer composites in emerging new application areas. This review discussed natural fibers, their types, surface modification of fibers, physical and mechanical properties, advantages, and disadvantages. Due to their durability, renewable nature, and fully or partially recyclable qualities, research on natural fiber-based composite materials has expanded in recent years. The automobile and space sectors are the most pushing composites, which have been reinforced with natural fiber manufacturing components that are non-structural and semi-structural. Other major industries, such as sports, medicine, furniture, and packaging materials, also adopt natural fiber composites.

CRediT authorship contribution statement

Ajay Kumar: writing – review & editing, writing – original draft; conceptualization and data curation; **Raman Bedi** Sc: supervision and review of entire manuscript.

Conflict of interest

The authors declare that they have no conflict of interest.

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