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Sustainable Fiber-Reinforced Composites: A Review

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Sustainable fiber reinforced polymer (FRP) composites from renewable and biodegradable fibrous materials and polymer matrices are of great interest, as they can potentially reduce environmental impacts. However, the overall properties of such composites are still far from the high-performance conventional glass or carbon FRP composites. Therefore, a balance between composite performance and biodegradability is required with approaches to what one might call an eco-friendly composite. This review provides an overview of sustainable FRP composites, their manufacturing techniques, and sustainability in general at materials, manufacturing, and end-of-life levels. Sustainable plant-based natural fibers and polymer matrices are also summarized, followed by an overview of their modification techniques to obtain high-performance, multifunctional, and sustainable FRP composites. Current state-of-the-art mechanical and functional properties of such composites are then surveyed, and an overview of their potential applications in various industries, including automobile, aerospace, construction, medical, sports, and electronics is provided. Finally, future market trends, current challenges, and the future perspective on sustainable natural FRP composites are discussed.

1. Introduction

Fiber-reinforced polymer (FRP) composites, comprised of a polymer matrix reinforced with fibers, have drawn significant interest in the last two decades as an alternative to traditional monolithic materials due to their higher specific strength and modulus, lightweight, adjustable deformation behavior, and good corrosion resistance. Consequently, they have been used in many applications, for example, in aerospace, automotive, medical, energy, and sports industries.[1–3] To manufacture FRP composites, synthetic fibers (e.g., carbon, glass, aramid) are the most popular choice as reinforcement materials due to their superior physical and mechanical properties compared to other fiber types. Additionally, synthetic polymers have widely been used as polymer matrices because of their higher adhesive properties, chemical resistance, moisture resistance, excellent mechanical properties, good fatigue resistance, low shrinkage, and strong durability at low and high temperatures and high electrical resistance. However, the environmental impacts of synthetic fibers and polymers are widespread and substantial, as they are non-biodegradable and can stay in the environment for hundreds of years.[4,5] Additionally, the production of such materials from fossil fuels releases a significant amount of excess greenhouse gases (GHG), contributing to global warming. Furthermore, fossil fuel depletion, increasing awareness of the environmental crisis, and sustainability, in general, have forced researchers and innovators to shift towards manufacturing sustainable, biodegradable, and recyclable FRP composites.[6] Indeed, the use of renewable and biodegradable fibrous materials and polymer matrices to manufacture FRP composites could considerably reduce the environmental impacts of such composites.

As sustainability is rapidly becoming a global priority across many industries, composite industries are also driving towards manufacturing sustainable composites. The development of such composites with lightweight and high performance at a lower cost is mainly dominated by the utilization of biodegradable and sustainable materials. Composites containing at least one of these components (matrix or reinforcement) obtained from natural resources are categorized as partially bio-degradable composites, and composites with all constituents from natural resources are called fully bio-degradable green composites.[7] Natural fibers, in this context, are proven

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to be proficient materials for producing sustainable composites due to their abundance, lightweight properties, and low cost along with good mechanical properties, high specific strength, non-abrasive nature, eco-friendliness, and biodegradable features.[8,9] Additionally, the environmental impacts of such fibers are significantly lower than that of synthetic fibers.[10] Furthermore, the lower abrasive characteristics of natural fibers compared to glass fibers make their processing easier. Therefore, natural fibers (e.g., jute, flax, kenaf, hemp, and sisal) are becoming increasingly attractive to composite manufacturers[11,12] as a replacement for glass and mineral fillers[13] for producing sustainable composites.

An approach to an environmentally friendly composite could be manufactured via using natural fibers reinforced with biodegradable polymers as a matrix that can be composted at the end of their life cycle. Also, the addition of nanomaterials with either natural fibers or the matrix material or with both the components increases the strength of the composites significantly.[14]

However, the overall properties of such composites are still far from the high-performance conventional glass-fiber reinforced composites. Therefore, a balance between composite performance and biodegradability is required with approaches to what one might call an eco-friendly composite.[15] Many research are currently in progress to improve the durability and strength of bio-composite for effective dynamic applications.[16] Additionally, the composites industry is forecasted to grow significantly with the development of new fibers and polymer matrices. The industry will need to shift towards biodegradable and/or recycled fiber and polymers, with a lowering of the environmental impact to contribute towards global net-zero targets to obtain zero or no excess GHG emissions by 2050 for the sustainable composite industry.

While several reviews have focused on natural fiber reinforced composites,[17–20] there remains a lack of a comprehensive review on the recent progress of sustainable fibers and polymer matrices to produce eco-friendly FRP. This review provides a brief overview of sustainable FRP composites (**Figure 1**) and their manufacturing techniques. We then review materials sustainability for FRP composites, followed by sustainable fibers and polymer matrices for sustainable FRP composites. The mechanical, physical, and thermal properties of sustainable FRP composites and its potential applications are then discussed. Finally, we present our recommendations and perspectives on future research directions. We believe, the review will be helpful to enhance scientific research and upgrade the technological capabilities of composite industries and facilitate reliable and

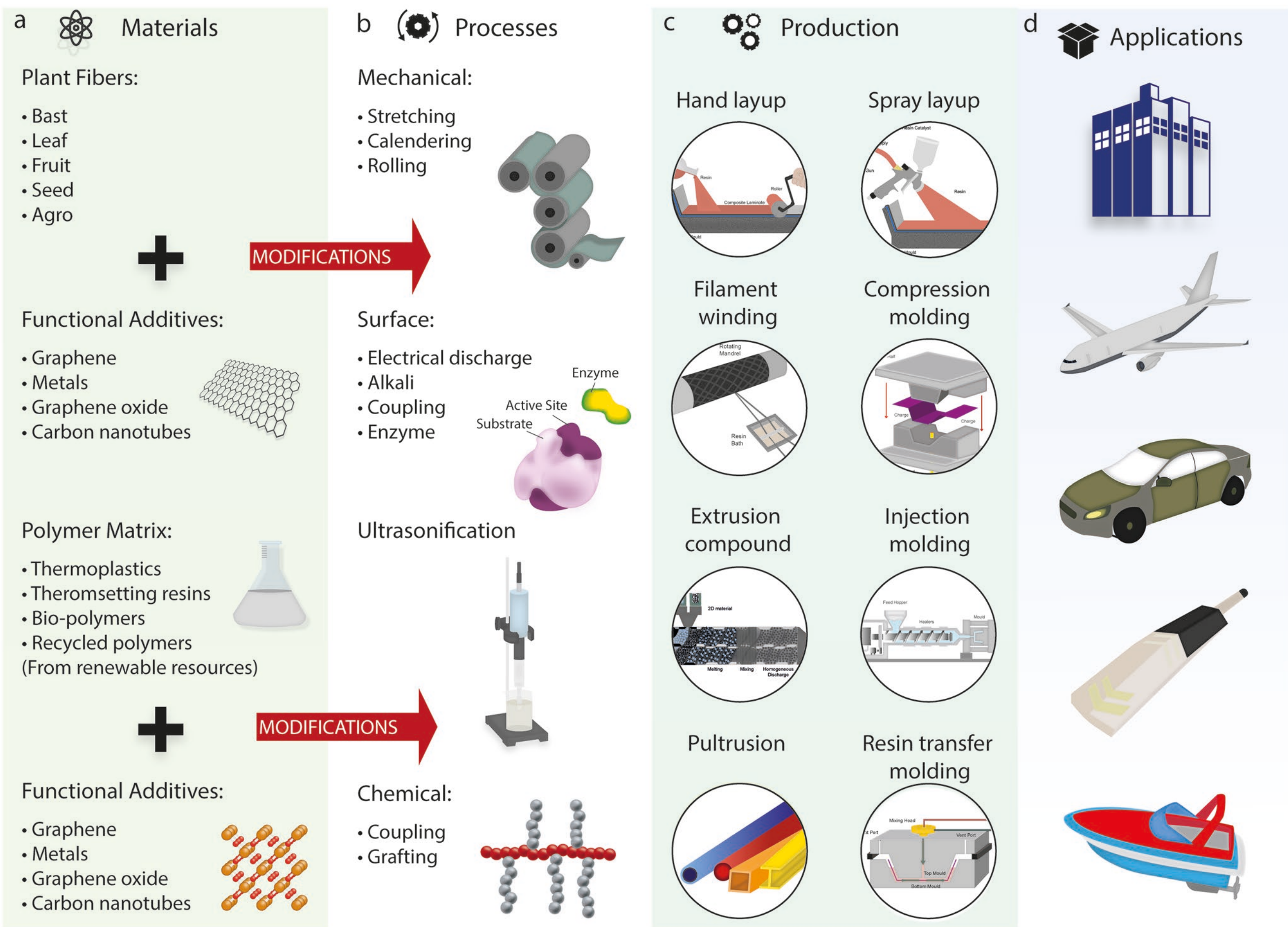

Figure 1. Sustainable FRP composites: a) materials, b) modification processes, c) composite manufacturing methods, and d) applications of fiber reinforced composites.

sustainable infrastructure development (United Nation Sustainable Development Goals (SDG) 9). It will also encourage the efficient use of natural resources, reduce synthetic waste generation and encourage the composite industries to adopt sustainable practices (SDG 12).

2. Fiber Reinforced Polymer (FRP) Composites and Manufacturing Techniques

2.1. Introduction to Composites

Composites are bi-phase or multi-phase materials, composed of two or more different materials, offering significantly improved bulk properties than their constituent materials. These enhanced properties include a high strength to weight ratio, high toughness, excellent dimensional stability, high durability and stiffness, flexural strength, and resistance to corrosion, wear impact, and fire.[21–23] The evidence of combining different materials in buildings and construction works was found in ancient times. The earliest examples of composites in construction were found in 3400 B.C. when the Mesopotamians created plywood by gluing wood strips at different angles.[24,25] The advances in material science have accelerated the development of synthetic materials, as an alternative to natural materials for composites manufacturing. During this phase of development, the invention of polymer resins and glass fibers around the 1930s, and the development of carbon fibers in the 1960s laid the groundwork for the manufacture of modern FRP composites. Additionally, the industry has developed expertise since then in specialized molding processes, allowing manufacturers to tailor several unique properties of composites. **Figure 2** summarizes the chronological development of composites materials and their applications in several industries including infrastructure, transport, and communication from 3400 B.C. to the present day.[24,25]

Composite materials can be classified according to their constituents, that is, base material and reinforcement material. The base material, also termed as a matrix or a binder material, binds or holds the reinforcement materials in composite structures. Depending on the matrix materials, composites may be classified as: organic matrix composites, metal matrix composites, and ceramic matrix composites. Organic matrix composites could further be classified as carbon matrix composites and polymer matrix composites. The reinforcing materials may be present in the form of fibers, particles, platelets, or whiskers of natural or synthetic materials. Based on these reinforcement materials, composites can be classified broadly either as fiber reinforced composites, particle reinforced composites, and laminar or sheet-molded composites.[22,26,27] In this review,

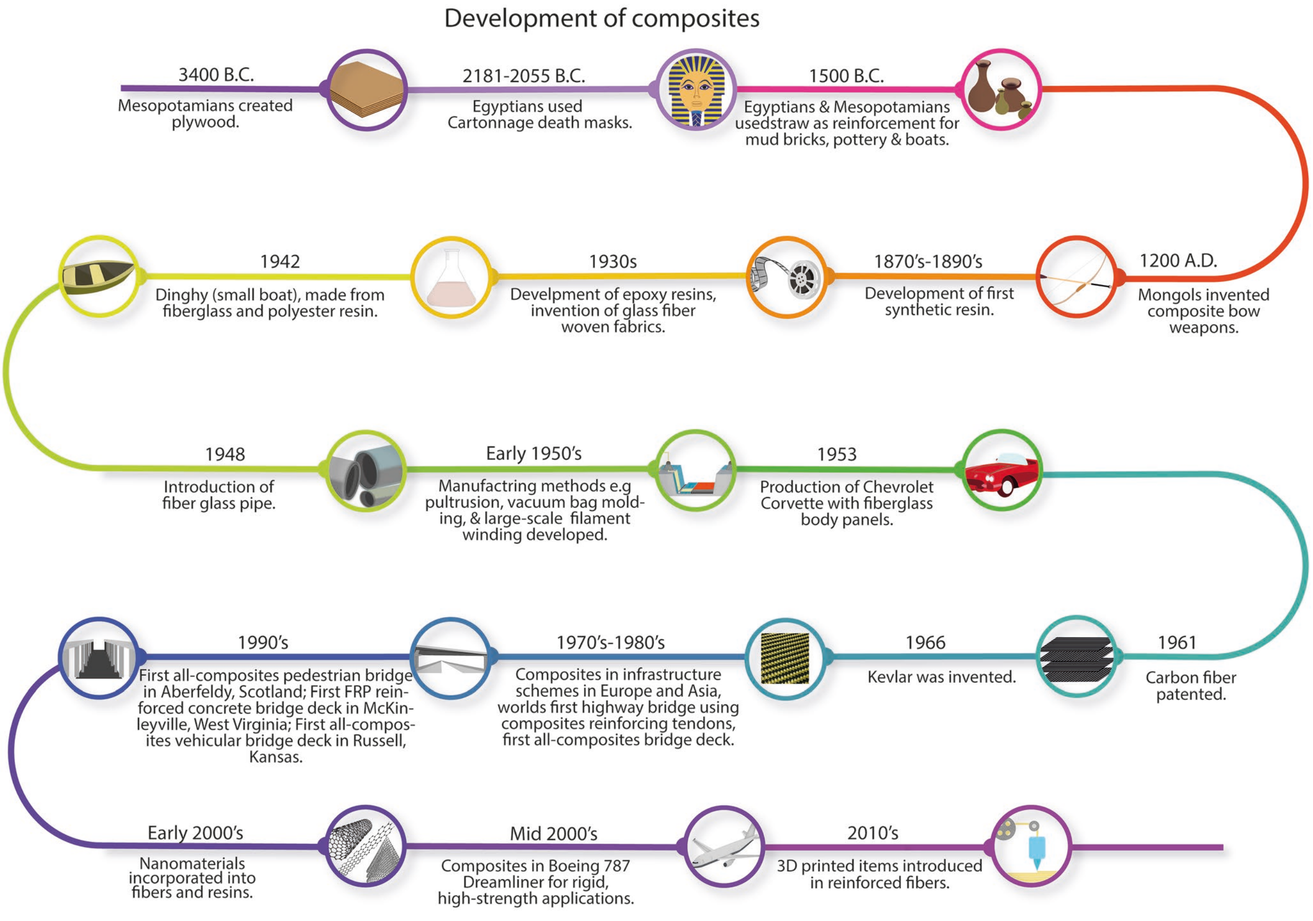

Figure 2. Chronological development of composites manufacturing for several industries.

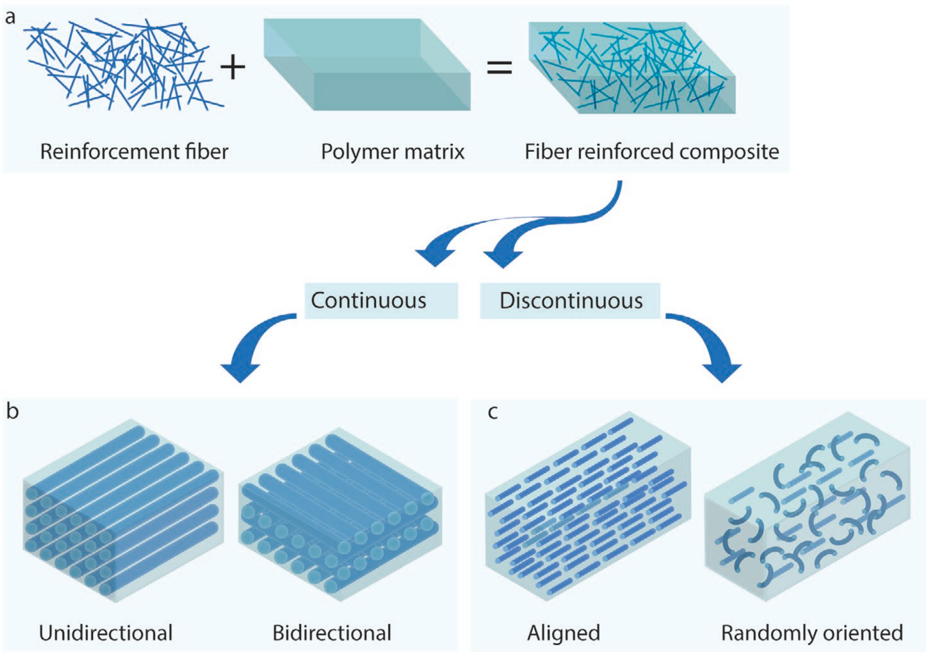

Figure 3. The general structural assembly of FRP composites and their classification: a) Basic constituents of FRP composites, b) continuous FRP composites: unidirectional and bi-directional, and c) discontinuous FRP composites: aligned and randomly oriented composites. Reproduced with permission.[35] Copyright 2022, Wiley-VCH.

we mainly focus on sustainable FRP composites, due to their intriguing properties.

2.2. Fiber Reinforced Polymer Composites

FRP composites comprise a polymer matrix reinforced with fibers (**Figure 3**a).[28] The fibers may either be natural or synthetic, and the most common polymer matrices used are thermosetting plastics (e.g., epoxy, vinyl ester, polyester) and phenol formaldehyde resins. FRP composites are used for lightweight structural applications because of their superior mechanical properties and lightweight nature.[29] These composites exhibit superior material properties, which in most cases the traditional engineering materials (e.g., metals) cannot provide at low weight. These can be manufactured to any geometry to obtain maximum efficiency in terms of the utilization of material strength.[30] Since lightweight design is important in various industries, including aerospace, wind energy, and automotive applications, FRP composites are attracting significant interest for such weight-sensitive applications due to their excellent stiffness and strength combined with a low density.[31] Additionally, FRP composites offer several other advantages, including that they are typically electrochemically non-corrosive and have tunable properties for specific applications.[32] Natural/plant fiber composites, another significant branch of FRP composites, have experienced a rapid expansion over the last few decades due to the advantages offered by plant fibers. These advantages include abundance, environmental friendliness, biodegradability, nontoxicity, low cost and density, flexibility during processing, and high tensile and flexural modulus.[33] Furthermore, they provide better flexural and impact strength, higher moisture resistance, less shrinkage, and improved weatherability.[34] Therefore such composites offer greater efficiency based on their life cycle assessment (LCA) compared to traditional engineering materials, and represent a promising alternative for future sustainable industries.[30]

FRP composites can be classified according to the length of the constituent fibers.[35] Composites with long fiber reinforcements are termed as continuous FRP composites (Figure 3b), while composites with short fiber reinforcements are termed as discontinuous FRP composites (Figure 3c). The placement of fibers can be unidirectional or bidirectional in the case of continuous (Figure 3b), and aligned or random for discontinuous (Figure 3c) FRP composites. When more than one fiber is used as reinforcement in a single polymer matrix, it is then termed as hybrid FRP composite. The type, length, arrangements, and orientation of fibers define the properties and structural behavior of the final composites.[35]

2.3. Manufacturing of FRP Composites

For manufacturing FRP composites, a fiber preform is prepared first by weaving, knitting, braiding, or stitching fibers in long sheets or matt structures, followed by reinforcement with a polymer matrix. Prepregs, composed of fiber materials pre-impregnated with a thermoplastic or thermoset polymer matrix, are sometimes used, which need a certain temperature to get cured and formed into a desired composites shape. Depending on the curing conditions, the composite manufacturing techniques can be categorized either as open or closed molding. In open molding, reinforcement materials and resins are exposed to air as they cure or harden. The most common open molding processes for manufacturing FRP composites are hand lay-up,[36] spray-up,[37] and filament winding.[38] In closed-molding, raw materials, that is, reinforcement materials and resins get cured inside a two-sided mold or within a vacuum bag. The closed molding process requires special equipment and is mainly used in large plants that produce large volumes of materials. Compression molding,[39,40] extrusion compounding,[41] injection molding,[42] pultrusion,[43] resin transfer molding (RTM),[44] and vacuum assisted resin transfer molding (VARTM)[45] are closed molding processes used for the manufacture of FRP composites.

Hand layup is the most widely used technique for manufacturing open mold composites. It involves manually laying down the individual layers of reinforcement materials and pouring the liquid resin over them. A roller is usually used to force down the resin to consolidate the laminate, thoroughly wetting various reinforcement layers and removing the entrapped air (**Figure 4**a). The relatively low start-up cost allows the manufacturing of a wide range of products with various geometries, new parts, and designs. However, there are several disadvantages of this method, including lower production rate and lower reinforcement volume fraction, nonuniform distribution of reinforcement materials and matrices due to lack of precision in human hand, which make the process inappropriate for large-scale manufacturing of high-quality composites.[46–49]

Spray layup is another open molding technique where a handgun is used to spray the resin instead of pouring it over the laid-up fibers. The use of a roller can assist simultaneously by fusing the fibers into the matrix material and enabling the removal of bubbles and voids before curing (Figure 4b). The operator controls thickness and consistency, therefore such a process is more dependent on the operator than the hand lay-up itself. The fiber orientation and the fiber constraints determine the mechanical properties of the final product.[47,50] Filament winding is another open molding technique that is useful when manufacturing axisymmetric components such as pipes, tubes, tanks, vessels, driveshaft, missiles, and pressure vessels. This technique offers certain advantages over other manufacturing approaches, such as producing components with higher fiber volume fractions (60–80%) and higher specific strengths.[51] In this technique, resin-impregnated continuous fiber is wound on a rotating mandrel with a certain winding angle, controlled by the mandrel speed and that of the carriage guide. After winding, the resin is solidified via curing for thermosetting resins, followed by removal of the mandrel,[52] (Figure 4c).

Compression molding is a very popular manufacturing technique among closed molding techniques, due to the precise and rapid production of quality composite parts at a high volume.[53] In such a technique, prepregs are placed into an open heated mold cavity, then closed with a top plug, and compressed with a large hydraulic press to ensure the uniform distribution of materials all over the mold[54] (Figure 4d). The combination

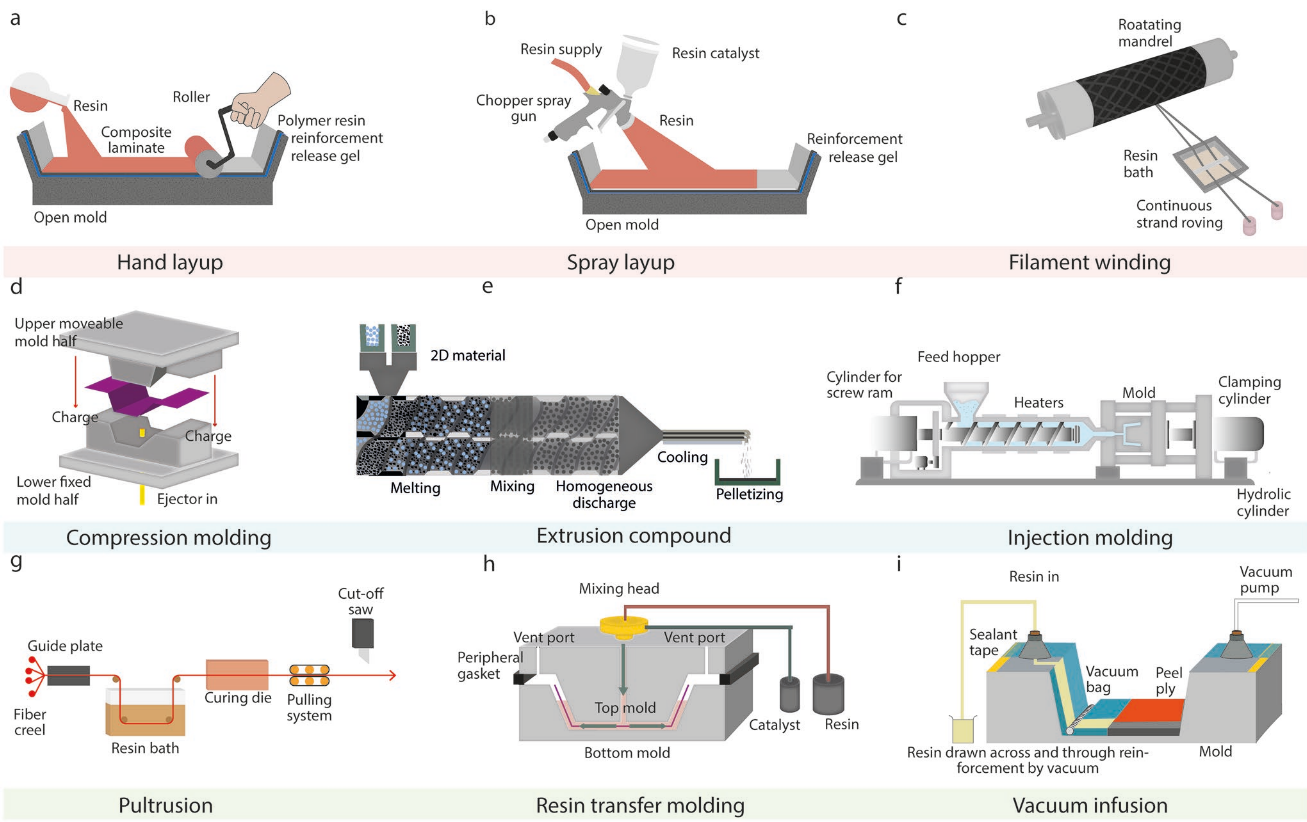

Figure 4. Schematics of common composite manufacturing techniques, a) hand layup, b) spray layup, c) filament winding, d) compression molding, e) extrusion compound, f) injection molding, g) pultrusion, h) RTM, and i) vacuum infusion or vacuum assisted resin transfer.

of two manufacturing techniques has also been reported: for example, the use of hand lay-up followed by a compression molding technique.[55]

Extrusion compounding, another closed molding technique, is a continuous process of manufacturing composites with constant cross-sections including rods, sheets, pipes, and films, and produced by forcing softened polymer through a 2D die opening. Fiber and polymer matrix mixtures are fed into the extruder through a hopper, conveyed forward by a feeding screw, and forced through a die, converting them to a fiber-reinforced polymer product (Figure 4e). Heating elements placed over the barrel soften and melt the polymer. Both stiff and soft materials can be formed into any shape with a smooth surface finish. With a similar principle, in injection molding, after metering the required amount of material into the barrel, the screw injects the material into the mold through a nozzle where the material gets cooled and achieves its final shape[56] (Figure 4f). Injection molding can fabricate complex 2D composite parts of various shapes and sizes with high precision at a very low cycle time.[47,57]

Pultrusion is another continuous process of manufacturing FRP composites with constant cross-sections. The term pultrusion comes from combining the words, "extrusion" and "pull". Unlike extrusion, where profiles are manufactured by pushing material through a die, the material is pulled instead. Layers of fibrous materials are impregnated with resin, followed by drawing through a stationary, temperature-controlled die that polymerizes the resin. A continuous profile is pulled from the production line using a pull-off unit (Figure 4g) and cut off at the required length.[58,59] Pultrusion offers manufacturing of very lightweight but high-strength composites with great uniformity and extremely low manufacturing defects in products. **Figure 5** represents a comparative schematic of the tensile strength versus fiber volume fraction for several composite manufacturing methods.[60]

RTM is another closed-mold process, which can manufacture high-performance composite components in medium volumes (1000s to 10 000s of parts). The basic principle involves the laying of dry reinforcement materials inside a closed mold cavity, adjusting the mold to a predetermined cavity height followed by the injection of a low viscosity resin (typically 0.1 to 1 Pas^{-1}) through a channel to the laid reinforcement material under moderate pressure (usually 3.5–7 bar; Figure 4h). High pressure is avoided due to the risk of displacement and disorientation of the reinforcement material inside the mold cavity.[61,62] Vacuum infusion or VARTM is an improved version of the RTM process. The preform reinforcement materials are sealed in a vacuum bag and a perforated tube is placed between the vacuum bag and the resin container. The vacuum force draws the resin through the perforated tubes over the reinforcement materials leaving no room for excess air, thereby consolidating the laminate structure (Figure 4i). The high-quality products obtained due to the reduction in void content make the VARTM process popular for large objects.[47,63,64]

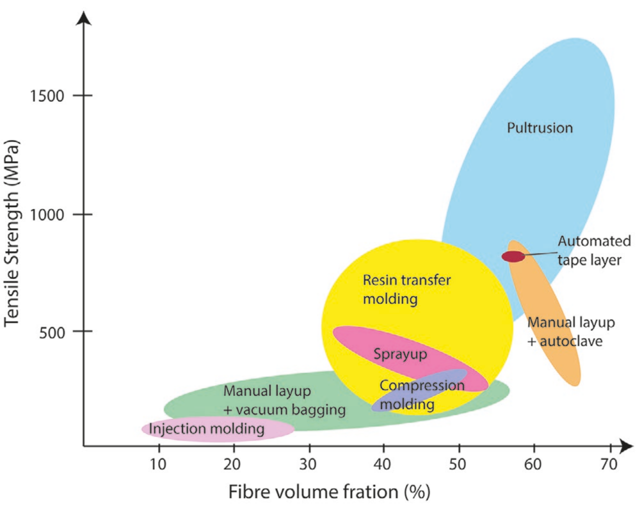

Figure 5. A comparison of tensile strength versus fiber volume fraction for several composite manufacturing techniques. Reproduced with permission.[60] Copyright 2018, Wiley-VCH.

Several automated techniques are emerging to manufacture composites that can replace the human operator; they are advanced in process control and repeatable with reduced manufacturing times. For example, robotic filament winding (RFW) technology, uses an industrial robot to place fibers impregnated with resin along the required direction.[65] Automated tape lay-up (ATL) is an automated process for laying up prepreg tapes, which can save more than 70–85% of the time and cost spent on manual lay-up. This process is used in the manufacturing of carbon–epoxy prepreg components for both military and commercial aircraft, such as the wing skins of F-22 Raptor, tilt-rotor wing skins of V-22 Osprey, and the skins of wing and stabilizer of the B-1 Lancer and B-2 Spirit bombers, empennage parts (e.g., spars, ribs, I-beam stiffeners) for the B777, A340-500/600, A380 airliners, etc. Automated fiber placement (AFP) is another automated production method for large aircraft structures from prepreg.[66] Such automated processes can produce composites with comparable strength to traditional techniques, making the processes beneficial in speeding up the production and cutting labor costs. However, their use is limited due to the cost of specialized machinery and constraints in the fabrication of complex parts.[67]

2.4. Outlook on Manufacturing

The manufacturing of composites has always been an issue in terms of large-scale production. A common realization within the composites industry is that a true breakthrough in the use of composite materials will not be realized solely via a reduction in raw material costs but, more importantly, by reduced manufacturing costs.[68] However, the present trend for reducing the manufacturing cost is focusing on more automation during FRP composites manufacturing. It is also worth noting that, challenges not only exist with the technical aspect but also the lack of knowledge and expertise about novel and sustainable composites manufacturing among the designer and manufacturers of FRP composites. Additionally, developing countries are aiming for fast industrialization, improved exports, self-reliability, and lessening the import for a better economy. Therefore, there remains a need to exploit locally available resources, and invest in local research and development for composites manufacturing in the future.

3. Sustainability and Sustainability Index for FRP Composites

3.1. Sustainability

Sustainability, and more precisely "sustainable development", has developed an increased focus in the last four decades, which arose from the environmental movement in the late 1960s and 1970s. It became a focal point in 1987 with the landmark report entitled "Our Common Future", where it was defined as "Sustainable development is the development that meets the needs of the present without compromising the ability of future generations to meet their own needs".[69] This Brundtland definition created a global explosion of academic debate and policy issues, leading to the United Nations Conference on Environment and Development (UNCED) in Rio in 1992.[70] The concept of sustainability expanded to a balance between economic, social, and environmental factors to meet future demands of resources.[71] As such, the sustainability parameters for the environment include the use of water, energy, and chemicals, emission of GHGs, and generation of wastewater, waste materials, and chemicals throughout the supply chain. Nowadays, sustainability is measured using LCA in various forms such as cradle to cradle, cradle to grave, and cradle to gate, which have helped to understand environmental impacts more holistically.[72,73]

3.2. Sustainability for FRP Composites

There are more than 40 000 industrial composite products in various sectors such as energy, transportation, medical, construction, etc.[73,74] The use of composites is increasing every day, particularly for industrial applications.[75–77] For example, in airplanes, approximately 38% of the major components are now made from composite materials.[78] As a result, the environmental impact of the composites has become a reality along with the other sectors, where increasing environmental awareness, growing waste management issues, and rising crude oil prices have created great concerns from both legislative and consumer perspectives.[79,80] For FRP composites, natural plant fibers have been used as bio-composites in automotive, interiors, construction, transport, insulation material, etc., in combination with other synthetic fibers.[80–82] The carbon footprint of such fibers is in the range 0.3 to 0.5 tons CO_2eq, which is significantly lower in comparison to that of glass fibers (≈1.7 to 2.5 tons CO_2eq) during the fiber production stage. However, the footprint is higher for natural fibers than for glass during composite manufacturing, which somewhat offsets the advantages.[83] The comparison of LCA of bio-composites and glass FRP composites can be reviewed in this context (**Figure 6**),[80] which reveals—a) natural fiber composites have a lower environmental impact than glass-based materials; b) an increase of natural fibers could reduce the base fiber content; c) the final product could have lighter weight adding fuel efficiency; d) recycling could be extended for biobased parts in other applications. Environmental sustainability in the composites sector can therefore be divided into three categories: a) materials used, b) manufacturing process of composites, c) end-of-life and recycling.[81]

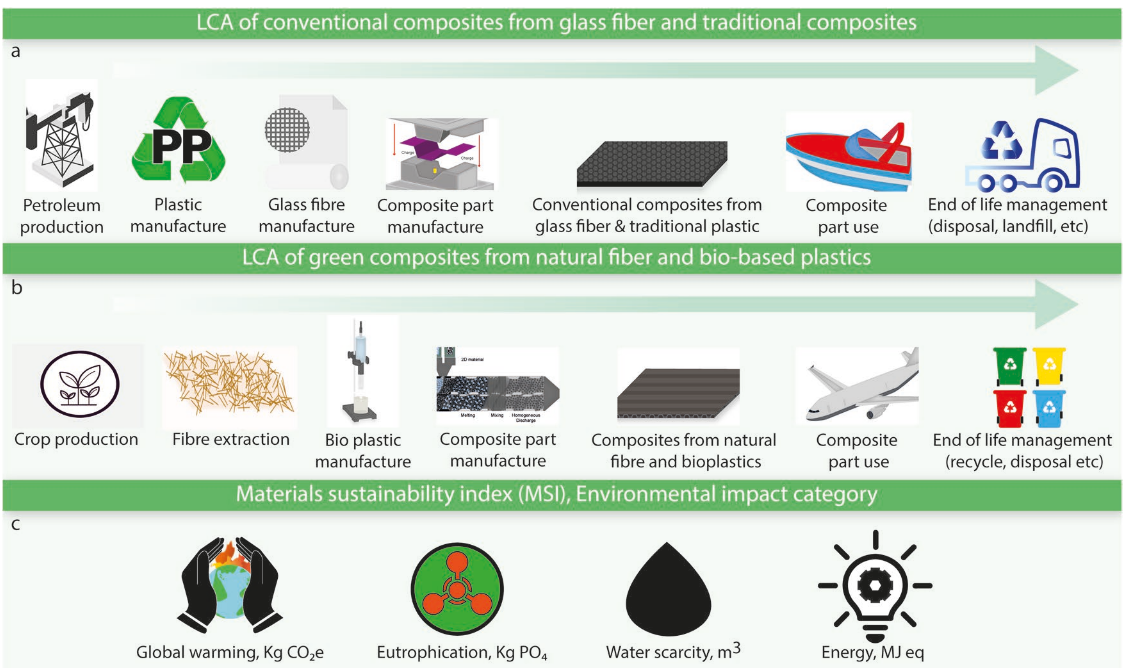

Figure 6. Simplified LCA study approach for a) conventional composites from glass fibers and traditional plastics, b) green composites from natural fibers and bio-based plastics, and c) environmental impact category for materials sustainability index (MSI).

Table 1. Carbon footprints of FRP composite components: Natural and synthetic fibers and Conventional epoxy resin polymer.

Composite components	Energy intensity [MJ kg^{-1}]	Ref	CO_2 emission [kg CO_{2eq}/Kg of fiber]	Ref
Glass fiber	13–32	[93,94]	1.4–2.95	[91,92,95]
Carbon fiber	183–286	[96]	29.4	[84,97]
Recycled carbon fiber	<250	[97]	4.65	[98]
Jute fiber	9.6	[84]	0.97	[84]
Flax fiber	6.5	[99]	0.90	[83]
Epoxy resin	76–80	[96]	6.70	[100]

3.2.1. Material Used

Glass and carbon fibers dominate composite applications due to the demand for high-performance and lightweight materials.[84,85] Natural fiber-based composites are limited in use as their mechanical, thermal, electrical, and chemical properties for particular applications are often less than glass and carbon fibers.[86] However, the biodegradability of plastics and synthetic fibers is of great concern nowadays. As a result, the demand for natural fiber-based composites is rising.[84] Different polymers, mostly synthetic, are used to produce composites for various applications, and are typically non-degradable such as epoxy, unsaturated polyester, vinyl ester. Epoxy, the most widely used matrix material, has many other uses, including as adhesives, coatings, and casting materials. These resins are derived from petroleum-based sources.[86] To improve their performance, several modifiers such as rubbers and elastomers are also used. It is possible to use bio-resins such as tannin, lignin, furan, rosin, and bio-modifiers in a ratio to the synthetic resin to reduce their environmental impacts.[87,88]

Since sustainability of any composite material depends on the input resources and biodegradability, a comparative index, known as Higg material sustainable index (MSI), was developed by the Sustainable Apparel Coalition (SAC). It is based on a cradle-to-gate material impact assessment, which assists in identifying and comparing materials and processes with the highest/lowest impact.[89] MSI assesses five environmental impacts: global warming potential (GWP), nutrient pollution in water (eutrophication), water scarcity, fossil fuel depletion, and chemistry, and features 80+ example materials, including natural and synthetic fibers, leather, and metals. In addition, material traceability is also getting more focus as it can improve the value during the end-of-life stages.[90]

Jute and flax are increasingly used in technical applications; their environmental impact is extensively studied compared to synthetic counterparts. Jute and flax production are usually water-intensive, mainly required during land preparation, plant growth, and the retting process compared to the corresponding glass and carbon fiber.[84] In addition, the production of natural fiber also requires fertilizers and pesticides to some extent—the impact of this can be countered by using organic fertilizers such as meat slurry. However, the carbon footprint of these natural fibers is much lower than that of glass and carbon fibers, **Table 1**. The production of 1 kg of glass fiber emits between 1.4 to 2.95 kg of CO_{2eq},[91,92] whereas their natural fiber counterpart has a carbon footprint of ≈ 0.9 CO_{2eq}.

3.2.2. Manufacturing Process of Composites

The environmental impacts in the processing stages are dominated by the use of energy, along with GHG emissions.[73] The European Composites Industry Association (EuCIA) has created an online tool, "Eco Impact Calculator for composites", to measure the impact of the production of composites, which allows companies to quantify the manufacturing part of the life cycle.[101] Although the composite manufacturing process adds carbon emissions in the production of natural fiber-based composite; it is still 50% lower than glass fiber-based composites. Even virgin carbon fibers release a significant amount of GHG during production, with a total value of 29.45 kg CO_2eq, which is approximately 60 000 times higher than natural fibers. A recycled carbon fiber-based composite has significantly low GHG emission during production which is five times higher than jute and flax fiber,[98] and up to half that of hemp-based biocomposites.[102] In addition, natural fiber can act as a carbon sink- during its growth, although, the embedded carbon will be released back into the atmosphere at the end of life.[83] Water use is a concern for jute and flax; these biodegradable fibers can enhance soil quality at the end of their life. Therefore, replacing synthetic fibers with natural plant-based biocomposites could pave the way for sustainable composite applications. On the other hand, epoxy resin, which is very popular for composites applications, has several disadvantages in terms of GWP and biodegradability as this resin is very difficult to recycle.[103,104] Different alternatives have been researched, such as epoxidized sucrose soyate resin (ESS) (0.287 kg CO_{2eq}/kg of ESS),[105] vegetable oil-based resin (5.7 kg CO_{2eq}/kg of resin),[106] supersap bioresin (5.7 kg CO_{2eq}/kg of resin),[107] or bio-based ECH (epichlorohydrin) resins with ≈61% reduction of the GWP.[108]

3.2.3. End-of-Life and Recycling

Sustainability is becoming a focal point in the material selection process, particularly in the use phase.[73] Additionally, there are other specific concerns for particular types

Figure 7. Criteria for sustainable fibers.

of applications; for example, fluoride emissions during the manufacturing of traditional glass-composite materials are significantly hazardous to the environment.[77] Waste disposal and waste management are key aspects of the sustainability of composites. FRP composites have different end life compared to traditional textiles. First, because the composites are made of different parts, which means the end-of-life for the product does not mean the end-of-life for the parts.[73] Second, there are limited options for the reuse and recycling of composites due to the poor performance properties of reused or recycled material. As a result, only 15% of the composites produced in the UK are reused or recycled each year at their end-of-life.[109] Moreover, recycling is not commercially viable in some cases, particularly for glass fiber composites in wind-turbine applications.[110] Innovation would be key in the area of recycling and end-of-life management for better material circularity.[110]

4. Sustainable Fibers

There have been several efforts across the world to grow fibers that are sustainable, biodegradable, and lightweight for structural composite applications, and could be manufactured at high volume and low cost. Sustainable fibers, commonly known as "Eco-friendly" fibers, have an insignificant impact on the environment during their production processes, and meet at least half of the criteria as illustrated in **Figure 7** including low water and energy consumption, being made from waste materials and used renewable resources, have control on chemical consumption, and soil erosion. Such fibers are biodegradable, and many of them often originate from bio-based resources. The growing interest towards the use of sustainable fibers from renewable and biodegradable sources to produce bio-based and high-potential green products have driven many researchers to investigate the possible use of natural fibers as reinforcement materials for green and potentially sustainable bio-composites.[111]

Natural fibers are categorized mainly based on their origin from plants, animals, or minerals. Some natural fibers are produced from the different sections of plants and trees. Plant-based cellulosic fibers can be classified primarily into seed fibers (cotton, coir, and kapok), bast fibers (jute, flax, ramie, hemp, and kenaf), and leaf fibers (sisal, pineapple, banana, abaca), **Figure 8**. Natural plant fibers are entirely derived from vegetable sources, therefore completely biodegradable and sustainable. The major chemical components of natural plant fibers are cellulose, hemi-cellulose, lignin, pectin, and wax.[112–115] The presence of predominantly hydrophilic cellulose affects the interfacial bonding between the fibers and a polymer matrix because, typically, these matrices are hydrophobic. Therefore, chemical treatment of such fibers is one of the ways to optimize the bonding between the fibers and polymer matrix. Such treatments modify −OH functional groups present on the surface of the fibers, and increase the surface roughness, thereby enhancing the interfacial interaction between the fibers and a matrix.[116–120]

The properties of composites significantly depend on the properties of reinforcement materials, that is, the natural fibers,[121–124] **Table 2**. Therefore, the selection of such materials during the design and manufacturing of sustainable composites plays a crucial role in determining their performance. Various natural fibers have been used or have the potential to be used

Sustainable Fibers

a) Bast fibers

Fiber	Cellulose, %	Lignin, %	Y Modulus, GPa
Flax	~70	~2.2	25-80
Jute	60-70	12-14	10-30
Ramie	68-75	0.6-0.7	44-128

b) Fruit fibers

Fiber	Cellulose, %	Lignin, %	Y Modulus, GPa
Coir	32-43	40-45	4-6
Kapok	~43	13-15	~4
Palm	~65	~29	~1.7

c) Leaf fibers

Fiber	Cellulose, %	Lignin, %	Y Modulus, GPa
Sisal	~65	~10	9.38
Pineapple	~80	~12.7	~82
Banana	~61.5	~15	3.5-32

d) Agro wastes

Fiber	Cellulose, %	Lignin, %	Y Modulus, GPa
Bagasse	25-45	15-25	20-27
Corn	35-40	7-18.5	2.38-4.5
Rice	41-57	8-20	~2.5

Figure 8. Chemical compositions of several sustainable natural fibers: a) bast fibers, b) fruit fibers, c) leaf fibers, and d) agro waste fibers.[126]

Table 2. Mechanical properties of different natural fibers compared to conventional reinforcing fibers.[19,27,121,125,136–139]

Fiber		Density [g cm^{-3}]	Tensile Strength [MPa]	Young's Modulus [GPa]	Elongation at break [%]	Equilibrium moisture content [%]
Eco-friendly sustainable natural fibers	Cotton	1.5–1.6	287–597	5.5–12.6	7–8	–
	Jute	1.44	393–773	10–30	1.5–1.8	12
	Flax	1.54	345–2000	27–85	1–4	7
	Hemp	1.47	368–800	17–70	1.6	9
	Kenaf	1.2	240–930	14–53	1.6	–
	Ramie	1.5–1.56	400–1000	27–128	1–4	9
	Coir	1.25	220	6	15–25	10
	Abaca	1.5	980	–	–	15
	Oil palm (empty fruit bunch)	0.7–1.55	248	3.2	2.5	–
	Sugar palm	1.5	421.4	10.4	9.8	–
	Baggase	1.2	20–290	19.7–27.1	1.1	8.8
	Sisal	1.33	600–700	38	2–3	11
	Pineapple	1.5	170–1627	82	1–3	13
	Banana	1.35	355	33.8	5.3	–
	Bamboo	0.6–1.1	140–230	11–17	–	8.9
	Henequen	1.4	500	13.2	4.8	–
	Nettle	1.51	650	38	1.7	–
Conventional fibers	E-glass	2.5	2000–3500	70–77	4.5–4.9	–
	S-glass	2.5	4570	86–90	4.5–4.9	–
	Carbon	1.8–1.9	3400–5400	230–440	1.4–1.8	–
	Aramid	1.45	3400–4000	130–185	2.5	–

for sustainable FRP composites, which have been discussed here.

4.1. Bast Fibers

Bast fibers are potential sustainable fibers produced from the outer layers of plant stems. They are mainly composed of cellulose, hemi-cellulose, and various proportions of lignin. The most valuable fiber crops that have attracted interest as reinforcements for composites are jute, flax, ramie, hemp, and kenaf.[125]

4.1.1. Jute (Corchorus Capsularis/Olitorius)

Popularly known as "Golden Fiber", Jute is extracted from the bark of the white jute plant (*Corchorus capsularis*), and is a 100% biodegradable, recyclable, and environmentally friendly natural fiber. After cotton, jute is the second most produced natural fiber globally, mainly in developing countries including Bangladesh, India, and China. Jute is a lignocellulosic fiber and is at least ≈50% cheaper than flax and other similar natural fibers. Jute plants can grow up to 15–20 cm in four months, and the fibers are extracted post-harvesting. Such fibers are characterized by high aspect ratio (i.e., length to diameter ratio, l/d), high strength to weight ratio, and have good insulation properties.[127] Jute has widely been used in the manufacturing of flexible packaging materials, carpet backing, geo-textiles, and green composites.

4.1.2. Flax (Linum Usitatissimum)

The cloth made from flax fibers is popularly known as "Linen" in the textile industry. Flax fibers are also cellulosic fibers but have higher crystallinity than jute fibers. The technical fibers extracted from the plant can be very long (up to 90 cm) with a diameter of 12–16 µm. France, Belgium, and Netherlands are the leading manufacturers of flax fibers.[125] They have higher specific tensile properties than glass fibers in addition to low density, higher strength, and stiffness. The higher strength makes them attractive for composite applications. There have been extensive studies investigating flax as reinforcing materials in the form of non-woven mats and combined with a natural resin to develop sustainable composites for the future.[125,128,129]

4.1.3. Ramie (Boehmeria Nivea)

Ramie is a strong, lustrous, soft, and fine bast fiber, extracted from the inner bark of the ramie plant. It is one of the oldest vegetable fibers, and was used to wrap mummies in Egypt.[130] China is the pioneer of ramie fiber production. The challenge with this fiber processing is removing its gum content which is about 30% of the total fiber weight, making it very difficult to be spun. This extracted gum can sometimes be used as a natural resin to develop a green particle board or other biocomposites. Ramie is often used in the manufacturing of fishing nets, ropes, tents, household furnishings, and composites.[131]

4.1.4. Hemp (Cannabis Sativa)

Hemp, most widely grown in Asia and Europe, can grow up to ≈1.2–4.5 m with plant stems of ≈2 cm in diameter.[132,133] A core usually covers the inner girth, and the outer layer is the bast fiber attached to the inner layer via a glue-like substance called pectin. The fibers have excellent mechanical strength and Young's modulus as well as good insulation properties.[134] Such fibers are generally used in ropes and mulching, and have found applications as reinforcement for composites.[135]

4.1.5. Kenaf (Hibiscus Cannabinus)

Kenaf is a strong, stiff, and tough bast fiber, with high resistance to insects. It is mainly extracted from the flowers of the plant with an outer fiber and an inner core. The outer fiber is known as the bast, which makes 40% dry weight of the stalks, and the inner core covers the remainder. Kenaf fibers have low density and high specific mechanical properties[127] and are completely biodegradable. It has traditionally been used for making cords, ropes, and storage bags. Nevertheless, it has found recent applications as composites reinforcements for automobiles, construction, furniture, and packaging applications.[140,141]

4.2. Leaf Fibers

Using lignocellulosic leaf fibers (sisal, pineapple, and banana) as reinforcements in thermoplastic and thermosetting resins to develop low-cost and lightweight composites is an emerging field of research. Such fibers have several advantages: low density, low cost, non-abrasive nature, low energy consumption, high specific properties, and biodegradability.[125]

4.2.1. Sisal (Agave Sisalana)

Sisal, originating from Mexico, is one of the most widely used leaf fibers and consists of the rosette of leaves with a height of up to 1.5–2 m. Sisal fiber is easily cultivated with short renewal times. The fiber has high tenacity and tensile intensity, abrasion, saltwater, acid, and alkali resistance.[127] It was originally used to make ropes and twines. Recently, it has seen applications as a reinforcement for composite materials, and furniture.[142,143]

4.2.2. Pineapple Leaf Fiber (PALF-Ananas Comosus)

Pineapple leaf fiber is a crop waste after fruit cultivation and is white, creamy, and lustrous like silk, and ten times coarser than cotton fiber. It is a multi-cellular lignocellulosic fiber. It shows excellent mechanical, physical, and thermal properties.[144] Some of the major applications for such fibers are in automobiles, mats, construction, and advanced composite materials.[145]

4.2.3. Banana Fiber (Musa Acuminata)

Banana fiber, also known as "Musa fiber" is one of the world's strongest natural fibers.[146] It is biodegradable and extracted from the pseudostem of the banana tree, which is incredibly durable. It consists of thick-walled cell tissue bonded together by natural gums and is primarily composed of cellulose, hemicellulose, and lignin. It is often assumed that banana fibers have similar properties to natural bamboo fiber; however their fineness, spin ability, and tensile strength are much better than that of bamboo fibers.[147] Banana fibers can produce a few different types of textiles with various thicknesses and weights depending on what part of the banana stem the fiber has been extracted from. Banana fibers can be used to make ropes, mats, and woven fabrics as well as handmade papers. These fibers have been used as reinforcement materials for manufacturing green composites.[131]

4.2.4. Bamboo Fiber (Bambusoideae)

Bamboo fibers are also known as natural glass fibers due to their fiber alignment in the longitudinal direction.[148,149] Such fibers are extracted from natural bamboo trees via different physical and mechanical methods. Bamboo fibers have received interest due to their high aspect ratios as well as high strength-to-weight ratios.[150] Additionally, the surface of such fibers are round and smooth. They are lighter, stiffer, and stronger than glass fibers, which make them attractive reinforcement materials for making advanced composite materials for various industries.[151–153]

4.3. Fruit Fibers

The fruits and seeds of plants are often attached to hairs or fibers or encased in a husk that may be fibrous. Such fibers are an attractive combination of cellulose and lignin. They are widely known for their durability and thermal insulation properties and possess high potential as reinforcements for composite applications.[115]

4.3.1. Coir (Cocos Nucifera)

Coir is a short and coarse fiber extracted from the outer shell of coconuts. The thickness of coir fiber is very high as compared to that of other natural fibers and shows very good chemical resistance. The slow decomposition rate of coir fibers makes them suitable for making durable geo-textiles. There are two types of coir: coarser brown coir and finer white coir.[154] More commonly used coir is brown coir obtained from mature coconuts. Coir fibers are mostly used for making rugs, mattresses, doormats, building boards, insulation boards, cement boards, building panels, and composites.[125]

4.3.2. Kapok (Ceiba Pentandra)

Kapok fibers are obtained from the seed hairs of kapok trees. They are cotton-like fibers with colors varying from yellowish to light brown. They are extremely lightweight and hydrophobic.[155] Conventionally, kapok fibers are used as buoyancy materials, oil absorbents, biofuels, insulation materials for heat and sound, and reinforcement materials for composites.[156–158]

4.3.3. Palm (Phoenix Dactylifera)

Palm fibers are usually obtained from the fruits, rachis, and leaves of the date palm trees. Such fibers can potentially be used as sources of cellulose and lignin. They can be used as reinforcements for thermoplastic and thermosetting polymer-based composites in the automotive sector.[159]

4.4. Agro Waste Fibers

The primary fiber wastes produced from agricultural activities are a rich source of cellulose. They have potential as reinforcing materials due to their wide availability and benefits of being renewable, degradable, and economical. Such biomass wastes provide several excellent specific properties especially mechanical, thermal, and biodegradability, making them suitable as reinforcements for FRP composite applications.[160]

4.4.1. Sugarcane Fiber (Bagasse)

Sugarcane bagasse is a fibrous material obtained as a residue from sugarcane after crushing to extract the juice. It is mainly composed of two components, that is, the outer rind and inner pith. The rind consists of a strong fibrous structure, which protects the inner components of a soft spongy structure. It contains long, fine fibers that are randomly arranged throughout the stem and bonded together via hemi-cellulose and lignin.[161] The inner component contains smaller fibers, mainly composed of cellulose. Sugarcane bagasse approximately comprises of 50% cellulose, 25% hemicellulose, and 25% lignin. It has widely been used as a composite reinforcement due to its high cellulose content which exhibits a highly crystalline structure.[162] Cement composites, false ceilings, particle boards, and lightweight structures are some of the applications using bagasse as one of the reinforcement materials.[163]

4.4.2. Corn Stalk

Corn stalks are composed of a pith and rind. The pith of corn stalks is rich in hemicellulose, fat, protein, and sugar.[164] However, the primary components of corn stalk rind are cellulose and lignin, thereby resulting in high strength and toughness. Because of the weakly bonded pith, corn stalks can exhibit poor strength. So the pith needs to be removed when preparing corn stalk fibers. The rind of corn stalk has been used for manufacturing corn stalk fiber-based composite materials.[165]

4.4.3. Rice Straw

Rice straws (RS) belong to the family of non-wood bio-fibers. It is a residue obtained from the agricultural production of rice. Rice is the major crop in Vietnam, India, and Bangladesh, therefore a huge quantity of rice residues are available after rice husking.[166] Such residues are either used as animal feed or disposed of as waste by burning them in the field, associated with carbon emissions. However, embedding of such residues in polymer composites as reinforcement materials could reduce environmental pollution and find potential applications in the reinforcement of composites.[160]

4.5. Outlook on Sustainable Fibers

The application of natural fibers in FRP composites results in various limitations because of their reduced durability, high moisture absorption, and moderate mechanical properties. One of the major challenges identified with natural fibers when used as reinforcements for FRP composites is the incompatibility between hydrophilic natural fibers and hydrophobic matrices during mixing, leading to undesirable properties of the final composites.[167] The weaker adhesive nature of the polymer matrices introduces lower strength to the composite. Additionally, degradation due to the presence of water is another major challenge due to the higher hydrophilicity of the fibers. The presence of hemi-cellulose increases the swelling capacity of the natural FRP composites which makes them unsuitable for outdoor applications.[167] The variability in natural fiber properties like length and diameter is also a very challenging aspect during composite manufacturing. Therefore, to develop high-performance sustainable FRP composites, the current challenges with sustainable fibers such as poor interfacial properties, water absorption, materials inconsistency, and swelling need to be addressed.

5. Polymer Matrix

An FRP composite is a physical mixture of at least two components with different physical and chemical properties: reinforcement materials which are usually fibers, and a matrix which is usually a polymer. Polymer matrices are an essential part of a FRP composite, which can protect the fiber surface from mechanical abrasion, as well as hold and protect the reinforcing material from adverse environmental effects without disturbing its orientation and position within the composite (**Figure 9**a). Additionally, it acts as a stress transfer medium by distributing the applied load equally within the composite. Furthermore, it helps with the isolation of fibers, so that the individual fibers can act separately to stop or slow down crack propagation.[168,169] In this review, based on the sources, polymer matrices have

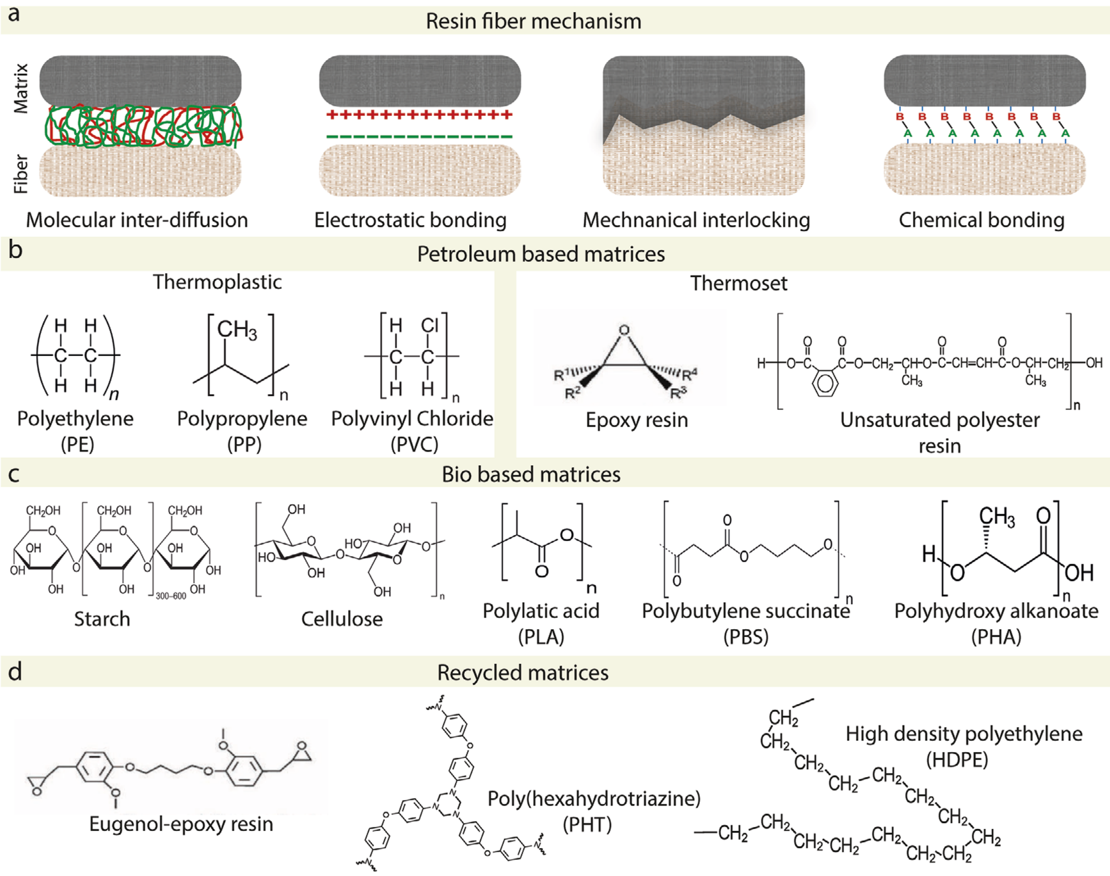

Figure 9. a) Resin-fiber mechanism for FRP composites. Examples of several polymer matrices used for manufacturing of fiber reinforced composites: b) Petroleum-based, c) bio-based, and d) recycled matrices). Reprinted with permission.[170] Copyright 2019, SAGE Publications.

been categorized into three major classes: synthetic, bio-based, and recycled. Major types of polymer matrix are shown with some examples (Figure 9b–d) and some of their key properties are illustrated in **Table 3**.

5.1. Synthetic Matrices

Synthetic or petroleum-based polymers, produced from abundant petroleum-based resources, are the most commonly used matrices for FRP composites. The common synthetic polymers are usually thermoplastics and thermosets (Figure 9b).

5.1.1. Thermoplastic Polymers

These are melt-processable plastics that can be easily heated and softened for shaping or molding. Thermoplastic polymers are generally high molecular weight compounds. The polymer chains in thermoplastics are associated with each other through intermolecular entanglements. When they are subjected to a raised temperature, such intermolecular entanglements weaken easily, resulting in a viscous liquid. In this state, thermoplastic polymers can be easily reshaped and are typically used in composites by various polymer processing techniques such as injection molding, compression molding, calendering, and extrusion. Such polymers have approximately ten times more resistance to impact, greater tolerance to damage, higher re-formability, as well as higher processing temperature and pressures as compared to thermosets.[182] Most commonly used thermoplastic polymers for fiber-reinforced composites are: polypropylene (PP), polyvinyl chloride (PVC), and polyethylene (PE). Among them, PP is perhaps the most widely used because of its moderate to good mechanical properties. Additionally, PP provides moderate dimensional stability, higher resistance to thermal deformation, and higher flame resistance.[171] Therefore, PP has become an obvious choice as a matrix for manufacturing natural fiber-reinforced composites. PVC is another thermoplastic polymer that is popular for building structures and construction sectors due to its compatibility with natural fibers, economical, durability, flame, and chemical resistance.[171]

5.1.2. Thermoset Polymers

These polymers undergo an irreversible chemical reaction during molding (often known as curing/cross-linking) to change from a liquid or rubbery state to the final solid state. Once they are fully cured, the shape of such materials cannot be changed significantly via heating. Thermoset polymers contain small, unlinked molecules, which usually require the addition

Table 3. Summary of some key properties of polymer matrices.

Matrices	Resin	Density [g cc^{-1}]	Tensile strength [MPa]	Elongation at break [%]	Young's modulus [GPa]	Compression strength [MPa]	Properties	Reference
Thermoplastic	Polyethylene (PE)	0.91–0.95	25–45	150	0.3–0.5	–	Low cost, good solidity, chemical resistance, ageing resistant	[171,172]
	Polypropylene (PP)	0.90–0.91	20–40	80	1.1–1.6	–	Low cost, good solidity, chemical resistance	[171,173]
	Polyvinyl chloride (PVC)	1.3–1.5	52–90	50–80	3.0–4.0	–	Low cost, weather resistant, non-inflammable, good haptics	[171,172]
	Polystyrene (PS)	1.04–1.05	35–60	1.6	2.5–3.5	–	Lightweight, water resistant, excellent shock absorption, anti-bacterial	[115,171]
Thermoset	Unsaturated polyester (UPE)	1.2–1.5	40–90	2	2.0–4.5	90–250	Poor linear shrinkage, excellent wettability of fibers, room temperature curable by addition of hardener	[171,174]
	Epoxy (EP)	1.1–1.4	28–100	1–6	3.0–6.0	100–200	Low cost and low toxicity, high strength, low shrinkage, excellent adhesion to fibers, chemical, and solvent resistant	[175,176]
	Phenolic (PH)	1.3	35–62	1–2	2.8–4.8	210–360	Good strength and dimensional stability, heat, solvents, acids, and water resistant	[176,177]
	Vinyl ester (VE)	1.2–1.4	69–86	4–7	3.1–3.8	86	Good strength and mechanical properties	[171,178]
Bio-based	Polylactic acid (PLA)	1.2–1.3	57–185	2.1–30.7	5.1–19.5	–	High strength, high modulus, good appearance, highly biodegradable, less green-house gases emission	[115,179]
	Polybutylene succinate (PBS)	1.26	39–55	5–12	3.6–7.4	–	Inherent biocompatibility and biodegradability	[115,180]
	Polyhydroxy alkanoate (PHA)	1.2–1.3	10–39	2–1200	0.3–3.8	–	Biodegradable with lack of toxicity, reduction of fossil usage	[115,179]
Recycled	High-density polyethylene (HDPE)	0.9–1.0	32.0–38.2	150	1.3	–	Stiff, durable, high-temperature stability, UV resistant, easily recycled	[115,181]
	Polyethylene terephthalate (PET)	1.5–1.6	55–159	300	2.3–9.0	–	Highly rigid, good tensile strength, good barrier effect, easily recycled	[115,181]

of another material (curing agent/cross-linker/catalyst) and/or heat to initiate the chemical reaction. During such reactions, the molecules cross-link, and form significantly longer molecular chains and a cross-linked network, which results in a solidified material. Thermoset matrices provide better resistance to creep, higher modulus, good stability to thermal variations, and higher chemical resistance than thermoplastic matrices.[183] However, such matrices are fragile and provide very low fracture toughness at room temperature. Most commonly used thermoset polymer's matrices for FRP composites are unsaturated polyesters, phenolics, epoxy, and vinyl esters.[184]

Epoxy Resin: Epoxy resins show excellent mechanical and chemical properties, corrosion resistance, good dimensional, and thermal stability, and are widely used as matrices for FRP composites.[185] Such polymer matrices have been an obvious choice owing to their good stiffness, dimensional stability, and chemical resistance.[186]

Unsaturated Polyester Resin: Unsaturated polyester, commonly known as polyester resin, is another popular and versatile synthetic co-polymer that is often used as the matrix for composites manufacturing. It is a linear chained polymer having an ester bond formed by polycondensation of unsaturated dibasic acids with diols or by saturated dibasic acids with unsaturated diols.[174] It is primarily used in compression molding, injection molding, RTM, hand lay-up, filament winding, and pultrusion processes.[187] Such resin is used in the manufacturing of about 85% FRP that is used in cars, boats, and aircrafts.[188]

5.2. Bio-Based Polymers

The sustainable approach to reducing the environmental impacts of synthetic polymer-based composites is to replace them with bio-based or biodegradable polymers. Such bio-based materials not only help in reducing carbon emissions by substituting fossil carbon but also impart added advantages including biodegradability, biocompatibility, carbon dioxide sequestration, and the reduction of global warming.[189] Bio-based polymers are biodegradable in nature as they can undergo deterioration upon exposure to aerobic, anaerobic, or microbial processes. Bio-based polymers are usually produced via three most common methods: 1) The direct use of natural bio-based polymers (starch and cellulose) with partial modifications; 2) The production of polymers from organic waste residues such as polylactic acid (PLA) and polybutylene succinate (PBS); and 3) The direct synthesis via microorganisms such as polyhydroxy alkanoate (PHA).[190]

5.2.1. Polylactic Acid (PLA)

Among various bio-based matrices, PLA is the most commercially popular polymer, which is also a feasible alternative to petroleum-based polymers. In terms of its biodegradability, PLA is a flexible polymer that can be optimized to degrade either over a very short period or a long period of time via varying their composition during synthesis.[191] It is well-known for good structural stability with high transparency, and is mainly used for making composites for packaging in the food industry.[192]

5.2.2. Polyhydroxy Alkanoate (PHA)

PHA polymers are aliphatic polyesters that can biologically be produced as cytoplasmic aggregations in various bacteria with specific nutrients and growing conditions, like abundant amount of carbon and deficiency of one or more important nutrients such as sulfur, phosphorus, nitrogen, oxygen, and other trace elements like iron, calcium, magnesium.[190] They possess high thermal stability, and are completely biodegradable and biocompatible polymeric systems which are perfectly suitable for an extensive range of composite applications.[193]

5.3. Recycled Polymers

Currently, polymers are either burned or end up in landfill at the end of their lifecycle, which is not environmentally friendly. In recent years, there has been a focus on alternative matrix materials that can be recycled and reused easily after the end of their life.[194] Commercial companies like Connora Technologies and Adesso Advanced Materials Co. Ltd. have developed acid-degradable and amine-based hardeners which allow thermoset epoxy to be broken apart upon the addition of a suitable acid.[195–197] A similar strategy has been developed using anhydride hardener that can easily be broken down by an acidic solution in the presence of a zinc chloride catalyst.[198]

Another approach includes the rebuilding of matrix materials entirely, developing new kinds of polymers that can be recycled or reused in a simple way. A eugenol-derived epoxy vitrimer has been developed, in which cross-links can be broken down at elevated temperature, allowing remolding.[199] In addition, several recyclable polymer materials have been developed in which monomers are reversibly linked through a boroxine ring.[200] Such polymers behave like classical thermosets, showing a good solvent resistance and mechanical properties but can be broken down in boiling water, reshaped, and reformed. Perhaps the most appealing fact from an environmental point of view is the idea of developing a polymer matrix that can be easily recycled back to its monomeric units for making into new polymer matrix.[201] Various approaches have been made for such polymers, like poly(hexahydrotriazine) (PHT) and self-immolative polymers.[202]

5.4. Outlook on Polymer Matrices

Recently, extensive research has been taking place in the field of composites to find suitable polymer matrices. The incorporation of these polymers in the composites shows several advantages like low cost, low density, and reduced abrasiveness.[171] However, further research is required to explore the scope and limitations of such polymeric materials. The research direction must focus on the development of materials that maintain a balance between the composite structure and properties, as well as the capability to be manufactured at scale and lower cost. As most of the polymer matrices used for manufacturing FRP composites are non-biodegradable, the first step towards sustainable FRP composites could be the use of bio-based polymer resins and/or partially degradable composites.

6. Modifications for Sustainable FRP Composites

6.1. Surface Functionalization of Natural Fibers

Jute, flax, hemp, and sisal are the four main dominating bast fibers, used as reinforcing materials for natural fiber reinforced composites. Among them, jute has gained significant attention due to its prospect to replace partially or fully conventional glass fibers owing to lower specific gravity and higher specific modulus. It's also the second-most produced natural fiber after cotton, found mainly in Bangladesh, India, and China, and is at least 50% cheaper than flax and other natural fibers.[13,84]

Natural fibers could offer an attractive alternative to synthetic fibers, due to their lower production cost, lower density, and long individual fiber length. However, natural fiber reinforced composites still suffer from poor mechanical properties in comparison to conventional synthetic fibers.[13] The mechanical

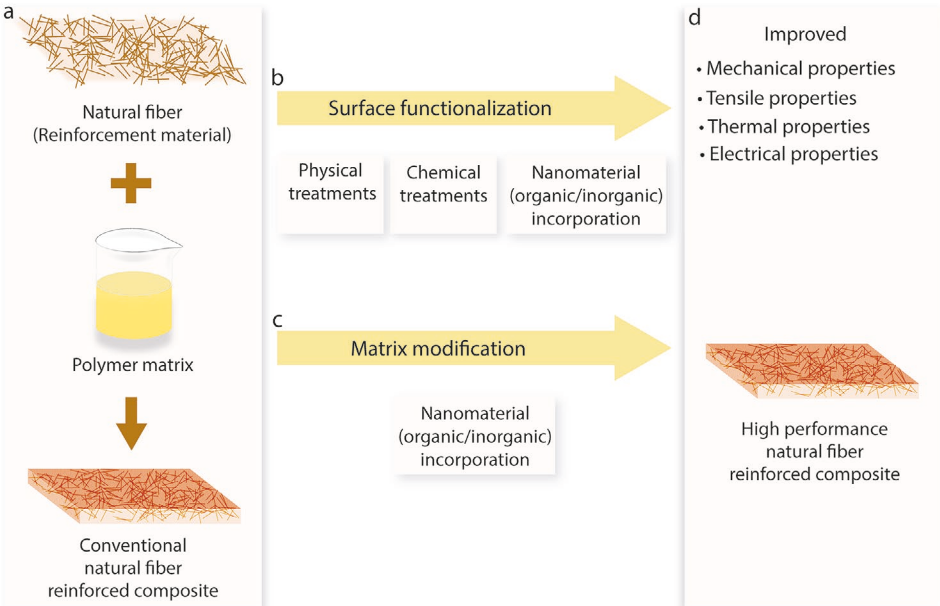

Figure 10. Improvement of a) conventional natural FRP composite properties by b) surface functionalization of fibers and/or c) matrix modification to produce d) high performance natural FRP composites.

properties of FRP composites depend on the proportion and properties of the constituent fibers and matrix materials, the orientation of fibers through the matrix, and the manufacturing methods.[17] However, being the main load-bearing constituent, the properties of reinforcement materials, that is, the fiber in the case of fiber reinforced composites, are dominant among all. **Figure 10** illustrates the scope for altering the properties of fiber-reinforced composites.

Several physical and chemical treatments have been reported to enhance the mechanical performance of natural fibers and their composites. Physical treatments do not change the chemical composition of fibers. They only modify the surface and structural properties of fibers. Some physical treatments include stretching, calendaring, combing, cold plasma treatment, and electric discharge.[203–205] In contrast, chemical modifications of natural fibers permanently alter the nature of fiber cell walls either by grafting polymers on the fiber surface, bulking or cross-linking within the fiber cell wall. Such treatments provide more dimensional stability, reduce water absorption capacity and give resistance to the fibers against fungal decay.[206] Alkali treatment is the most popular chemical treatment,[207] which removes impurities including wax, hemicellulose, and lignin from the fiber surface, separating elementary fibers from technical fibers to improve the fiber packing in composites, resulting in a strong fiber-matrix interface and improving the load bearing capacity of the reinforcement materials. It was evident that the treatment with a lower alkali concentration (≈0.5 wt.%) for a prolonged period of time is an effective technique to enhance the mechanical properties of jute fiber, though micro-voids can still produce a weak fiber/matrix interface. Attempts to overcome such flaws, either by nanomaterial grafting or other surface modifications including silane treatment, acetylation, etherification, peroxide and plasma treatment to enhance the composite's performance have been reported.[33,206] Additionally, several combined surface treatments have also been investigated[33] including alkali-silane,[208] alkali-plasma,[209] alkali-bleaching,[210] alkali-acetylation,[211] etc. Unfortunately, many such treatments are expensive and time-consuming but provides a minor improvement in composite performances.[12] Therefore, the field of research to investigate newer surface treatments to enhance composite performances still remains attractive.

Nanomaterials, due to their higher specific surface areas of up to 1000 $m^2 g^{-1}$, and for their unique mechanical, electronic, and thermal properties are presently considered high-potential filler materials for the improvement of mechanical and physical properties of multifunctional polymer material.[212] Several carbonaceous materials were reported to be utilized for the modification of natural fiber reinforced composites, in particular carbon nanotubes (CNTs).[35] Shen et al.[213] treated ramie fibers/epoxy composites with different CNT contents ranging from 0 to 0.6 wt.% and the investigation showed that, interlaminar shear strength (ILSS), flexural strength, and flexural modulus were improved by about 38%, 34%, and 37% respectively. Islam et al.,[214] increased the mechanical strength of jute fibers composites by dip-drying jute fibers with CNTs. They also demonstrated enhanced thermal stability, flame retardancy, electrical conductivity and showed potential to be applied in different electrical and electronic devices as well as in polymer composites as conductive fillers. Zhuang et al.,[215] dip coated hydrophilic jute fibers/fabrics in interconnected multi-walled carbon nanotube (MWCNT) networks to obtain electrically semiconducting jute fiber surfaces which were further used as temperature, relative humidity, and stress/strain sensors. Saiteja et al.,[216] also demonstrated an enhancement of mechanical properties for jute fiber reinforced composites by treating with a dispersion of MWCNT.

Graphene, since its isolation in 2004, has received huge interests from researchers attention due to its outstanding mechanical, thermal, electrical, and other properties.[217,221] Such incredible properties of graphene and its derivatives[222–224] were utilized for designing high-performance natural fiber composites.[35] Ganapathy et al.[225] used graphene as a filler for banyan aerial root fibers reinforced epoxy composites at various compositions. They reported that hybrid composites containing 4% of graphene achieved the highest tensile (40.6 MPa) and flexural strength (163.23 MPa). Alkali-treated jute fibers were coated with graphene flakes and graphene oxide (GO) by Sarker et al.[13] to enhance interfacial shear strength up to ≈236% and tensile strength up to ≈96%. Karim et al.[84] introduced reduced GO into jute fiber to improve the tensile strength and Young's modulus of the composites significantly by up to ≈183% and up to ≈450%, respectively. Additionally, organic materials, inorganic nanofillers such as alumina, magnesia, silica, zinc oxide, titanium dioxide, and calcium carbonate have also been used for fiber surface modification.[226]

6.2. Polymer Matrix Modification with Nanomaterials

Like fiber modification, many research groups have reported matrix modifications to improve the performance of FRP composites. Gojny et al.,[227] applied a standard shear-mixing technique to disperse double-wall nanotubes in an epoxy resin. They demonstrated that, in comparison to a carbon black filled epoxy, the addition of only 0.1 wt.% of CNT with epoxy resin leads composites to an increased tensile strength and Young's modulus while also retaining ductility. In another study,[212] they significantly improved the matrix-dominated properties (e.g., ILSS), by treating the glass FRPs with CNT/epoxy matrix although tensile properties were not affected. Costa et al.,[228] exhibited that, compared to control curaua fibers (CF) reinforced composites, CF reinforced composites having polymer matrix functionalized with GO offer superior tensile performance, higher yield strength (64%), tensile strength (40%),Young's modulus (60%) and toughness (28%). As found for the reinforcement modification, inorganic nanoparticles are also used for matrix modification of fiber reinforced composites.[229,230] Prasad et al.[231] introduced titanium dioxide (TiO_2) nanoparticles for the epoxy matrix reinforcement. Adding only 6% TiO_2 nanoparticle in flax fiber reinforced epoxy composites improved the tensile strength up to 12% and Young's modulus up to 23%. In another study, TiO_2 filled (4 wt.%) jute epoxy composite showed an increase in tensile and compressive strength by 31% and 34% compared to an unfilled composite.[232]

6.3. Hybridization with Synthetic Fibers

The inherent limitations of natural fibers such as having inferior mechanical properties along with a higher water absorption limit their use in comparison with commonly used glass and carbon fiber-based composites. Hybrid composites consisting of two or more fibers in one matrix are therefore seen as a solution to enhance the properties of natural fiber-reinforced polymer composites.[233] Such composites have already achieved mechanical properties equal to or sometimes even greater than conventional fiber-reinforced polymer matrix composites. Synthetic fibers are introduced along with natural fibers by an optimal stacking sequence, which imparts advantages of both fibers to the resultant composites.[234] Glass fiber is the most used fiber for hybridizing purposes due to its low cost, great availability, and ease of manufacturing.[8] However, several research groups have reported the hybridization of various natural fibers with different synthetic fibers. Examples include natural sugar palm fiber hybridized with glass fiber,[235] kenaf fiber hybridized with glass,[236] or Kevlar fiber,[237] oil palm empty fruit bunches fiber with glass fiber,[238] pineapple leaf fibers with glass fibers,[239] bamboo fiber with glass fiber,[240] jute fiber with glass,[241] or carbon,[242] hemp fiber with carbon fiber,[243] flax with glass fiber,[244] ramie fiber with glass fiber,[245] and sisal with glass,[246] carbon,[247] or aramid fiber.[248] Synthetic fiber films are also reported as hybridization materials. For example, Shamsuyeva et al.[249] investigated the effect of the epoxy-based coating of flax textiles on their tensile properties and corresponding PA6 biocomposites after thermo-oxidative aging.[250] They demonstrated, that the treated flax fiber reinforced composites demonstrate increased tensile strength and modulus for all test conditions. A combined treatment of silanization and partially bio-based epoxy resin coated flax fiber composites with PA6 film was also investigated.

6.4. Outlook on Modifications

From the above review, it is evident that the surface modification of natural fibers in composites is justified for engineering applications owing to their mechanical properties. However, there are a lot of challenges involved in controlling and improving the mechanical properties of natural FRP composites. Further exploration is also required from the research forum to support and motivate the utilization of natural fibers as well as novel modification techniques in advancements of natural FRP composites.[251] With the significant progress in materials science, it can be assumed that in near future these advancements will lead us towards the enhanced properties of natural FRP composites, particularly for novel applications. Future research is required to overcome the impediments like moisture absorption for long-term stability in outdoor applications, interfacial compatibility, swelling, etc.

7. Properties of Fiber Reinforced Composites

7.1. Mechanical Properties

Mechanical properties of sustainable FRP composites are the most important factors, determining their potential applications in various sectors including automobiles, aerospace, household, and sports. **Figure 11** illustrates important parameters, that generally influence the mechanical properties of FRP composites. They can be grouped into three categories a) reinforcing fibers; b) polymer matrices; and c) composites manufacturing processes. As fibers are used as reinforcement materials for FRP composites, the first thing to look out for is the

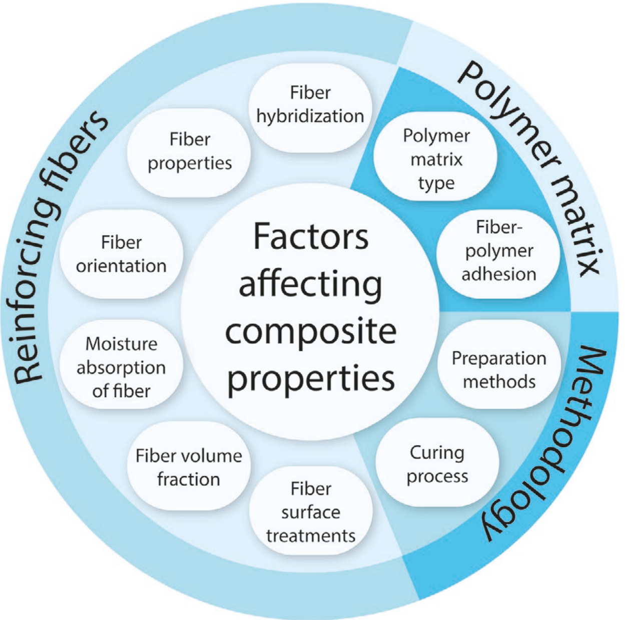

Figure 11. Influencing factors for mechanical properties of FRP composites.

fiber properties, including their orientations, moisture absorption, surface treatment, and hybridization.[252–254]

Enhanced mechanical properties of composites are achieved when the fibers are aligned in a direction parallel to the applied load. However, it is very challenging to obtain such alignment with natural fibers, as they tend to orient themselves randomly. The tensile strength and Young modulus of FRP reduces significantly with an increase in fiber orientation angle relative to the test direction.[254] Natural fibers are often carded and placed in sheets prior to matrix impregnation, in order to obtain a high degree of fiber alignment.[255]

It is often observed that the presence of moisture in the fiber hinders the ultimate strength of composites. Natural fibers are usually hygroscopic in nature, which has an effect on the mechanical properties of their composites. Fiber dispersion and volume fraction are the other two important factors that influence the properties of short natural fiber reinforced composites, which commonly have hydrophilic fibers and hydrophobic matrices. Longer fibers can increase their tendency to agglomerate. The good fiber dispersion promotes good interfacial bonding, thereby reducing voids by ensuring that the fibers are surrounded by the matrix.[256] Fibers can also undergo certain surface treatments or modifications in order to impart specific functionalities and enhance mechanical properties.

The polymer matrix plays a major role in fiber-reinforcement in a composite. They protect fiber surfaces from mechanical damage, act as a barrier against adverse environments, and transfer loads to the fibers. The most commonly used matrices in natural FRP composites are thermoset or thermoplastic polymers, as they are lightweight and can be processed at lower temperatures.[257] Fiber-matrix adhesion plays a significant role in determining the mechanical properties of composites.[258] As the stress is transferred between the fiber and the matrix, a good interface bonding between them is required to achieve optimum reinforcement and enable crack propagation. However, natural FRP composites have a limited interaction between the hydrophilic fibers and hydrophobic matrices, leading to poor interfacial bonding and mechanical performance. Such interfacial bonding can be improved via surface modifications or functionalization.[9,12,13,33,84]

As already mentioned, the common methods used for the manufacture of FRP composites are vacuum infusion, extrusion, compression molding, and injection molding. The speed, pressure, and temperature of such processes are key factors that influence the mechanical properties of FRP composites. It is possible for the fibers to degrade when they are subjected to high temperature, which limits the use of thermoplastic matrices as degradation can occur around the melting point of the polymer. In an extrusion process, thermoplastic polymers are usually in the form of pellets or beads, which are softened and mixed with the fiber by means of a single or two co-rotating screws, compressed and forced out at a steady rate through a die. High screw speeds result in fiber breakage, air entrapment, and excessive melt temperatures.[259,260] Whereas low screw speeds lead to poor mixing and insufficient wetting of the fibers. Better fiber dispersion and mechanical performance are achieved through the twin screw system rather than a single screw extruder.[261]

Recently, the mechanical properties of natural FRP composites have been enhanced substantially via better raw materials extraction and selection, interfacial engineering, and improved composite manufacturing processes.[262] In a previous study,[263] both untreated and treated banana fibers have been considered to develop hybrid composite material. The untreated banana fiber was treated with caustic soda in order to increase the wettability, and then used as a reinforcement material. It was found that after alkali treatment, there was an improvement in the mechanical properties including tensile, flexural, and impact strength of the hybrid composites. In another study,[264] natural bamboo fiber reinforced composite was used, investigating its tensile and flexural strengths, and surface hardness. The effect of fiber loading on mechanical properties of the composite was studied. It was found that maximum values of tensile & flexural, and surface hardness of the composite were achieved at a 25% wt.% loading. Hybridization of sisal fiber with coconut spathe and ridge gourd was reported by Girisha et al.,[265] which resulted in a significant enhancement in tensile properties when compared to the individual components. The tensile strength was found to be increased by ≈65%.

7.2. Tensile Properties

Tensile properties of natural FRP composites are primarily affected by the fiber-matrix interfacial adhesion.[182] They can also be improved via physical and chemical modifications of fibers and polymer matrices. It is largely dependent on the volume fraction of the fiber in matrix. Generally, when there is an increase in the fiber volume fraction up to an optimum level, there is more distribution of load among the fibers, and the applied force can be carried even after the fibers fracture which can lead to a higher tensile strength.[266] However, with further increases in the fiber volume fraction beyond the optimum level, the composite will become brittle, and the matrix is unable to withstand the additional load from the fibers, thereby resulting in a lower tensile strength.[267] Additionally, irregular trends for the tensile properties are also sometimes observed because of several factors that include fiber degradation, non-compatibility between the fibers and the matrix, and an inappropriate selection of manufacturing processes.[266]

7.3. Flexural Properties

Flexural stiffness is one of the important properties to measure the resistance of a composite against bending deformation. It mainly depends on two parameters: the moment of inertia and the modulus of the composite. It has been found that the flexural strength of natural FRP composites increases with the increase in fiber content up to an their optimum level.[268] However, further increases in the fiber content beyond the optimum level reduces the flexural strength, due to defects in the wetting of fibers that can create stress concentrations in the composites.[268] The fracture toughness of composites, which means a composite's resistance to crack propagation, is primarily affected by the fiber-matrix interfacial bonding strength, fiber volume fraction, and the intrinsic properties of the matrix.[183]

7.4. Compressive Properties

The compressive behavior of natural fiber reinforced composites mainly depends on fiber volume fraction and the reinforcement architectures.[183,269] The compressive strength of composites is found to be higher at a higher fiber volume fraction within its threshold point, due to a reduction in void.[183] Furthermore, high compressive strength can be attributed to a high fiber-matrix compaction and good homogeneity of composites. However, as the fiber volume fraction crosses the optimum level, the compressive strength decreases. Generally, when the fiber volume fraction crosses 1.5%, a reduction of compressive strength of nearly 8.5% occurs for every 0.5% increase in fiber volume.[270,271] However, it has been observed that the compressive strength of natural FRP composites gradually decreases when there is an increase in the fiber volume fraction.[271,272]

7.5. Impact Strength

The impact behavior of natural FRP composites primarily depends on the fiber-matrix bonding level, which is another important parameter for assessing the mechanical properties of composites. The impact properties of composites can be enhanced by various modification methods. The impact loading may be the result of debris, falling objects, crashes, and impacts. Some of the parameters that influence the impact strength of composites are energy absorption, fiber pull-out, favorable bonding, and the level of adhesion.[183]

7.6. Inter-Laminar Properties

In a study on interface modification and characterization of natural FRP composites, it has been observed that the quality of the fiber-matrix interface is required to identify the application of natural fibers as composite reinforcement.[273] Some researchers have also concluded that, because the fibers and matrices are chemically different in nature, strong adhesion at fiber-matrix interfaces is necessary for the successful transfer and distribution of stress. Fiber-matrix interfacial characteristics affect the behavior of various natural FRP composites, such as jute, flax, yarn, and woven fibers reinforced with PP, polyester, epoxy resins.[274] It has been noted that such natural FRP composites possess a high critical load for damage initiation, strong fiber-matrix adhesion, and high fiber strength and modulus. The alkali treatment of natural fibers like jute, kapok, hemp, and woven fibers can change their structure, which can increase fiber-resin adhesion as well as interfacial energy.[275]

7.7. Hardness

Lundquist et al.[276] investigated that a pulp fiber reinforced thermoplastic composite results in a strength increase by a factor of 2.3 and stiffness increase by a factor of 5.2 relative to the virgin polymer. Kenaf-maleated PP composites have a higher strength and specific modulus, at a lower cost than those reinforced with coir, sisal thereby making them good alternatives to the existing materials.[277] Ramanaiah et al.[278] in 2012, developed a new natural borassus seed shoot fiber reinforced composite with varying fiber volume content ranging from around 12–31%. The hardness of the composites decreased with an increase in the fiber content. Sreekala et al.[279] in 2002, investigated the mechanical behavior of hybrid phenol-formaldehyde-based composites reinforced with oil palm fibers and glass. Hybridizing oil palm fibers with glass fibers demonstrated improved properties, such as tensile strength, modulus, and flexural strength but poor hardness properties.

7.8. Tribological Properties

Wear and friction are the two vital tribological phenomena that occur due to the relative motion of solid surfaces, deteriorating materials, and dissipating energy.[280] Such properties of sisal fiber reinforced composites were studied at high temperatures. After studying the effect of different fiber volume fractions on the friction coefficient and the wear rate of such composites, it was found that the friction coefficient showed multiple trends at different temperatures whereas the wear rate significantly increased at higher temperatures. Generally, at higher fiber volume fractions, defects are found in the composites due to the poor dispersion of fibers in the matrix.[281]

The coefficient of friction (COF) is a quantitative number defining the frictional behavior of a material. In wear tests, a friction value is either the number determined at the end of the test or an average of values collected during the test. Wear is a progressive loss of material from the surfaces during chemical and/or mechanical processes.[282] The worn-out surfaces of kenaf fiber reinforced epoxy (KFRE) composite have been observed at different operating parameters. It is found that high thermo-mechanical activity is responsible for the debonding and formation of micro-cracks thereby deteriorating the fiber-matrix interfacial bond and increasing the wear on the application of high load.[283]

The tribological properties of brake pads were significantly enhanced by the addition of RS and rice husk dust into the composites.[284]In another study, the friction coefficient and wear rate of chopped sugar cane fiber reinforced polyester (CSCRP) were compared with those of chopped glass fiber reinforced polyester (C-GRP) composites at different lengths of fiber. For 1 mm of fiber it was found that the wear rate of CSCRP composites was nearly 10 times higher than C-GRP composites.[285]

7.9. Flame Retardancy

The potential applications of natural fibers have been limited by their poor flammability. Numerous research has been carried out to improve the flame retardancy of natural FRP composites.[286,287] Natural polymers and fibers are organic materials that are highly sensitive to fire. In the presence of a flame, the burning of a composite material occurs in five consecutive steps: heating, ignition, combustion, decomposition, and propagation.[288] The burning of composites mainly results in two forms of products with a high cellulose content and high lignin content. High cellulose content gives rise to higher flammability, whereas higher lignin content means that there is a greater chance of char formation.[289]

The flame retardancy of composites can be achieved by undertaking different procedures like the introduction of fire barriers, such as coatings and additives used as intumescents. However, another method of reducing combustion is by increasing the stability and char formation in the composite, resulting in reduced flammability, decreased visible smoke, as well as restricting the volume of products produced because of combustion.[290] Flame retardant coatings are another method that helps in the improvement of flame retardant properties of composites. Flame retardants like magnesium hydroxide ($Mg(OH)_2$), ammonium polyphosphate (APP), and a mixture of these two were incorporated into sisal fiber reinforced composites for the enhancement of flame retardancy.[291] Natural fiber PP composites containing $Mg(OH)_2$ were analyzed by Sain et al.,[288] where the flame retardant properties with boric acid were studied. It was found that 25% $Mg(OH)_2$ could reduce the flammability effectively almost by ≈50%. In another study involving kenaf/PP fiber reinforced composites,[292] APP was used as a flame retardant with three different compositions. The results showed that the flame retardant property of kenaf/PP composites was improved with an increase in concentration of APP irrespective of the types used.

Flame retardancy is mainly evaluated in terms of Limiting Oxygen Index (LOI), that is, the minimum amount of oxygen (vol%) required to sustain a stable combustion of any material. The higher the LOI value indicates better flame retardant properties of a material as illustrated in **Table 4**.

Table 4. Classification based on LOI values.[293]

LOI	Remark
< 21	Flammable
= 21	Marginally stable
21–28	Slow burning
28–100	Self-extinguishing
> 100	Intrinsically non-flammable

Another most important standard assessment method of flame retardancy is a UL 94 classification (**Table 5**). UL 94 is a plastic flammability standard released by the Underwriters Laboratories (USA).[294] It classifies plastics according to the manner in which they burn in various orientations and part thicknesses from the lowest to the highest flame retardancy in six different ratings.

Thermo-mechanical studies of bio-based composites derived from lactic and thermoset resins and flax and flax/basalt fiber reinforced composites were carried out, where mechanical and thermal properties of such composites were found to be better than the pure flax FRP composite.[295] Srinivasan et al.[296] found that a hybrid composite of banana, flax, and glass fibers has better thermal stability and flame resistance than the flax and banana fiber reinforced composites. In another study,[297] it was demonstrated that the thermal treatment of ramie fiber was efficient in modifying the inner structures and surface of the fibers.

7.10. Thermal Conductivity

The use of heat-insulating materials is a well-known approach to reducing energy costs and improving manufacturing efficiency in various sectors like packaging, construction, automobile, and aerospace. Natural fibers contain lumens which are an air-filled hollow portion, therefore the thermal conductivity of their composites decreases with an increase in fiber content. With an increase in fiber volume fraction, the amount of air contained in the composite also increases, thereby resulting in heat insulation.[298] Pujari et al.[299] investigated the thermal

Table 5. Classification based on UL 94 rating.[294]

UL 94 Rating	Remark
HB	Slow burning on a horizontal part
V-2	Burning stops within 30 s on a part allowing for drops of vertical flammable plastic
V-1	Burning stops within 30 s on a vertical part allowing for drops of plastic that are not in flames
V-0	Burning stops within 10 s on a vertical part allowing for drops of plastic that are not in flames
5VB	Burning stops within 60 s on a vertical part allowing for drops of plastic that are not in flames; plaque specimens may develop a hole
5VA	Burning stops within 60 s on a vertical part allowing for drops of plastic that are not in flames; plaque specimens may not develop a hole

conductivity of randomly-oriented banana and jute fiber-based epoxy composites and reported it to be very low at the maximum fiber volume fraction. Li et al.[300] found that the thermal conductivity of flax fiber/HDPE composite reduced with the increase of fiber volume fraction. Agrawal et al.[301] studied the influence of fiber treatment on the thermal conductivity of an oil palm fiber/phenolic composite and found that silane and alkali treatments increased the thermal conductivity of the composite more than an acetylation treatment, signifying that the latter was more appropriate than the former treatments for enhancing the heat-insulating characteristics of a natural fiber composite.

7.11. Acoustic Properties

Sometimes vibration is a desirable physical phenomenon. However, continuous vibration in structural materials often leads to problems like noise pollution and the formation of fatigue cracks.[302] Noise pollution is a very concerning problem in urban life. It causes not only sleep disturbance and annoyance, but also leads to severe health issues such as heart attacks and hearing loss.[303] Therefore, the development of materials with enhanced vibration damping properties is becoming increasingly important now-a-days. Presently, in many buildings, automobiles, and movie theatres, sound absorbing materials are used to mitigate the noise and vibration through the absorption of acoustic waves, as noise in the form of sound waves propagates through the sound absorber and gets absorbed.[304] Natural fibers are known to have ease in handling and good acoustic insulation properties owing to their viscoelastic behavior as compared to inorganic fibers like glass and carbon.[305] Because of such phenomena, natural fiber reinforced composites show better vibration damping properties. Researchers have studied the free vibration characteristics of banana/sisal fiber reinforced composite beams and found that the chemical treatment enhances the free vibration and mechanical properties due to the improvement of the interfacial bond of fiber-matrix system.[306] In other research it was found that the damping properties of flax fiber composites increased with the twist and crimp because of the enhanced intra-yarn and inter-yarn friction.[307]

7.12. Electrical Properties

Natural fiber reinforced composites can be used for electrical applications for shielding the cables, or as an insulator for wires.[308] The demand for natural fiber reinforced composites as dielectrics is increasing, because of their electrical insulating properties. The dielectric constant of any material depends on its polarizability and it increases with the increase in the polarizability of a material. However, the dielectric loss factor decreases with the increase in frequency at a constant temperature.[309] Joseph & Thomas[310] investigated electrical properties of banana fiber reinforced composites. Treatments with caustic, silanes, acetyl, and latex under heat, decreased the dielectric constant. It was also observed that the dielectric constant increased with a rise in temperature and decreased with a rise in frequency. After the removal of water, the value of the dielectric constant was decreased. To make wires, flexibility is also one of the vital criteria. The whole length of the wire can be easily accommodated in a small space by using flexible polymers. In the case of natural FRP composite manufacturing processes, hardeners are often used. When hardeners are used composites become brittle in nature. But when the same brittle composites show good electrical properties along with mechanical properties, they can be used for the fabrication of switchboards and other electrical panels.[20]

7.13. Water Absorption

One of the major problems with natural FRP is their high moisture regain.[311] Mechanical properties of FRP have a strong dependency on the fiber-matrix interface adhesion.[312] It is well known that the fibers are rich sources of cellulose, hemicelluloses, lignin, and pectins, all of which are having hydroxyl groups. Therefore, they are strongly polar while most polymers show considerable hydrophobicity. Water absorption of a fiber reinforced composite will depend on the absorption characteristics of both the reinforcing material, that is, the fiber as well as the polymer matrix. Generally, the high water intake by composite materials results in the development of pressure on nearby structures, swelling, increments in their deflection, a conceivable decline in their strength, and increased weight of wet profiles. Excessive water absorption of a composite will also create a suitable platform for microbial attack on the composites.[313]

7.14. Biodegradability

An important and characteristic functionality of sustainable fiber reinforced composites is their biodegradability, which originates mostly from microbial action.[124] Biodegradability is an indispensable and effective function for the waste treatment of green composites after completing their service life. However, the introduction of biodegradability in composites effectively brings about lower durability. Thus, there must be a control over the biodegradability behavior. Synthetic fibers like carbon, glass, and aramid reinforced composites are causing problems to the environment as they are not as easily degradable as natural fibers. The development of natural fiber reinforced composites promotes the use of environmentally friendly materials due to their biodegradability. Biodegradable materials, which are also bio-based in nature, are ideal for the development of a sustainable world. They have the potential to be used as recyclable products, as they can be collected, processed, and reused or left in the environment for natural degradation.[314] Rwawiire et al.[315] analyzed polymer matrices derived from natural resources like polyhydroxyalkanoates (PHAs), cellulose, thermoplastic starch (TPS), and PLA. They found that biodegradable composites have comparable properties to synthetic-based materials. Wang and Shih,[316] analyzed bamboo fiber structure and extraction methods. They reinforced the fiber with polymers and analyzed their mechanical properties, cost, and energy of extraction, CO_2 absorption, renewability, recyclability, and biodegradability. They found that bamboo fiber-based composites are comparable with glass composite in terms of their performance properties due to their low cost, lightweightness, and most importantly biodegradability.

There are mainly five types of biodegradable bio-based polymers commercially available: protein-based polymers, bacterial PHA-based polymers, cellulose ester-based polymers, PLA-based polymers, and starch-based polymers. In a study involving starch-based biodegradable reinforced composites with date palm and flax fibers, it was found that TPS composites are hydrophilic and biodegradable. The rate of biodegradation and water uptake was reported to be inversely proportional to the fiber fraction.[317]

8. Applications of FRP Composites

Natural FRP composites have become popular in automotive, aerospace, aircraft, biomedical industries, electrical parts, packaging, and construction sectors (**Table 6**), owing to their good mechanical performance, economical production, vibration damping, lightweight, sound attenuation, eco-friendliness, and biodegradability.[138,318] Natural FRP composites have widely been applied in the automobile industry for a long time. Henry Ford, and George Washington Carver, made the first attempt to use natural fibers in the automobile industry using hemp and flax fibers in 1941.[319] Similarly, Audi launched the A2 midrange car in 2002, where door trim panels were made of flax/sisal reinforced polyurethane composites.[320] In the past decades, legislation from North American and European governments have encouraged the application of natural FRP composites in the automotive sector. Biocomposites obtained from natural fibers are extensively applied in automobile parts, such as interior parts, dashboards, trays, headliners, seat backs, and door panels by various manufacturers and suppliers all over the world. Almost all major car manufacturers such as Toyota, Mercedes-Benz, General Motors, Ford, Chrysler, Daimler, BMW, and Audi are using bio-based composites in multitudinous applications.[321]

Usually, natural FRP composites are predominantly applied for interior automotive parts due to their relatively low mechanical properties and intrinsic moisture sensitivity. Various automobile interior parts including storage bins, package shelves, floor mats, seat backs/fillers/liners, dashboard, and indoor panels are typically made of natural fibers.[322] Examples of natural FRP composites used for automobile exterior components include flax/polyester composites in the engine and transmission enclosures for sound insulation, abaca–reinforced composites for the spare tire well covers, fender components, spoilers, bumpers, seat frame, and load floors.[323] Owing to a low environmental impact, low thermal conductivity, and the light weight of renewable materials, natural FRP composites have advanced their applications in architecture, building construction, furniture, soil blending, and masonry.[138]

8.1. Automobile

Traditionally composites used in the automotive sector are mostly made up of synthetic fibers such as carbon and glass as reinforcing materials. However, renewable and bio-based reinforcing materials are becoming increasingly attractive as alternatives to reduce environmental impacts.[358,359] Several natural fibers such as jute, hemp, flax, kenaf, sisal, and coir have been investigated to produce bio-composites for automobile applications, **Table 7**. Such biocomposites are popular for automobile applications, as they provide lightweight, improved fuel efficiency, and low production costs. They are used to manufacture various automobiles components such as seat pads, armrests, headrests, trunk covers, cup holders, door panels, and bumpers.[360] Additionally, biocomposites are effective in reducing noise and vibrations through damping.[361] For example, Ford uses bio-based cushions, seats made of soy foam, and hemp fiber composites in the front grills.[362] Similarly, interior panels of Mercedes-Benz use jute-based biocomposites, flax reinforced composites for trunk covers and shelves, and rear panel shelves are made up of sisal fiber composites.[363] Toyota uses kenaf fibers in their covers for tires, soy foams for seats, and PLA/PP-based biocomposites inside package trays, toolbox areas, and trims. Volkswagen also uses biocomposites for the construction of package trays, door inserts, panels, and flap linings.[322]

8.2. Aerospace

Almost half of the components of an aircraft are made of composites. The potential benefits of those composite components are they are lightweight and only require a simple assembly. Natural FRP composites have found use on a large scale for the development of aircrafts for civil transport and military fighters, helicopters, and the launching of satellites and missiles. Some of the aircraft components are made of conventional composites such as propellers, turbine engine fan blades, main wings, wing ribs, rear bulkhead, keel beam, engine cowlings, doors, elevators, airbrakes, spoilers, and the rudder.[364,365]

8.3. Construction

In the construction industry, biocomposites are often used to manufacture roof tiles, floor matting, ceilings, doors, windows, and window frames. Load-bearing applications include the manufacturing of tanks, pipes, beams, and floor slabs.[366] Furthermore, biocomposites are employed in the rehabilitation and repairing of various structural components. Due to better acoustic and thermal properties, natural FRP composites are used as soundproofing and insulating materials.[367] Hemp-lime-concrete composites have shown better sound absorption ability than any other binders.[368] LCA, ecological aspects, and durability properties are taken into consideration for the selection of any biocomposites for the construction sector. Lightweight and good mechanical properties are very important for such applications.[321,369]

8.4. Medical

Composites are often used in medical devices where they are progressively replacing glass and metals. Over the years, the

Table 6. Potential applications of natural FRP composites.

Sector	Fibers	Applications	Ref.
Civil construction	Banana	Compressed earth block	[324]
	Jute, sisal, ramie, pineapple	Cementitious materials	[325,326]
	Flax, jute, sisal, hemp, coir, palm	Masonry	[327]
	Jute	Deck panel	[328]
	Kenaf	Ceiling	[329]
	Wheat straw, corn husk	Thermal insulation materials	[330]
	Wood cellulose, cork	Thermal insulation materials	[331]
Furniture and architecture	Lignocellulose, straw	Lounge furniture	[332]
	Hemp	Chair furniture	
	Hemp, flax	Ignot bio- and Polycal acoustic panel	
	Lignocellulose	BioMat research pavilion	
Sports and clothes	Hemp, jute, bamboo, sugarcane bagasse, coconut, banana	Footwear	[333]
	Flax, hemp	Racing bicycle	[334]
	Flax	Bicycle frame	[335]
	Jute	Winter overcoat	[336]
	Jute, sisal, coconut, areca, banana	Helmet shell	[337]
	Kenaf	Ballistic armor materials, mobile phone casing	[334,338,339]
	Kenaf, pineapple	Recurve bow	[340]
	Palm	Sports utility	[341]
Aerospace	Hemp	Electronics racks for helicopter	[342]
	Ramie	Aircraft wing boxes	[343]
	Kenaf	Aircraft materials	
Biomedical and pharmaceutical	Sugarcane	Drugs, antimicrobial, antibiotics	[344]
	Flax, ramie	Bone grafting, orthopedic implants	[345]
	Hemp, sisal, coir	Orthoses materials	[346]
	Jute	Enzyme	[344]
	Jute, sugarcane, flax, bamboo	Biomedical nanoparticles, antibiotics	
	Sisal	Drug delivery	
Others	Bamboo	Packaging	[347]
	Banana, bamboo, flax, jute, kenaf, palm, sisal	Dielectric materials	[348,349]
	Flax	Electrodes	[350]
	Flax, jute, coir, sisal	Wind turbine blades	[351]
	Flax, seagrass	Marine materials	[352,353]
	Jute	Solar parabolic trough collector	[354]
	Jute, flax, kenaf, hemp	EMI shielding	[355,356]
	Wood cellulose	Battery	[357]

advancement in natural materials, sterilization techniques, and surgical methods have allowed the use of composite materials in many ways. At present, medical practice utilizes a large number of medical implants and devices. Composites in the form of artificial hearts, pacemakers, heart valves, biosensors, dental implants, intraocular lenses, vascular grafts, joint and bone replacements, and sutures are widely used to restore and/or replace the function of degenerated/disturbed organs or tissues, to help in healing, to enhance function, and to rectify abnormalities, thereby enhance the quality of patient's life.[370]

8.5. Sports

Composites are used for sports equipment because they are lightweight, strong, highly resistant to wear and tear, are fatigue and friction resistant, are easily transportable, thermally stable, highly durable, and have good shock absorption properties.[341] They also provide flexibility in design and fabrication, therefore can be processed and shaped very easily. There are multitudinous goods made of natural FRP composites materials that include bows and arrows, hockey sticks, softball bats, tennis and badminton rackets, sailing and planning boats, and sailboards.[371,372]

Table 7. Natural FRP composites in automotive.[320]

Automobile	Fibers	Applications
Audi	Sisal, flax	Seat back, side, and back door panel, boot liners, hat rack, spare tire liners
BMW	Sisal, flax	Door panels, headliner panels, boot liners, seat backs, noise insulation panels, molded foot well liners, bumpers, fender liners, shields
Chevrolet	Flax	Trim panels
Citroen	Vegetable, wood	Interior door paneling, parcel shelves, boot liners, mudguards
Daimler	Sisal, hemp, flax, coir	Door panels, engine and transmission covers, pillar cover panels, windshields, dashboards, business tables
Fiat	Bamboo	Door panels
Ford	Wheat straw, Tomato, rice husks, hemp, kenaf	Floor trays, door panels, B-pillars, boot liners, wiring brackets, storage, front grills
General Motors	Kenaf, hemp, flax	Seat backs, inner door panels, cargo area floors
Lotus	Kenaf, sisal, hemp	Body panels, spoilers, seats, interior carpets
Mercedes-Benz	Banana, flax, sisal, hemp	Inner door panels, door panels, rear panel shelves, engine encapsulation, trunk covers
	Wood, sisal, flax, abaca	Door panels, engine encapsulation, spare wheel pan covers, door liners, seat backrests, parcel shelves, trunk covers
Toyota	Sweet potato, Sugarcane, kenaf	Door panels, seat backs, floor mats, spare tire covers, and package shelves
	Kenaf	Spare tire covers
Volkswagen	Sisal, Flax	Door panels, seat backs, boot-lid finish panels, boot liners
Volvo	Wood, rapeseed, jute, hemp	Seat padding, natural foams, cargo floor trays, dashboards, ceilings

8.6. Electrical and Electronics

Natural FRP composite materials are characterized by high strength and modulus, low thermal expansion, good electrical and thermal conductivity, and a low dielectric constant. The application of composites in electrical and electronics include inter-connections in housings, heat sinks, connectors, electrical contacts, thermal interface materials, light emitting diodes (LEDs) interlayer dielectrics, and printed circuit boards.[373–375]

8.7. Others

Natural fiber reinforced composites are extensively used in storage tanks, columns, reactors for acidic and alkaline environments, structural supports, pumps and blowers, exhaust stacks, piping, ducting, scrubbers, industrial gratings, composite vessels, casings, stacks, ducts, fan blades, and drive shafts.[376]

9. Outlook

The application of sustainable and natural FRP composites is increasing rapidly owing to their numerous advantages including their eco-friendly nature, biodegradability, and low cost, with relatively good mechanical properties. Thus, the natural FRP composites market is estimated to reach nearly 3.7 million tons in 2022, and is expected to register a compound annual growth rate (CAGR) of more than 9% during the forecast period (2022–2027).[20] Globally, the primary manufacturers of natural FRP composites include Trex Company, Inc., The AZEK Company, and Fiberon LLC. Among them, Trex Company, Inc. has the largest market share of over 10%. Asia-Pacific is the largest market that holds a share of production of over 30%. It is anticipated that the biggest market for natural FRP composites will be the building and construction sector followed by the automobile sector. Natural fibers are becoming an integral part of replacing synthetic fibers as the reinforcing materials for composites due to their great contributions towards a less polluted environment through eco-friendly materials consumption in different sectors.

Synthetic fibers have traditionally been an integral part of composite materials. However, the global scenario and outlook has completely turned into approaches leading toward biodegradability, recyclability, and sustainability. New environmental challenges and concerns have triggered the search for novel and sustainable products and processes. Engineering natural fibers with functional nanomaterials improves the mechanical properties of the composites. Instead of conventional petroleum-based polymers, using bio-based polymer matrices potentially reduces environmental hazards as well as enhancing biodegradability. Newer fibers have been developed which have opened up new market opportunities, since there has been a growing awareness about the importance of biodegradability and sustainability among the general population. It is an interesting fact that natural FRP composites are emerging as a realistic alternative to glass and carbon fiber reinforced composites (**Figure 12**a). Additionally, researchers have been investigating continuously to improve the manufacturing strategies to optimize material wastage, manufacturing time, and overall composite costs.

Figure 12. Future research direction of natural FRP composites.

Biocomposites could potentially replace petroleum-based composite materials for smart and multi-functional applications.[377] If we investigate the applications of biocomposites, it has demonstrated potential for the automotive and construction sectors in a major proportion, which is currently the leading market for these materials. However, the challenge remains in replacing conventional synthetic FRP composites with those that exhibit comparable functional and structural stability upon use, storage, and environmental degradation on disposal.[378] In addition to an improvement in the functional performance, research attention is also needed to overcome current barriers to using natural fibers. One of the barriers could be the compatibility of natural fibers in comparison to conventional glass or carbon fibers in terms of processing, functionalization, etc. Also, the low annual availability, as well as seasonality hinders the expanded use of natural fibers as reinforcing materials for composite manufacturing. Since, "sustainability" has become a key talking point for the industry in very recent years, researchers and composite manufacturers have been trying to find a balance between the performance of composites and sustainability. However, the relative advantages of natural fiber composites in comparison with conventional materials must be emphasized when developing new products, especially keeping in consideration the growth of the synthetic composites industry and their corresponding environmental effects. Though benefits such as reduced carbon footprint, biodegradability, and renewability are important, they are not easily recognized and appreciated by potential users, which significantly limits the spread and adoption of natural FRP composites. However, the market and commercialization of biocomposites are expected to expand in the near future owing to the identification of novel applications and the development of efficient manufacturing technologies. Research is being carried out on bio-based materials to overcome some hindrances such as high moisture absorption, inadequate toughness, and reduced long-term stability that are required mainly for outdoor applications. It is reported that one of the major hurdles for commercialization of sustainable FRP composites until recently is the inadequacy in the recognition of research and development in developing nations, where such fibers are abundantly available. At the same time, it is noteworthy that such a hurdle has been overcome by many industrialized nations, especially in UK and Europe who are taking the lead in the development of sustainable FRP composites in recent years.[379] New generations of biocomposites are eventually expected to be used in a wide range of applications majorly in mass-produced consumer products for short-term as well as long-term indoor applications[380,381] (Figure 12b).

Biocomposites are sustainable and can be totally recyclable (Figure 12c), but at the same time can be more expensive. Because of their recyclability, such composites are getting extensively popular in the packaging sector. Upon the selection of a proper matrix, biocomposites can also be totally biodegradable, but their biodegradation will be difficult to control. Biocomposites possess good specific properties, but there is a high variation in their properties. However, such problems can be overcome by suitable modifications and advanced processing of natural fiber and their composites. Recent advancements in genetic engineering, the development of natural fiber, and composite science and technology will offer valuable opportunities for enhanced value-added materials from renewable resources with improved support for global sustainability campaigns. Natural fibers are mainly biodegradable, but renewable resource-based biopolymers can be generated to be either biodegradable or non-biodegradable as per the specific demands for a particular application. Such a unique balance of properties can generate novel applications and at the same time can open up new market development opportunities for biocomposites in the 21st-century green materials world.

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Conflict of Interest

The authors declare no conflict of interest.

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