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PROPERTIES AND APPLICATIONS OF COMPOSITES REINFORCED WITH NATURAL FIBERS – A BRIEF REVIEW

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Abstract: The current concern on the environmental appeal provides a search for the development of eco-friendly materials that are applied in the most diverse areas of knowledge. In this scenario, natural lignocellulosic fibers (NLFs) are highlighted, because in comparison to conventional synthetic fibers they have several advantages, such as lower density, biodegradability and abundant availability, cause low damage to equipment. Additionally, their wastes do not pollute the environment. These characteristics make them a promising alternative for use as reinforcement in composite materials. There is a growing interest on the part of the academic community in investigating the properties presented by composites reinforced with natural fibers (CRNFs), thus generating a high number of published works related to this topic. Various methodologies are investigated with the aim of causing a significant improvement in the properties presented by the CRNFs, such as fiber surface treatments, manufacturing processes or processing temperatures. The properties presented by these composites are directly dependent on the matrix, the fiber reinforcement and the interface region between these phases. CRNFs are growing in number and degree of applications, being observed in different areas such as civil construction, naval, military, medical, fashion, automobile and several others, where all of them need materials that exhibit exceptional properties allied to low weight. The present work intends to present an up dated brief review on CRNFs, where due emphasis are given to the manufacturing methods, properties and main applications for these materials.

Keywords: Natural fiber, composites, properties, applications.

ABBREVIATIONS

The following abbreviations are used in this manuscript:

d Fiber diameter

E_{Comp} Longitudinal modulus of elasticity of the composite

E_f Modulus of elasticity of fiber

E_m Modulus of elasticity of matrix

L_c Critical fiber length

V_f Fiber volume fraction

V_m Matrix volume fraction

σ^*_{cl} Longitudinal tensile strength

σ^*_f Fiber tensile strength limit

σ'_m Stress in the matrix at the point of failure

σ_f Fiber tensile strength limit

τ_f Shear stress in the fiber

τ_m Shear stress in the matrix

τ Matrix shear yield stress

AFM Atomic force microscopy

CM Ceramic matrix

CMC Ceramic matrix composite

CRNFs Composites reinforced with natural fibers

DMA Dynamic mechanical analysis

DSC Differential scanning calorimetry

DTG Derived thermogravimetry

FM Flexural modulus

FR Flexural resistance

FRPMC Fiber reinforced polymer matrix composites

G Graphene

GFRP Glass fiber reinforced polymers

GO Graphene Oxide

MASs Multilayer armor system

ME Modulus of elasticity

MMC Metal matrix composite

NLF Natural lignocellulosic fiber

PAC Polyacrilato

PA Polyamide

PALF Pineapple Leaf Fiber

PES Polyethersulfone

PI Polyimide

PLA Lactic Polyacid

PMC Polymeric matrix composite

PPS Polyphenylene Sulfide

rGO Reduced graphene oxide

TG Glass transition temperature

TGA Thermogravimetric analysis

TS Tensile strength

RT Room temperature

INTRODUCTION REMARKS

In recent decades, efforts to develop new materials have been transferred from conventional monolithic materials to composite materials with the purpose of meeting emerging industrial needs (REDDY, YOGANANDAM and MOHANAVEL, 2020).

The evolution of fiber composites began alongside the discovery of plastic materials, although natural resins were already used as bonding materials. During the 1900s, synthetic plastics such as vinyl, polystyrene, phenolic and polyester were discovered. However, they could not meet certain applications, such as load transfer parts in automobiles, aircraft, sports equipment, wind turbine blades and many others. Due to the low resistance presented by plastics, reinforcements were introduced to improve their properties (VIGNESHWARAN et al., 2020).

Synthetic fibers such as glass, carbon and aramid fibers are the most common reinforcing materials among composites due to their unique mechanical behavior in various applications (VIGNESHWARAN et al., 2020). Composites reinforced with synthetic fibers provide high strength, rigidity and have been widely used in aerospace and automotive applications (AHMED et al., 2021).

Although synthetic fibers are attractive in their mechanical characteristics, they are neither biodegradable nor ecologically friendly. In search of alternatives to synthetic fibers, natural fibers have emerged as possible substitutes (ZHAO et al., 2022). As they present availability, renewability, cost-benefit and high specific properties, natural fibers have proven to be a promising substitute for synthetic fibers in the reinforcement of composites (ARULVEL et al., 2021).

Composites reinforced with natural fibers have attracted increasing interest as an alternative to metals and synthetic fiber-reinforced composites, mainly due to increasing demands for lightweight materials and an environmental appeal (LI et al., 2020). Natural fibers compared to synthetic fibers have several advantages, such as: lower density, biodegradability, abundant availability, good cushioning properties, less damage to equipment due to abrasion and high health safety (low skin irritation), making them a promising alternative reinforcing material in composites (REDDY, YOGANANDAM and MOHANAVEL, 2020). A wide variety of natural fibers are currently available as reinforcement for polymer composites, the most used of which are flax, hemp, jute, kenaf and sisal, due to their properties and availability (ABBAS et al., 2022).

Composite materials cover a wide range of applications, such as water treatment, photocatalyst, cable rails, usage cells, windows, chimneys, door panel, partitions, adsorbent, swimming pools, furniture, bathrooms, robotic arms, insulation, armor, covers, antennas, boxes, windmills, gears, bearings, golf clubs, steering wheels, tubes, drawing tables, wheels, bows, arrows, protective helmets, oars and many others (AKTER and HOSSAIN, 2021).

Owing to the existence of a weak interface interaction, CRNFs can promote failures in low-load applications. Thus, the strength of the interface between fibers and matrix plays a crucial role in the mechanical and wear properties of composites. In the search to improve the mechanical resistance of composites, several authors have addressed various fiber cutting methodologies, chemical treatments and/or surface coatings (ARULVEL et al., 2021).

Due to the high importance of studying the characteristics presented by CRNFs, the present work aims to present an updated brief bibliographic review regarding topics related to these. The review highlights the main research carried out today, with special emphasis on the properties and applications listed by various authors.

LITERATURE REVIEW

BRIEF HISTORY OF COMPOSITE MATERIALS

The development of new projects is intrinsically linked to the use of materials that can satisfy the properties inherent to the desired application. In this context, as Chawla (1993) presents, there would be no point in developing an advanced and innovative design of a turbine or aircraft if suitable materials were not available for use to meet the needs, related to supporting the loads and service conditions. Given this, it can be inferred that, in any area of technological development, the final limitation of advancement depends directly on the materials. Thus, composite materials present themselves as a major step towards the optimization and applicability of materials (CHAWLA, 1993).

Humanity has benefited from the use of composite materials for a long time, however, the understanding of the behavior presented by these materials occurred quite recently in history. One of the first uses of composite materials reported by archeology dates back to prehistoric times, where there is evidence of the use of straw to reinforce clay in the production of bricks (CHAWLA, 1993).

Generally, the term composite is associated with cutting-edge technological applications, where parts will be produced for use in components of satellites, aircraft, bioengineering, vehicles, maritime platforms, telescopes and intelligent structures. However, the origin of this class of materials dates back countless thousands of years, through the use of bones and wood (NETO and PARDINI, 2016).

Nature is responsible for a infinity of examples of composite materials, such as wood, leaves, bones, hair, animal hooves, horns and many other cases, thus showing that knowledge about the existence of these materials is not that current.

The development of our society was achieved by the use of accessible materials necessary for evolution. As such in prehistoric times ceramics, glass, natural polymers and composites were used (ASHBY, 2005). The polymer and composites industries show increasing evolution compared to the rate of development of new metallic alloys (ASHBY, 2005). This portrays the importance of using certain classes of materials in their respective technological eras, thus highlighting their high importance for the development of civilizations, as illustrated in Figure 1.

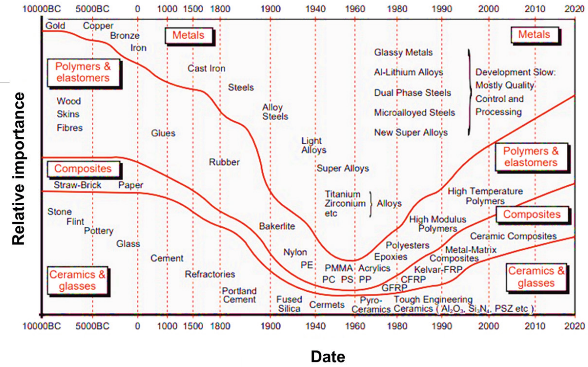

Figure 1: Importance of materials in their respective technological eras. Adapted with permission from Ashby (2005).

Composite materials presented themselves as an emerging class of distinct materials in the mid-20th century through the production of so-called multiphase composites, which were designed and "engineered". From then on, there was accelerated growth directed towards the applications of composite materials, especially polymers reinforced with glass fibers (CALLISTER JR and RETHWISCH, 2016).

According to the Brazilian Association of Composite Materials (ABMACO, 2008), composite materials produced in an "engineered" way appeared in the North American army at the beginning of the Second World War. This emergence occurred with the development of glass fibers reinforced polymers (GFRP), which were presented as light, strong materials resistant to corrosion caused by seawater. At the beginning of the 1950s, composite materials definitively entered the production lines of American automakers. Through their use, the bodies of the Chevrolet Corvette were shaped (a fact that continues to this day) (ABMACO, 2008).

Composite materials, especially GFRP, arrived in Brazil around 1954 with the creation of the first polyester resin factory. However, before that, enthusiasts for new products were already importing rolls of glass fibers and cans of resin to "play inventors" with composites

(ABMACO, 2008).

When it comes to the application of natural fibers in composites, this feat has been observed for centuries, including flax, hemp, jute, sisal, among others. Currently, there is considerable interest in the use of natural fibers for reinforcement in polymeric resins (BALEY, BOURMAUD and DAVIES, 2021). In fact, this interest was reported by De Bruyne (1939), when the author described the development of these materials aimed at aircraft applications.

In recent decades there has been a growing interest on the part of the scientific community in investigating the properties presented by CRNFs, this interest is partly due to the excellent properties presented by the combination of different materials. Several studies aimed at evaluating the properties of different CRNFs were observed, such as: carnauba (JUNIO et al., 2020), kenaf (SILVA et al., 2021), titica vine (CUNHA et al., 2021), caranan (SOUZA et al., 2020), seven-islands-sedge (NEUBA et al., 2020), hemp (RIBEIRO et al., 2021), sugar cane (ASYRAF et al., 2021), guaruman (REIS et al., 2021), banana (SRINIVASAN et al., 2021).

COMPOSITE MATERIALS

The emergence of new technologies is combined with the application and development of new materials, which generally must have their own characteristics to satisfy the needs required for practical use. In this scenario, the use of composite materials is presented as a means to overcome this lack. Therefore, composites arise from the need to improve the properties of pre-existing materials, such as metals, ceramics and polymers.

For millennia, the understanding for manufacturing multiphase materials, such as bricks and clays reinforced with straw, was not well established. Therefore, the strategy of combining different materials during the manufacture of a given component led to the identification of what is known as composite materials, which are distinct from conventional metals, ceramics and polymers. For (CALLISTER JR and RETHWISCH, 2016), the new concept of composite materials opened up an infinite number of design opportunities, thus being able to obtain a high variability of parts with different properties, characteristics that could not be achieved by unitary materials.

Initially, composite materials were also called conjugated (CHAWLA, 1993) or reinforced (MOROZOV and VAZILIEV, 1989). To avoid misleading comparisons, some definitions were raised to identify composite materials (CHAWLA, 1993):

a) The material must be manufactured, i.e., natural composites such as wood are excluded;

b) It must consist of at least two physically and/or chemically distinct phases, suitably arranged or distributed with an interface that separates them;

c) It must present properties/characteristics that are not exhibited by any of the components in isolation.

Composite materials must present a balance between the properties of their constituents, therefore, they must present the so-called principle of combined action (CALLISTER JR and RETHWISCH, 2016). In general, a composite is a multiphase material that presents a considerable fraction of the properties of the constituent phases, in order to obtain a better combination of these characteristics. The properties highlighted by the composites can be correlated to various factors, among the main ones the geometry and distribution of the phases present in the material stand out (NETO and PARDINI, 2016).

According to what has been discussed so far, composites are materials artificially produced by man, the phases that constitute them must be chemically different and thus separated by a region known as interface (CHAWLA, 1993; CALLISTER JR and RETHWISCH, 2016). This interface region is also responsible for providing the composites with variability in their properties.

Unlike traditional isotropic materials, like metals, composites, due to the variability of the properties of their constituents, present a high complexity (NETO and PARDINI, 2016). Therefore, isotropic materials have well-defined, repetitive and predictable properties, which cannot be observed in most composites. For Neto and Pardini (2016), this characteristic becomes a favorable factor for the production of composites, since it is possible to adjust the manufacturing parameters and thus achieve the required properties, meeting a specific parameter of a given project.

The vast majority of composite materials have two constituent phases, where the first is called the matrix phase and the second called the disperse or reinforcement phase (CALLISTER JR and RETHWISCH, 2016). The properties of composites are directly dependent on the characteristics exhibited by the phases present, making the relative quantity, geometry, distribution and orientation of reinforcement throughout the material structure.

MATRIX PHASE

Composites have a continuous phase called matrix, which can be a metallic, ceramic or polymeric material. The matrix has the main function of transmitting part of the mechanical stress imposed on the material to the dispersed phase (reinforcement) (CALLISTER JR and RETHWISCH, 2016). Generally, the matrix acts as a barrier to the propagation of cracks, as well as protecting the reinforcement phase.

In view of the above, composites can be classified as: polymer matrix (PMC), ceramic matrix (CM) and metal matrix composites (MMC). Among the PMCs most common are those reinforced with fibers (FRPMS), in these materials the reinforcement of a polymeric matrix occurs through the use of fibers, whether synthetic (MACIEL et al., 2018), natural (OKAFOR et al., 2022; MOHANKUMAR et al., 2022) or hybrid reinforcement (JAWAID et al., 2022; NAYAK et al., 2022).

CMCs are generally used in aggressive environments, as this class of material has resistance to both corrosion and high temperatures (VEIGAS, NAJIMI and SHAFEI, 2022). MMCs are considered advanced materials, as they present excellent mechanical properties with high stiffness, high specific strength, corrosion resistance, and in some cases desirable electrical and thermal properties (KANAKIN, SMIRNOV and KONOVALOV, 2022). The classification of composites is presented by Chung (2010), which can be seen in Table 1.

The matrix phase can be composed of the three main classes of materials, however, metals and polymers are more common for applications where the reinforcement phase presents a fibrous morphology. For these classes of composites, good ductility on the part of the matrix is desired. Such characteristic is not observed in CMC (CALLISTER JR and RETHWISCH, 2016).

In many applications, composite materials are superior compared to other materials, as they have high resistance, high rigidity, good moldability, weather resistance, all combined with great lightness (ABMACO, 2008).

Countless composite materials are known to present high levels of specific resistance and stiffness, even when subjected to combined tensile-compression, flexion, torsion efforts,

Class	Matrix Type	Reinforcement	Examples
PMC	Thermoplastics (e.g., PPS, PES)	Filling	Metal or ceramic powders, particles, spheres
		Fibers	Carbon monofilaments, glass fibers, natural fibers and whiskers
		Laminates	Glass sheets, aluminum sheets
	Thermosetting (e.g., epoxides, PI, PA, PAC)	Filling	Metal or ceramic powder, particles, granules
		Fibers	Carbon monofilaments, glass fibers, natural fibers and whiskers
		Laminates	Glass sheets, aluminum foil, honeycomb
	Elastomers (rubber)	Filling	Graphite powder, particles, granules
		Fibers	Carbon monofilaments, glass fibers, natural fibers and whiskers
		Laminates	Glass sheets
MMC	Metals (Al, Mg, Ti, Cu)	Particles or flakes	Ceramic, hard metal, diamond-like carbon;
		Fibers	SiC or B_4C or monofilaments, whiskers
	Alloys	Particles or flakes	Ceramic, hard metal, diamond-like carbon
		Fibers	SiC or B_4C or monofilaments, whiskers
		Others	Expanded metal, mesh, honeycomb
CMC	Ceramics	Particles or flakes	Ceramic, hard metal, diamond-like carbon
		Monofilaments	Carbon and/or whiskers
		Metal fibers	Cutting wires and whiskers
		Others	Expanded metal, mesh, honeycomb
	Glass or Glass-Ceramic	Particles	-
	Carbon-Carbon	Monofilaments	Whiskers, honeycomb fabric

Table 1: Classification of composites according to matrix and type of reinforcement. Adapted with permission from Chung (2010).

thus such properties are obtained through the combination of the characteristics presented by the matrix phase in association with the reinforcement phase (NETO and PARDINI, 2016).

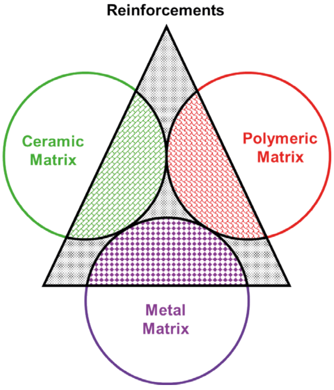

Figure 2: Possible combinations in composites with ceramic, metallic and polymeric matrices. Adapted with permission from Neto and Pardini (2016).

Among the composites, those with the greatest industrial employability are those made from polymer matrices, thus being used in a greater diversity of applications. This fact is directly linked to their properties at room temperature (RT), involving both ease and manufacturing cost (CALLISTER JR and RETHWISCH, 2016).

The increasing use of polymer matrices in the production of composites may be combined with the mechanical properties presented by these materials. Indeed, they present lower resistances than metals and ceramics, meaning there is a great need to improve these properties. Therefore, the viable alternative becomes the use of reinforcements to improve the polymer matrix.

Polymer matrices can be thermoplastic, thermorigid or elastomers, where the first is characterized by softening and fluidity with a substantial increase in temperature, they solidify with the removal of these variables, and can thus be recycled (CANEVAROLO JR, 2002). Thermosets are materials that present cross-links between the polymer chains after the curing process. They are network or lattice polymers and when subjected to increased temperature they soften and flow. However, unlike thermoplastics, after solidification the temperature variation no longer has an influence, making them infusible and non-recyclable (CANEVAROLO JR, 2002).

DISPERSED PHASE (REINFORCEMENT)

Some authors present different classifications for composite materials, where the morphology of the reinforcement phase can be used as a classification point (CALLISTER JR and RETHWISCH, 2016). Composites can be reinforced with particles, fibers, structural or nanocomposites.

For (CALLISTER JR and RETHWISCH, 2016), composites with the greatest technological value are those in which the dispersed phase is in the form of fibers. Fiber-reinforced composites can have a high modulus of elasticity and specific resistance, enabling various technological applications. A schematic representation for the classification of composites according to the morphology of the reinforcement phase can be seen in Figure3.

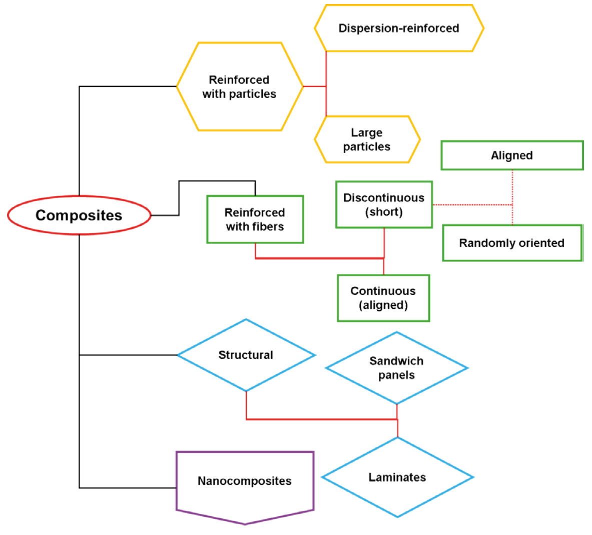

Figure 3: Classification of composite materials according to reinforcement geometry. Adapted with permission from Callister and Rethwisch (2016).

When looking at polymer matrix composites, it is clear that the most commonly used reinforcement fibers at a commercial level are carbon fibers, E-glass, aramid, ultra-high molecular weight polyethylene (NETO and PARDINI, 2016). Polymeric matrices, either thermoplastic or thermorigid, have low density (~1g/cm^3), as they are also much less resistant and rigid than fibers. This characteristic means that the mechanical properties exhibited by PMC are highly influenced by both the orientation and the volumetric fractions of the fibers present.

The arrangement or orientation of the fibrous fraction in composites, concentration and distribution have a significant influence on the properties presented by these materials (CALLISTER JR and RETHWISCH, 2016). Depending on the orientation, fibrous composites can have continuous or short fibers, which can be organized in a manner aligned with the longitudinal direction of the material or arranged randomly. Another way of manufacturing fibrous composites is the creation of fabrics, which have two-dimensional orientation of intertwined fibers throughout the composite. The schematic representation of the possibilities for organizing fibers throughout the composites is presented in Figure 4.

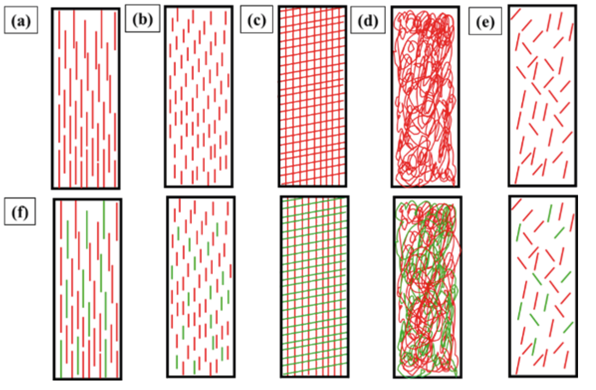

Figure 4: Schematic representation of fiber-reinforced composites. (a) continuous and aligned, (b) discontinuous and aligned, (c) fabric, (d) blanket, (e) random discontinuous and (f) hybrid composites. Adapted with permission from Callister and Rethwisch (2016), Neto and Pardini (2016).

Composites reinforced by longitudinal fibers Figure 4(a), bidirectional fabrics Figure 4(c) and longitudinal hybrids or fabrics Figure 4(f), tend to be much more structurally efficient in relation to composites obtained by discontinuous fibers Figure 4(b) and Figure 4(e) as well as blankets in Figure 4(d). Therefore, mechanical strength and longitudinal stiffness would have higher values compared to other cases. In transverse constraints, the best performance would be observed for fabric composites Figure 4(c), or hybrid fabrics Figure 4(f). These trends demonstrate that the orientation of the fibers in relation to the applied efforts, considering whether they are continuous or not, significantly influences the mechanical properties of the composites (NETO and PARDINI, 2016; CALLISTER JR and RETHWISCH, 2016).

The effectiveness of fibrous reinforcement is linked to the interface, requiring the fiber to have a certain critical length (L_c), which is directly dependent on the diameter of the fiber and the tensile strength presented by it. The relationship between L_c and the determination parameters are represented in Equation 1 (CALLISTER JR and RETHWISCH, 2016).

$$L_c = \frac{\sigma_f . d}{2\tau_i} \quad (1)$$

Where:

L_c: Critical fiber length;

σ_f: Fiber tensile strength limit;

d – Fiber diameter;

τ – Matrix shear yield stress.

Fiber-reinforced composites, as shown in Figure 4, are classified according to the length of the fiber used, thus continuous fibers are normally distributed aligned in the longitudinal direction of the composite, while discontinuous fibers may or may not have this alignment. Mechanical properties such as modulus of elasticity and longitudinal strength of composites reinforced with continuous and aligned fibers can be predicted according to the rule of mixtures according to Equation 2 and Equation 3, respectively (CALLISTER JR and RETHWISCH, 2016).

$$E_{Comp} = E_m . V_m + E_f . V_f \quad (2)$$

$$\sigma^*_{cl} = \sigma'_m (1 - V_f) + \sigma^*_f V_f \quad (3)$$

Where:

E_{Comp}: Longitudinal modulus of elasticity of the composite;

E_m: Modulus of elasticity of the matrix;

E_f: Modulus of elasticity of the fiber;

V_m: Matrix volume fraction;

V_f: Fiber volume fraction;

σ^*_{cl}: Longitudinal tensile strength;

σ'_m: Stress in the matrix at the point of failure;

σ^*_f: Fiber tensile strength limit.

Fiber-reinforced polymer resins tend to present the resulting properties depending on the intrinsic characteristics of the matrix and reinforcement phases (ABMACO, 2008). The representation of the stress-strain behavior for a brittle fiber and matrix with ductile behavior, together with the behavior of the composite subjected to uniaxial tensile, can be seen in Figure 5 (CALLISTER JR and RETHWISCH, 2016).

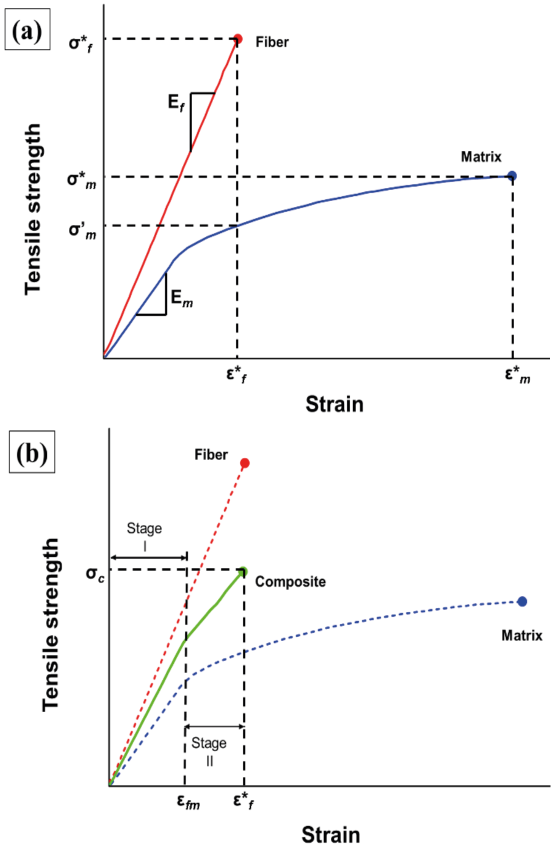

Figure 5: Schematic stress-strain curves for (a) brittle fiber and ductile matrix in (b) fibrous composite. Adapted with permission from Callister and Rethwisch (2016).

The structural performance of polymer composites reinforced with synthetic fibers is in many aspects superior to natural composites (NETO and PARDINI, 2016). There is a high demand for environmentally sustainable materials, with an extensive search to minimize the costs of conventional fibers (carbon, aramid, glass and steel), this interest directs research into the production of CRNFs (AHMED et al., 2021). Natural fibers are gaining more attention in the composites industries due to their specific properties and environmental benefits (KANDEMIR et al., 2020). Despite their low resistance properties, natural fibers partially replace synthetic fibers in structural applications due to their improved characteristics, such as specific modulus and elongation at break. Such information can be confirmed as presented (Furqan et al., 2015) by highlighting tensile modulus in relation to the volumetric cost of natural and synthetic fibers. The data obtained is illustrated in Figure 6.

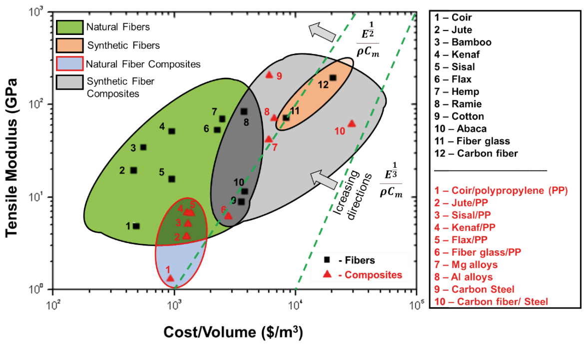

Figure 6: Tensile modulus versus cost per volume of natural and synthetic fibers. Adapted with permission from Furqan et al. (2015).

NATURAL FIBERS

Natural fibers are materials widely known across different industrial sectors, all this interest is partly due to their versatility and availability, as well as the concept of sustainability added to them (ROSLAN et al., 2021). The fashion, clothing and technical applications sectors are largely responsible for the high use of fibers, as they are very versatile materials (AWAIS et al., 2021).

Notably, there is a variety of materials with fibrous characteristics, however, the fibers themselves can be classified into two types, synthetic and natural. In recent decades, both synthetic and natural fibers have been widely used to manufacture high-quality materials, which exhibit exceptional properties (AWAIS et al., 2021). Fibers have a varied classification, so to work with these materials you must, in principle, know their main groups, therefore, Figure 7 highlights the different types of fibers.

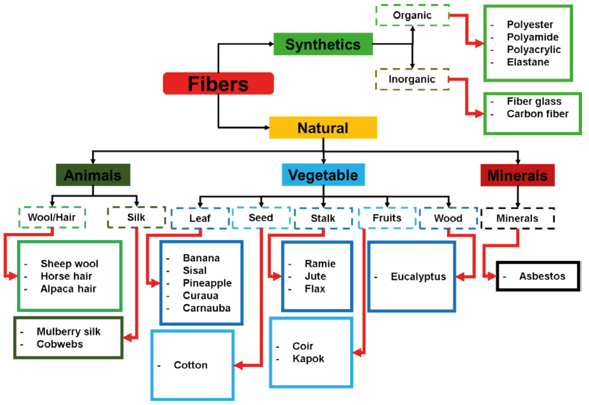

Figure 7: Classification of fibers. Adapted with permission from Awais et al. (2021).

When it comes to natural fibers, they can still be categorized according to their origins, such as animal, vegetable or mineral fibers. Fibers that are obtained from hair or secreted by animals are known as animal fibers, they are generally protein based (wool and silk) (NARAYANA and RAO, 2021).

Among natural fibers, those that have attracted the greatest technological interest from industries are natural fibers that is, obtained from plants (AHMED et al., 2021). The increasing employability of these materials in various sectors is partly due to their origin, as these fibers are generally obtained from agribusiness waste that would generally be destined for disposal (ZAMRI et al., 2021). Thus, agricultural waste is becoming a major problem for the environment, as large volumes of materials can be generated that will be discarded (KAMDA et al., 2021). These residues can originate from different stages, such as before or after cultivation, during storage, during or after consumption (KWOCZYNSKI e ČMELÍK, 2021).

The increase in the use of natural fibers can be correlated to their great use in the manufacture of composite materials, where their availability combined with abundant advantages becomes an imperative factor for their use (ASYRAF et al., 2021). As presented by (LOTFI et al., 2019), this high employability of lignocellulosic fibers can be understood through the evaluation of the annual production of these materials, the data presented by the authors were interpreted and presented in Figure 8. Thus, Figure 8(a) represents the annual production of NLFs in tons and Figure 8(b) shows the respective percentages of production and the countries responsible for the fraction of fiber produced.

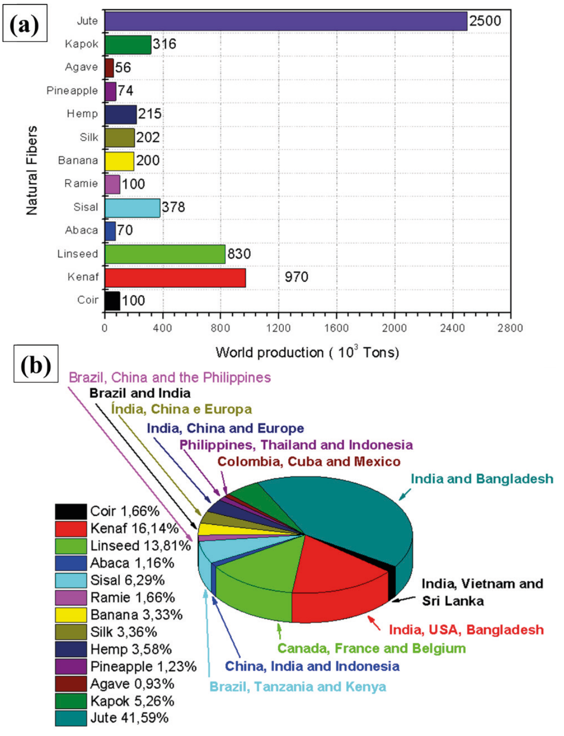

Figure 8: Annual production of the most commercially important natural fibers (a) and (b) the respective countries where they are produced. Adapted with permission from Lotfi et al. (2019).

Plant fibers can be obtained from various parts of plants, which act as a "skeleton" for plants, providing rigidity in their structure. The microfibrils present in the fiber are bound together by other natural substances, such as cellulose, lignin, hemicellulose, pectin, waxes and gums (AWAIS et al., 2021).

Natural fibers are materials that have a complex constitution, in their most basic constitution they present a matrix of hemicellulose and lignin, which is responsible for involving semicrystalline cellulose fibrils (JOHN and THOMAS, 2008). The morphological complexity of natural fiber is presented in Figure 9(a), taking as an example the cross section of a flax fiber in Figure 9(b).

The top layer is the bark or primary wall, which is primarily responsible for protecting the plant against moisture evaporation and unexpected changes in temperature (KARIMAH et al., 2021). In the middle layer where the fibrils are located in the phloem, it appears as a bundle. The bottom layer contains xylem, broad beans and woody core which plays a significant role in transferring water and nutrients from the center of the fiber (AHMED et al., 2021). The simple fibers are connected by the middle lamella, a substance composed of pectin, which works like a glue (HASAN, RABBI e BILLAH, 2022).

Fibers are made up of two significant walls, with a primary and secondary wall; These walls surround a protein- and pectin-filled channel called the lumen (KARIMAH et al., 2021). The primary wall has a rigid structure of cellulose microfibrils within a network of hemicellulose, pectin and glycoprotein compounds. Whereas the secondary wall is larger with a three-layer structure (S1, S2, S3). All these layers consist of cellulose, hemicellulose and lignin. The middle layer (S2) forms the maximum fiber volume (about 70-80% of the mass) (AWAIS et al., 2021). Thus, the highest attribute of a single fiber is controlled by the S2 layer which determines the overall mechanical strength (AHMED et al., 2021).

Natural fibers as a whole present a great variability in their properties, several factors are responsible for this variation, among which the following stand out: chemical compositions, species, extraction techniques, geographic factors, growth conditions, age of the plant and regions from which they were extracted (MAZLAN et al., 2021).

Quantifying the lignocellulosic constituents present in the fibers is extremely important, as they are directly responsible for the variation in the thermal and mechanical properties presented by these materials (JUNIO et al., 2022). The selection of fibers depends mainly on the physical and mechanical properties inherent to them. A significant variation in the properties of plant fibers was observed among the published literature, due to the influence of several factors from production to the processing stage (AWAIS, NAWAB, et al., 2021). Table 2 presents the variation in lignocellulosic constituents of some natural fiber evaluated by different authors.

Various natural fibers are harvested from crop residues, such as pineapple leaves, sugar cane fibers, coir sheaths and kenaf stems. A infinity of cellulosic fibers such as kenaf (SILVA et al., 2021), hemp (RIBEIRO et al., 2021), seven-islands-sedge (NEUBA et al., 2020), titica vine (CUNHA et al., 2021) and carnauba (JUNIO et al., 2020) they have been widely used for various engineering applications, especially the manufacture of composites.

Characteristics such as the lack of uniformity, dimensional limitations and constitutional heterogeneity can be correlated with the difficulty of applying natural fiber in relation to synthetic ones in industrial sectors. One of the main disadvantages of natural fibers in relation to their competitors lies in their hydrophilic properties, which increase their moisture absorption rate (ASYRAF et al., 2021). This factor combined with the manufacture of polymer matrix composites reduces wettability and interfacial interaction between the reinforcement phase and the polymer. This directly leads to non-uniformity in the final material, where a lack of thermal stability and limited mechanical resistance are generally observed in relation to composites

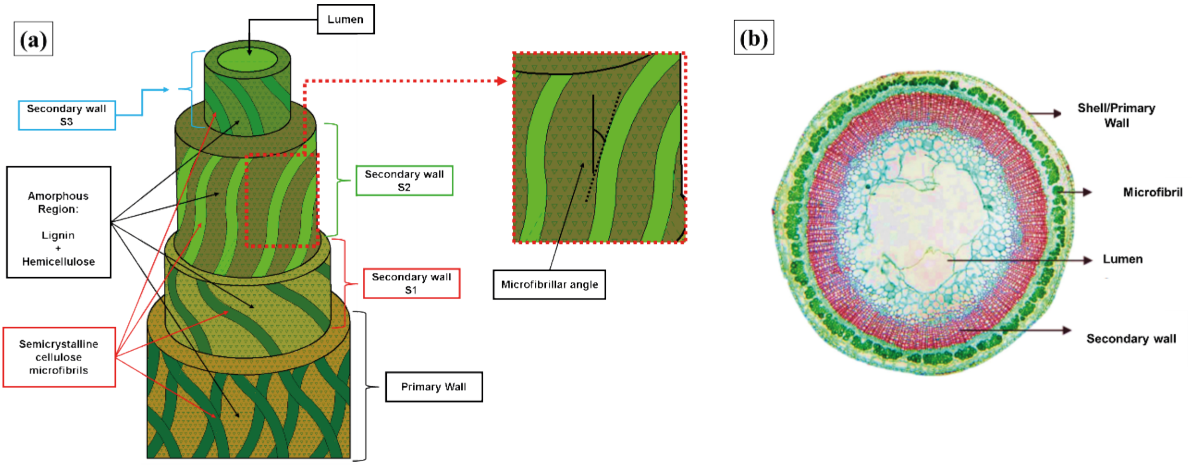

Figure 9: Schematic representation of a natural fiber (a) and (b) cross section of a flax fiber. Adapted with permission from John and Thomas (2008), Ahmed et al. (2021).

NLF	Cel. (%)	Hem. (%)	Lig. (%)	Ashes (%)	Moisture (%)	References
Carnauba	20.1	40.9	36.9	2.2	7.2	(JUNIO et al., 2022)
Bagasse	55.2	16.8	24.3	2.2	8.8	(GIRIJAPPA et al., 2019)
Hemp	68.0	15.0	10.0	0.80	9.1	
Flax	71.0	18.6	2.2	-	10.0	
Coir	36.5	15.0	43.0	-	-	(MOCHANE et al., 2019)
Jute	67.0	16.5	14.5	-	8.0	(GHOLAMPOUR e OZBAKKALOGLU, 2020)

Table 2: Chemical composition of some natural fiber evaluated in the literature.

Fiber	Density (Kg/m³)	Diameter (μm)	Tensile strength (MPa)	Modulus of elasticity (GPa)	Stretching (%)
Flax	1500	75	345-1035	50-70	2.7-3.2
Ramie	1500	34	560	24.5	2.5
Nettle	1500	50	650	38	1.7
Glass	2500	-	2000-3500	70	2.5
Carbon	1700	-	4000	230-240	1.4-1.8
Aramid	1400	-	3000-3150	63-67	3.3-3.7

Table 3: Properties of some natural and synthetic fibers. Adapted with permission from Mudoi et al. (2021)

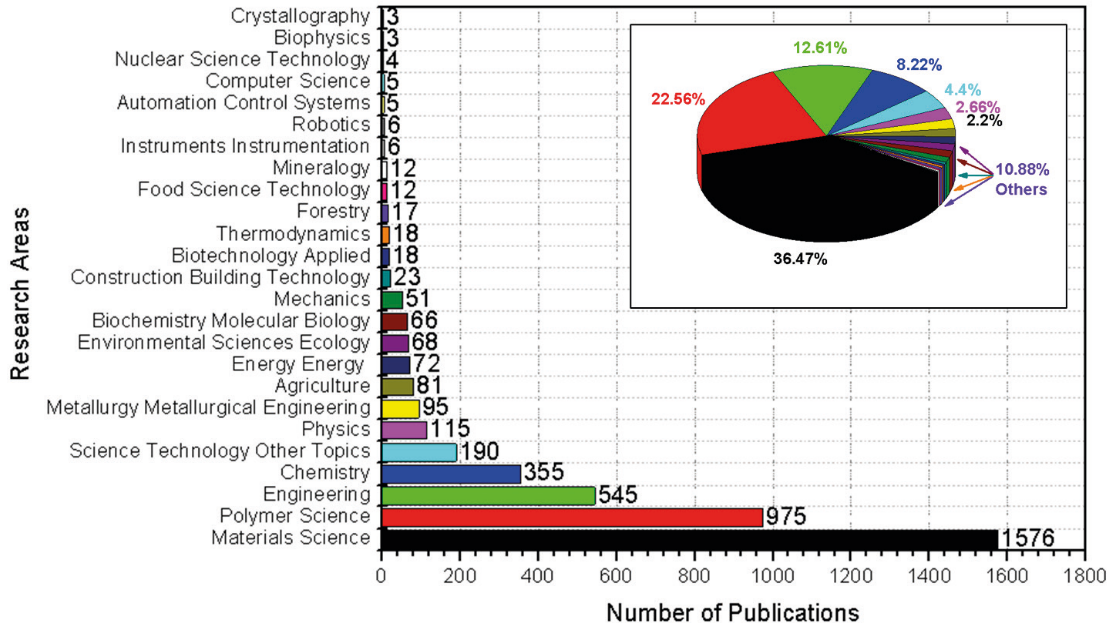

Figure 10: Works published in their respective areas of interest between 2003 to 2023. Adapted from Web of Science (2023).

A	B	C	D	E	F	References
Banana/Epoxy	CM	30.4	-	56.3	2.7 J/mm^2	(BALAJI et al., 2020)
Banana/Epoxy	HL	16.4	0.65	57.33	2.3 J/m	(VENKATARAJAN e ATHIJAYAMANI, 2021)
PALF/Epoxy	HL	22.0	0.57	31.0	-	(KUMAR et al., 2020)
Sisal/Epoxy	RTM	116.2	3.4	112.0	19.0 (kJ/m^2)	(SAHOO, MOHANTY e NAYAK, 2015)
Sisal/Epoxy	HP	380	5.6	325	-	(PAPPU, PICKERING e THAKUR, 2019)

A – Composite
B – Processing
C – Tensile strength (MPa)
D – Modulus of elasticity (GPa)
E – Flexural resistance (MPa)
F – Impact resistance

CM – Compression molding
HA – Hand Lay-up
RTM – Rotomolding
HP – Hot pressing

Table 4: Different methodologies for manufacturing composites reinforced with different natural fibers and their respective properties.

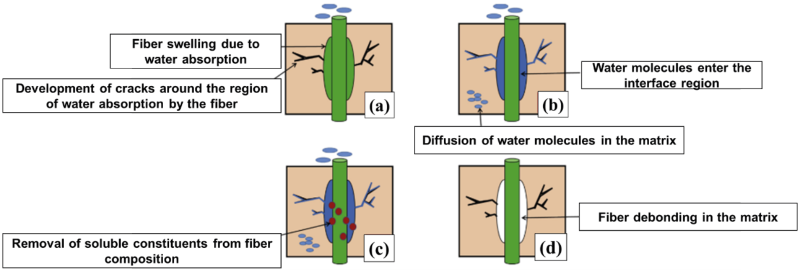

Figure 14: Effect of water absorption in CRNFs. Adapted with permission from Haameem et al. (2016).

made with synthetic fibers (ILYAS et al., 2021).

In order to improve the properties presented by natural fibers, several surface treatments are applied. Physical and chemical techniques are introduced to seek to solve, at least in part, the problem of fiber and matrix compatibility, thus promoting the manufacture of effective composites. These media provide the treated fibers with greater surface roughness, which would allow for additional activated sites in the fiber topography (VU et al., 2017). The comparison between some mechanical properties presented by natural fibers in relation to synthetic fibers can be seen in Table 3.

Synthetic fibers are generally stronger than natural fibers. The quality of synthetic fibers and fabrics can also be easily converted as needed by modifying the chemical properties and circumstances in which they are manufactured (CESA, TURRA and BARUQUE-RAMOS, 2017). Depending on their properties, they can be elastic, flexible, rigid or have a lesser or greater water absorption. When compared to natural fibers, synthetic fibers are typically more resistant to water, stains, heat, and chemical damage (AHMAD e ZHOU, 2022). In general, natural fibers are more susceptible to chemical decomposition than synthetic fibers because they are biodegradable and can be caused by various bacteria and fungi. In contrast, most synthetic fibers are not biodegradable and wear out over time, while natural fibers are easily decomposable (CAPPITELLI, VILLA and SANMARTÍN, 2021).

The drawbacks inherent to natural fibers are largely outweighed by advantages such as: low cost, greater flexibility, low specific mass, non-abrasiveness and possibility of social and economic development (JUNIO et al., 2020). Because they are biodegradable and come from renewable resources, they qualify as potential sources of income for developing regions, especially for the poorest countries. In view of the above, there is a tendency to replace synthetic fibers with natural fibers, especially for the manufacture of composite materials.

STATE OF ART

There is an incessant search by the academic community to replace synthetic fibers such as aramid, glass fibers and carbon fibers with natural fibers in the manufacture of composites (MUDOI, SINHA and PARTHASARTHY, 2021). Composites made using natural fiber as a reinforcing phase tend to exhibit better mechanical properties, such as lower density, non-abrasiveness, lower cost, high impact resistance, strong damping capacity, easy availability and recyclability (DEVNANI and SINHA, 2020). The use of natural fiber reinforced composites is growing in structural and non-structural, automotive, marine, wind, aerospace and biomedical device applications (SELDON e ABILASH, 2021).

All the benefits presented through the manufacture of composites with natural fibers further strengthen the interest of the academic community in investigating the properties presented by these materials. The growing need to evaluate the characteristics presented by CRNFs is explained by the gradual increase in the number of articles published in relation to this topic, mainly in the last 20 years (WEB OF SCIENCE, 2023). With the aim of evaluating the evolution of research aimed at the topic under study, a search was carried out in the Web of Science database (WEB OF SCIENCE, 2023). The following keywords were used for the search: Natural Fibers; Composites; Biocomposites, therefore, published results between January 2003 to November 2023 were evaluated. The results obtained for the number of works published in each area of interest are presented in Figure

10.

Between January 2003 to November 2023, around 2807 works were published related to the keywords proposed in the search, as shown in Figure 10. The area of Materials Science had the highest publication rate, with 1576 articles published. The areas of Polymer Science (975) and Engineering (545) present the second and third highest number of publications, respectively. It is also possible to evaluate the evolution in the number of annual publications during the search interval, thus observing the number of works published in the current year and the accumulation of citations over the years. The data obtained can be seen in Figure 11.

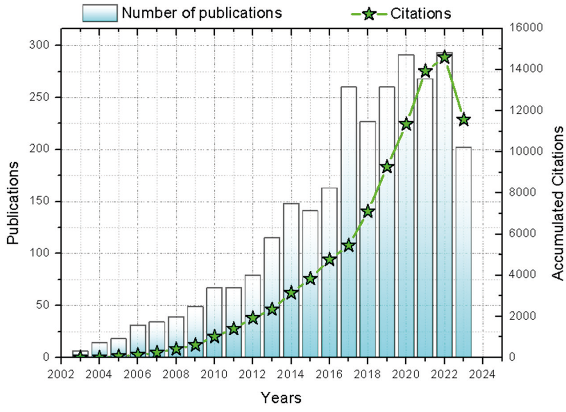

Figure 11: Number of works published annually and accumulated citations between 2003 and 2023. Adapted from Web of Science (2023).

When analyzing the data presented in Figure 11, it can be seen that until mid-2005 the number of works published annually did not exceed two dozen.

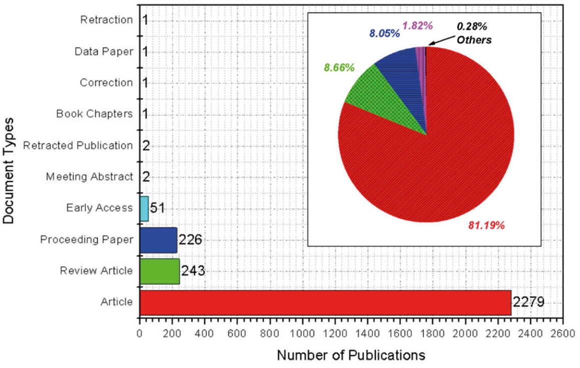

Figure 13: Types of works published between 2003 and 2023. Adapted from Web of Science (2023).

CRNFS PROCESSING

The correct selection of processing methods that fit the needs required for the manufacture of CRNFs becomes imperative for the manufacture of these materials. The methodology used to manufacture composite materials is an extremely important parameter that directly influences the final properties displayed by the finished products. The preliminary assessment for choosing the most suitable manufacturing process involves consideration of several key criteria, including shape, size and desired properties of the composites, as well as manufacturing cost, production speed and raw material properties (LOTFI et al., 2019).

The preliminary assessment to select the manufacturing technique is dominated by the size of the composites. For small and medium-sized components, simple and fast processing techniques such as compression and injection molding are used, while large components are manufactured by autoclave and open molding processes (PRAVEENA et al., 2020).

The applicability of different processing means for CRNFs was observed in the literature. The main manufacturing methods applied for the production of composites are represented in Table 4.

There are several forming techniques

for polymeric materials, however, when natural fibers are involved in processing, several points such as: temperature, shear rate and pressure must be taken into account (VIGNESHWARAN et al., 2020). Natural fibers are versatile materials, however, they are quite sensitive to temperature variations, and high compression or shear loads can lead these materials to sudden failure. Thus, the composite becomes brittle without it having been exposed to any mechanical or thermal stress, other than the parameters involved in processing.

Another extremely important parameter for CRNFs is the hydrophilic character of the fibers, as this characteristic directly affects the properties presented by the composite (VIGNESHWARAN et al., 2020). The presence of moisture in the fiber can develop a crack in the matrix due to poor interfacial bonding which can reduce the useful life of the composites by affecting the strength of the composite (GOWDA et al., 2018). The effect of diffusion of water molecules at the interface of polyester/napier grass fiber composite was studied by (HAAMEEM et al., 2016), the mechanisms addressed in the study are presented in Figure 14.

The moisture absorption means of polymer composites mainly follow three diffusion mechanisms. The first is the diffusion of water molecules due to the presence of micro gaps or voids in the polymer chain. The second is transport occurs through the spaces between the fiber and the matrix interface. The third mechanism involves the diffusion of water through microcracks on the surface of the composite (VIGNESHWARAN et al., 2020). (HAAMEEM et al., 2016) reported in their study that fiber treatment is effective in increasing the water resistance characteristics of the composite. The untreated short and long fiber composites absorbed about 13% and 20% of water, respectively, while the treated short and long fiber composites absorbed only 5% and 7%, respectively. Another similar example is presented by (WU et al., 2018), where hemp fiber composites were manufactured and coated with polyethylene film on the surface to study the water absorption property. The study reported that the polyethylene coating reduced the water absorption of the composites. Furthermore, they found that a higher rate of water absorption was observed along the edges of the composites.

Improving the interfacial bonds of fibrous composites can be achieved through surface treatments applied to the fibers. Surface modification of the fiber occurs with the action of chemical or physical treatments, and thus disadvantages such as the hydrophilicity of the fibers are overcome (MUDOI, SINHA and PARTHASARTHY, 2021). Various chemical treatments applied to nettle fibers and their respective effects are represented in Table 5.

Each treatment presents a different set of mechanisms of action, therefore, the final objective of all methods is to remove at least part of the OH groups, waxes, lignin, hemicellulose and pectin content. This factor provides an increase in surface roughness and thus obtains a strong matrix/fiber interaction in the composite (SATHISH et al., 2021).

Taking as an example the mercerization depicted in Table 5, where it acts to remove lignin, hemicellulose, wax and oils from the external surface, depolymerizes the cellulose, exposes the crystallites and fibrils, reduces the spiral angle and diameter and increases molecular orientation (VIJU and THILAGAVATHI, 2020 (b)). Reduction in fiber diameter is a common consequence of NaOH treatment, which has also been observed by other researchers (PRITHIVIRAJ and MURALIKANNAN, 2020). All these modifications lead to a better bond at the fiber-matrix interface, however, concentration,

Chemical T.	Effect	Ref.
Mercerization (NaOH)	The cellulose content of treated fibers (78.85% by weight) was increased compared to untreated fibers (72.12% by weight). The lignin, hemicellulose and pectin fractions were removed. The amount of water absorption was reduced (11.8%) compared to untreated fiber (12.3%). There was an improvement in thermal stability (260–340 °C) than untreated fibers. There was an increase in tensile strength (626.15 MPa) compared to untreated fiber (479.86 MPa). There was a reduction in the average diameter of the treated fiber (0.038 mm) to the untreated fiber (0.052 mm).	(VIJU and THILAGAVATHI, 2020 (a); VIJU and THILAGAVATHI, 2020 (b))
Silane	There was an increase in the flexural properties of the composite to 49.325 MPa. There was a reduction in fiber diameter after silane treatment.	(SUARSANA et al., 2021)
H_2O_2	The treatment increased the cellulose content and reduced the hemicellulose, lignin, wax and water absorption levels compared to untreated fibers. It provided better thermal stability in the temperature range (260–340 °C).	(VIJU and THILAGAVATHI, 2020 (a))
$NaClO_2$	It resulted in an increase in the cellulose fraction together with a decrease in the amount of lignin, hemicellulose and water absorption in relation to untreated fibers.	(VIJU and THILAGAVATHI, 2020 (b))

Table 5: Effect of chemical treatments on nettle fibers.

immersion time, treatment temperature are vital parameters to obtain an optimal interfacial adhesion efficiency.

With the search for better properties to be presented by CRNFs, the surface modification of the fiber or addition of fillers has become a viable point. In this regard, the insertion of graphene (G), graphene oxide (GO) or reduced graphene oxide (rGO) in the manufacture of high-performance composites stands out (LUZ et al., 2020 (a)).

The first strategy was to use GO as a filler in polymer matrix composites associated with some fiber reinforcement. The successful development of these hybrid composite structures lies in the synergy between the matrix and reinforcements, as well as the optimized composition of the constituents (LUZ et al., 2020 (a)). In a study carried out by (PRASOB et al., 2019), the authors investigated the mechanical behavior of a hybrid composite using jute fiber and rGO filler reinforcing epoxy matrix. Figure 15 presents the results for different mechanical properties measured at three different temperatures.

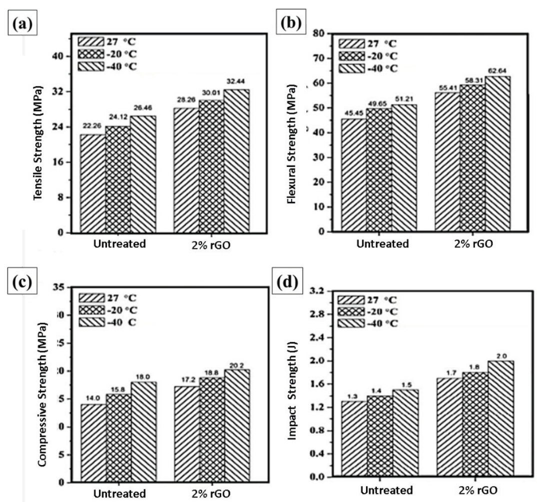

Figure 15: Properties of jute/epoxy composites incorporated and not incorporated with rGO: (a) tensile strength, (b) flexural resistance, (c) compression and (d) Izod impact resistance. Adapted with permission from Prasob et al. (2019).

It is noted that with the use of GO and its derivatives, there is an increase in the mechanical properties of polymer matrix CRNFs. However, the increase achieved can be considered low and in many aspects the amount of GO can negatively impact the properties of the composite. In fact, the amount of filler is something that must be carefully addressed, as additions on the order of 1% by weight can lead to agglomeration of nanofillers (LUZ et al., 2020 (a)).

In fact, it is observed that parameters adopted for the processing of CRNFs, whether for surface treatments of the fibers, manufacturing methodology or addition of fillers, are parameters of essential importance, and directly influence the final properties presented by these materials.

PROPERTIES OF CRNFs

The properties of CRNFs are directly affected by the fiber-matrix interface region, so the efforts transmitted to the material are shared between the fiber and the matrix (HUANG et al., 2021). The load transfer mechanisms in unidirectional fibrous composites are represented in Figure 16, as highlighted (HUANG et al., 2021).

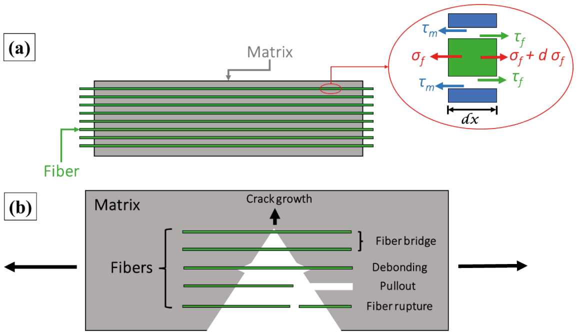

Figure 16: Schematic view of load transfer mechanisms at the fiber-matrix interface in composites reinforced with unidirectional fibers (a) and (b) failure mechanisms occurring in the composites. Adapted with permission from Huang et al. (2021).

When dealing with CRNFs, the vast majority of fibers have greater stiffness and resistance compared to the matrix used, thus, the simple addition of fibers leads to an improvement in the mechanical properties of the matrix, such as the modulus of elasticity or tensile strength. Among the functions of fibers, the most important are filling cracks in the matrix, dissipating energy when subjected to mechanical stress, carrying most of the applied load (tensile strength or compression) and thus, mitigate the growth of cracks internal to the matrix (HUANG et al., 2021).

As shown in Figure 16(a), when subjecting a fibrous composite to axial stress, tension is transferred to the fiber through the matrix that surrounds it. In the image, $\boldsymbol{\tau}_m$ and $\boldsymbol{\tau}_f$ are the shear stresses that act on the matrix and fibers, respectively. The $\boldsymbol{\sigma}_f$ and $d\sigma_f$ are the tensile stresses observed in a fiber element with length dx, respectively. The mechanical process initiates the deformation of the matrix, then the deformation of the matrix induces shear stress at the fiber-matrix interface. The tensile stress of the fiber is activated to balance the shear stress. Through this process, the fiber supports most of the tensile load applied to the composite (SETHI and RAY, 2015).

The fiber failure mechanisms are illustrated in Figure 16(b). Microcracks initially occur in the polymer matrix due to its lower fracture stress. A crack propagates when the strain energy near the crack tip is greater than the energy to form a new surface. If the fibers are filling a crack in the matrix, the stress carried by the matrix is transferred to the fiber across the interface. As tension increases, interface failure (e.g., fiber pullout and interface debonding) or fiber fracture will occur depending on the interfacial shear strength and tensile strength of the fibers. Interface failure and fiber fracture dissipate strain energy near the crack tip, leading to mitigation of crack propagation (HUANG et al., 2021).

The phenomena observed in the fiber-matrix interface region can be directly related to the properties presented by CRNFs. In this regard, several authors dedicate efforts to investigating characteristics related to the interfacial interaction between the fiber and matrix (JUNIO et al., 2020). So-called pullout tests are generally applied to evaluate the maximum pullout tension, critical fiber length and interfacial bond strength, parameters that are directly related to the mechanical resistance presented by the composites (LUZ et al., 2018). Figure 17 shows the pullout tests performed on different natural fibers in epoxy matrix composites.

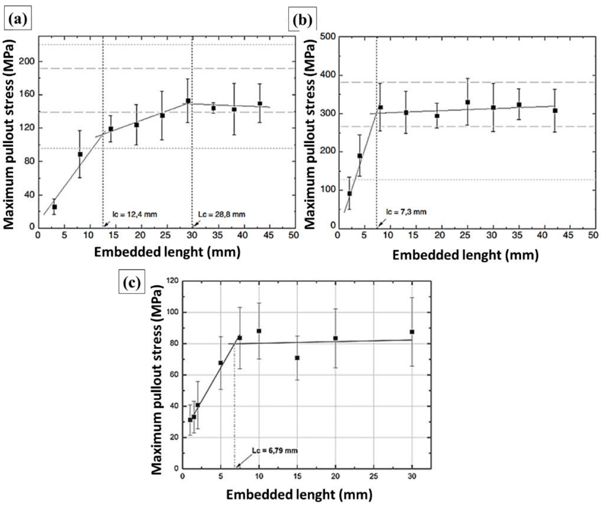

Figure 17: Results obtained for pullout tests. (a) coir fiber, (b) pineapple leaf fiber (PALF) and (c) carnauba fibers in epoxy matrix. Adapted with permission from Luz et al. (2018) and Junio et al. (2020).

Figure 17 (a) and (b) (LUZ et al., 2018) present the results obtained for the pullout test on epoxy matrix composites reinforced with coir fibers and PALF epoxy, respectively, where were the interfacial bond strength values of 1.42 MPa for the first case and 4.93 MPa for the second. The authors highlighted that the PALF/Epoxy interfacial adhesion was stronger than coir fibers/Epoxy, stressing that PALF can more efficiently transfer the mechanical strength to the composite matrix. The result can then be attributed to the rougher surface presented by the pineapple leaf fibers, where a surface morphology with greater irregularity allows greater penetration and more efficient anchoring of the epoxy matrix (LUZ et al., 2018). In Figure 17(c) (JUNIO et al., 2020), they present the results obtained in the same type of test for carnauba/Epoxy composites, where the interfacial bond strength was 3.95 MPa. The value obtained was considered relatively low, this fact was combined with the hydrophilic nature of carnauba fibers, furthermore, this type of fiber has a layer of wax (fatty acid) on its surface, which can act to impair adhesion in the matrix (JUNIO et al., 2020). However, the value obtained was relatively higher than the epoxy-coir fiber interaction (1.42 MPa) and very close to the epoxy-pineapple leaf fiber interaction (4.93 MPa).

Studies aimed at evaluating the surface roughness of natural fibers are important for verifying the mechanical properties of CRNFs. One option to investigate the variation in the surface morphology of natural fibers is atomic force microscopy (AFM), therefore, several authors use this technique to correlate the surface roughness presented by the fibers that will be used in CRNFs. Figure 18 presents AFM results obtained for some natural fibers in different studies.

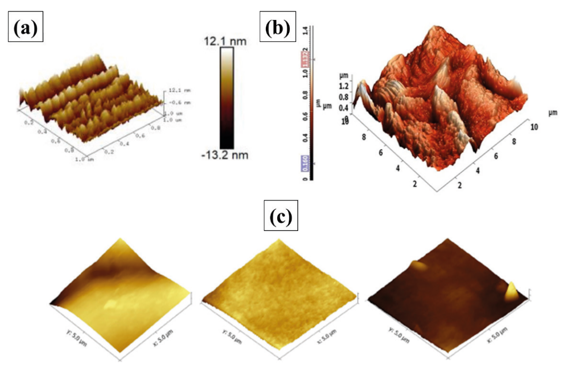

Figure 18: AFM images for different natural fibers. (a) PALF in natura, (b) Derris scandens and (c) Rubber tree treated by acid hydrolysis. Adapted with permission from Najeeb et al. (2020); Onkarappa et al. (2020) and Perumal et al. (2020).

In general, a rougher fiber surface is an important indicator to increase the compatibility of the matrix with the fiber (NAJEEB et al., 2020). Composites reinforced with surface treated fiber showed greater strength compared to untreated fiber (ONKARAPPA et al., 2020; PERUMAL and SARALA, 2020).

MECHANICAL PROPERTIES OF CRNFS

There are a variety of approaches that can be used to improve the mechanical properties presented by CRNFs. They are generally not rigid formulas that, when implemented, will result in substantial improvements; rather, improving the mechanical and physical properties of natural fibers is an active topic that is still being researched and developed (AHMAD and ZHOU, 2022). Better mechanical performance in one area of application may not equate to the same performance in another area of use and vice versa. For example, increasing the stiffness of a material may be beneficial in the construction industry, but may not be necessary or valuable in biological applications, which may prefer improvements in flexibility or toughness.

Regarding the mechanical properties presented by CRNFs, Figure 19 presents the tensile strength, flexural strength and impact resistance of polyester matrix composites reinforced with different natural fibers.

When observing Figure 19(a), it is noted that for the same matrix different tensile strength values are presented, this characteristic is related to all the parameters already discussed, such as the fiber resistance itself, interaction between the fiber/matrix, surface treatments applied, among other characteristics. It is very clear in the literature that the tensile property of CRNFs is directly dependent on the interfacial bond strength between the matrix and the fiber. Improving this property can maximize the mechanical strength of composites (VIGNESHWARAN et al., 2020). As each natural fiber has characteristics intrinsic to its constitution, they present better or worse interaction with the matrix, thus resulting in a greater or lesser value of the property.

Figure 19(b) presents the flexural resistance of the composites, directly observing that the value of the property will not be the same as for the tensile strength, thus not following the same trend. Fiber properties are the most influential parameters on bending, therefore, when fiber reinforcement is stronger, tensile strength is greater than flexural resistance. In the case where the fiber is brittle and breaks easily, the flexural resistance is greater than the tensile strength (VIGNESHWARAN et al., 2020).

Regarding the impact resistance (Figure 19(c)) of a material which is the amount of energy absorbed by the material under sudden load across its area. Composite materials are subjected to different types of impact stress during their useful life. Therefore, the behavior of composites under various loads needs to be understood. Therefore, depending on the speed, the impact is generally classified as low impact (<10 m/s), medium (50-200 m/s) and high speed (200-5000 m/s) (FATIMA, BACHIR and FABIENNE, 2019).

In general, the impact resistance of pure polymers is extremely low due to their brittle property. The introduction of fiber into polymers offers greater tenacity to composites, which is favorable for increasing impact resistance (VIGNESHWARAN et al., 2020). (ELANCHEZHIAN et al., 2018) reported that the impact resistance of fibrous composites is a factor influenced by the fiber-matrix bond. In this research, it was found that impact resistance increases with increasing fiber fraction due to the favorable adhesion between the fiber and the matrix.

Natural fibers in general provide different variations in the final properties of

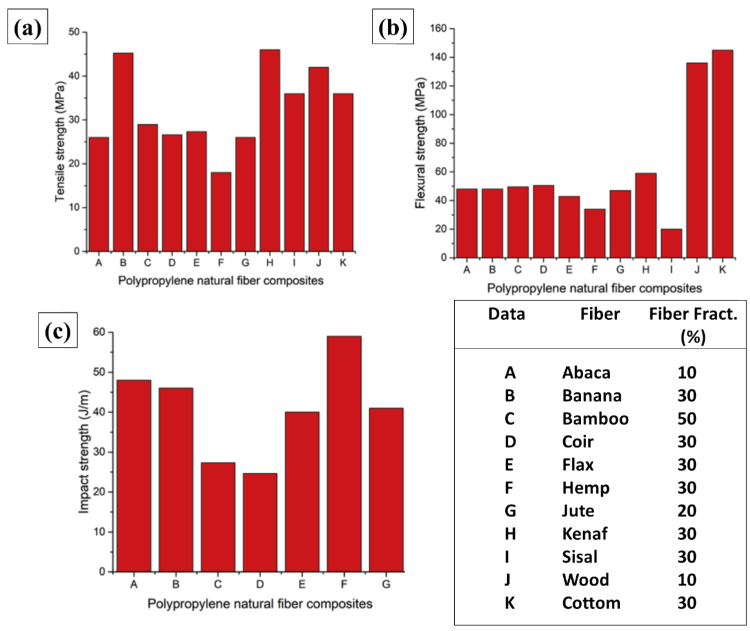

Figure 19: Mechanical properties of polyester matrix composites reinforced with different natural fibers. (a) tensile strength, (b) flexural resistance and (c) impact resistance. Adapted with permission from Vigneshwaran et al. (2020).

Fibers	Matrix	Properties	Improvement (%)	Ref.
Jute	Polypropylene	TS	150	(LIEW et al., 2020)
Jute	Polypropylene	ME	330	
Banana	Polyester	TS	100	(HOSSAIN et al., 2020)
Banana	Polyester	ME	53	
Banana	Polyester	FR	75	
Banana	Polyester	FM	55	
Jute and Sisal	Epoxy	TS	70	(CAVALCANTI et al., 2019)
Jute and Sisal	Epoxy	ME	75	

TS – Tensile strength
ME – Modulus of elasticity
FR – Flexural resistance
FM – Flexural modulus
Ref – References

Table 6: Effect of natural fibers on different types of matrices.

Fibers	Matrices	Applications	Ref.
Curaua, jute, sisal, ramie	Cement	Cementitious materials	(FERREIRA et al., 2018)
Jute	Hybrid + glass fibers	Vessel panel	(GOPINATH, POOPATHI e SARAVANAKUMAR, 2019)
Kenaf	Plater	Ceiling	(AKUBUEZE et al., 2019)
Hemp	Epoxy	Furniture	(DAHY, 2019)
Curaua	Epoxy + GO	Ballistic protection	(COSTA et al., 2019)
Jute, sisal, coir, banana	Epoxy	Helmet	(BHARATH et al., 2018)
Ramie	Kevlar + epoxy	Ballistic protection vests	(BRAGA et al., 2018)
Flax	Carbon nanotube	Electrodes	(ZHANG et al., 2017)
Cotton	Epoxy	Medical applications	(MORRIS et al., 2020)

Table 7: Applications of natural fiber composites in various areas.

composites, several authors evaluate different results of mechanical properties obtained for different CRNFs, some results observed in the literature are presented in Table 6.

As seen in Table 6, fiber-based residues that are added as reinforcing agents to the polymeric matrix act by providing an improvement in mechanical properties, such as: tensile strength, tensile modulus, flexural resistance, flexural modulus, elongation at break, impact resistance, compressive strength and toughness (AKTER and HOSSAIN, 2021).

THERMAL PROPERTIES OF CRNFs

The thermal properties of CRNFs, especially those made from polymer matrices, are highly influenced by the characteristics of the natural fiber. Characterization techniques such as thermogravimetric analysis (TGA), derived thermogravimetry (DTG), differential scanning calorimetry (DSC), and dynamic mechanical analysis (DMA) are some notable methods to evaluate the thermal behavior of CRNFs. Thermal degradation of CNF is categorized based on five main characteristics, i.e., weight loss of the composite, decomposition of fiber composition such as hemicellulose, cellulose and lignin, and depolymerization (MONTEIRO et al., 2012). The temperatures occurring during the manufacturing steps of CRNFs, such as curing temperature for thermosetting polymers and extrusion temperature for thermoplastic polymers, have a significant impact on the thermal degradation of the fiber and matrix. For these reasons, CNF processing normally involves temperatures below 200 °C, a range in which a large part of the natural fibers are degraded (VIGNESHWARAN et al., 2020).

Composites reinforced with natural fibers exhibit mass loss in a very characteristic way, with each constituent present in the matrix or reinforcement phase presenting a well-established degradation range. In the case of epoxy polymer matrix composites, it is thermally stable up to approximately 280 °C, with a negligible mass loss of 1.05% as presented by (NASCIMENTO et al., 2019). In the case of natural fibers, the thermal decomposition of some constituents such as cellulose (240–350 °C), hemicellulose (200–260 °C) and lignin (280–500 °C) degrade at these temperatures (JUNIO et al., 2022). The temperature ranges where degradation of the constituents present in the composites occurs is illustrated in Figure 20.

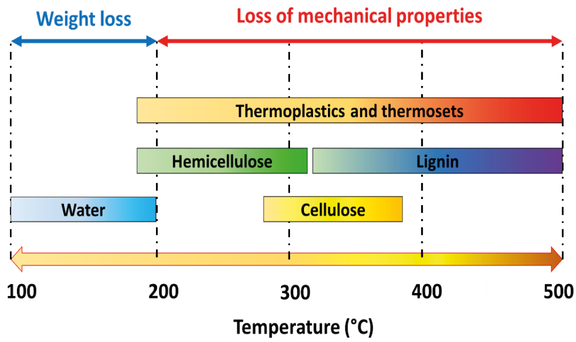

Figure 20: Degradation stages of CRNFs constituents. Adapted with permission from Vigneshwaran et al. (2020).

In studies carried out on epoxy matrix composites reinforced with Kenaf fibers portrayed by (SILVA et al., 2021), composites have a maximum working temperature of 200 °C. On the other hand, the addition of fibers to the epoxy matrix results in thermal stabilization of the composites. The authors raised this information due to the higher residue content observed at 900 °C. Thus, kenaf fibers provide better thermal stability to the composite, where the better interaction between the fiber and the matrix delays the total degradation of the composite at higher temperatures.

(RIBEIRO et al., 2021) evaluated the thermal behavior of epoxy-hemp fabric composites through DSC. The authors observed that the increase in the volumetric fraction of the hemp fabric in the epoxy matrix results in a shift in the endothermic peak that varies between 68.1 and 70.8 °C. This change may be contributed by the release of moisture inherent to the hydrophilic fibers at the glass transition temperature (T_g) of the epoxy resin. Thus, the T_g of the composites increases as the volumetric fraction of reinforcement increases.

(JUNIO et al., 2020) confirmed the reinforcement effect caused to epoxy-carnauba fiber composites through data obtained by DMA. The authors presented an improvement in the storage modulus with the incorporation of carnauba fibers, thus causing a better interaction between the fiber and the matrix, providing effective reinforcement to the composites in relation to the pure epoxy matrix. (JUNIO et al., 2020) also reported that the increase in the fraction of fibers in the composite generates a shift of the loss modulus peaks to higher temperatures compared to pure epoxy.

APPLICATIONS FOR CRNFs

Composites reinforced with natural fibers are often considered "green" or "sustainable" materials, partly due to the materials used in their manufacture, as well as their recyclability. Recycling CRNFs is becoming one of the most important points for these materials, so future processing methods will be designed to minimize and eventually remove waste from the process (MOAZZEM et al., 2021). In the future, it is expected that composite materials will be reused and/or repurposed, rather than relegated to landfills, Figure 21 highlights future flowcharts for recycling CRNFs.

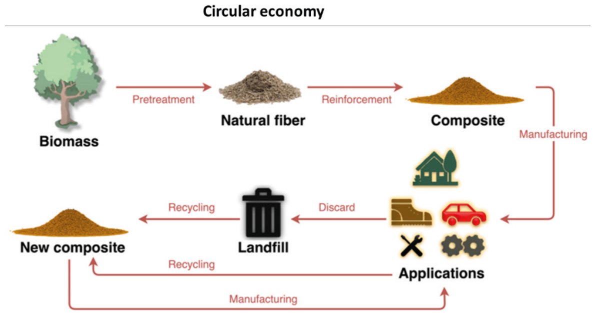

Figure 21: Future CRNFs recycling flowchart. Adapted with permission from Zhao et al. (2022).

As presented (ZHAO et al., 2022), in the automobile industry, cars at the end of their useful life are often sent to a shredder. During this process, polymeric materials must be separated from metal and glass. This is certainly possible, but the resulting polymer mixtures often cannot be recycled without additional separation steps.

When it comes to the automobile industry, some automakers highlight the use of CRNFs in their production lines. In 2019, Porsche announced that the 718 Cayman GT4 Clubsport race car was the world's first vehicle with a body constructed from hemp/flax reinforced composites (LI et al., 2020). These "green" composites derived from natural fibers are increasingly applied to automotive parts by manufacturers and suppliers, such as door panels, seat backs, headliners, trays, dashboards and interior parts.

For Mercedes-Benz, natural fibers have been widely applied in components of various models, as shown in Figure 22 (AKAMPUMUZA et al., 2017). Thus, for example, 20.8 kg of natural fibers (wood, banana and flax) are used in more than 20 components for the class A model; approximately 19.8 kg of natural fibers (wood, coconut and honeycomb cardboard) in more than 21 components for the class B model; around 17 kg of natural fibers (honeycomb cardboard, coking coal, sisal, wood, cotton) in more than 27 components for the class C model; in the class E model, 21 kg of natural fibers (jute, flax, hemp, sisal and olive coke) are used in more than 44 components; almost 43 kg of natural fibers in more than 27 components for the class S model (LI et al., 2020).

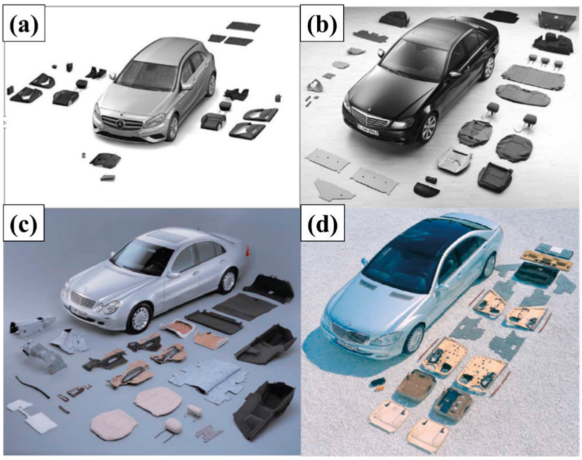

Figure 22: Components of Mercedes-Benz vehicles that feature natural fibers. (a) Class–A, (b) Class–C, (c) Class–E and (d) Class–S. Adapted with permission from Li et al. (2020).

Due to the light weight, low thermal conductivity and low environmental impact of renewable materials, Composites reinforced with natural fibers have also recently advanced for applications in masonry, soil mixture, cementitious materials, thermal insulating materials, decking applied to civil construction, furniture and architecture (AKTER e HOSSAIN, 2021). However, such materials are generally limited to thin sheet applications with high fiber volume fraction, making them particularly not useful for general construction purposes. Recently, natural fiber composites have been investigated for potentially extensive applications in several other areas, such as sports, clothing, recreation equipment, aerospace, biomedical and pharmaceutical, electrical, packaging, and electromagnetic applications, also presented in Table 7.

The global biocomposites market is expected to reach a net worth of US$ 41 billion by 2025 (ZWAWI, 2021). CRNFs have been widely used and have potential for a variety of applications, such as 3D printing (BHAGIA et al., 2021). Conventional composite processing technologies have limitations in efficiently manufacturing composites with different and complex shapes. However, 3D printing can easily manufacture different and complex parts through computer-aided design (LI et al., 2019).

(JIANG and RANEY, 2019) studied the 3D printing of amylopectin composites reinforced with cotton fiber to produce mortars in construction. Where cotton fiber reinforced amylopectin composites were stronger than common thermoplastic composites (wood fiber/PLA composites) to heat, flame and ultraviolet light. In a similar study (ZHAO et al., 2019) investigated the behavior of poplar fiber used to reinforce PLA for large-scale 3D printing application. A podium base, as shown in Figure 23, was successfully 3D printed using poplar fiber reinforced PLA composites.

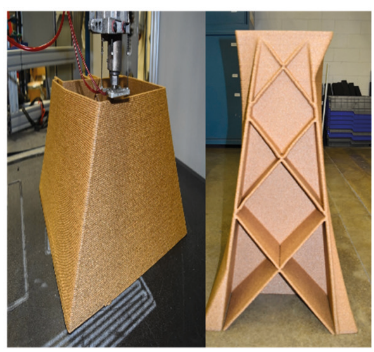

Figure 23: PLA-poplar composite produced by large-scale 3D printing. Adapted with permission from Zhao et al. (2019).

Threats related to firearms significantly affect human life, particularly those involved in public safety. So-called multilayer armor systems (MASs) offer an alternative against these threats (KHALID et al., 2021). Composites reinforced with natural fibers are considered as one of the most emerging materials for many engineering applications, especially in MASs, generally employed as the second layer of the shielding system (MONTEIRO et al., 2019).

(LUZ et al., 2020 (b)) carried out a comprehensive study using CRNFs and conventional materials (Dyneema) in ballistic armor, where the CRNFs were produced using pineapple leaves (PALF). The purpose of this armor system is to provide extra protection over normal level IIIA ballistic protection vests made with Kevlar. The incorporation of CRNFs along with the ceramic front panel results in level III ballistic protection. The results of this study show that this degree of ballistic protection was effective against 7.62 mm caliber rifle ammunition. The ballistic armor model proposed by (LUZ et al., 2020 (b)) is presented in Figure 24.

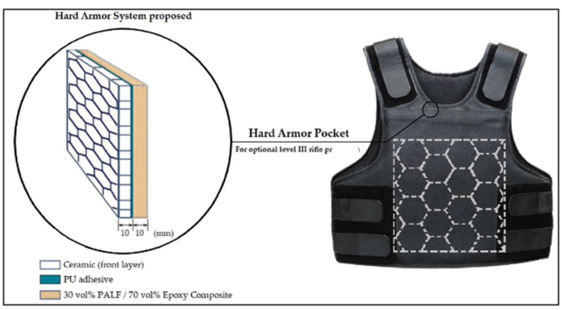

Figure 24: Schematic diagram of MASs with Epoxy/PALF composite intermediate layer. Adapted with permission from Luz et al. (2020 (b)).

In a similar study (PEREIRA et al., 2019) they evaluated polyester composites produced from fiber fibers for ballistic tests. The results showed that polyester composites with 30% fiber volume fractions are promising candidates for applications in MASs and can also replace Kevlar due to their high energy absorption and post-impact integrity.

(GARCIA FILHO et al., 2019) studied the ballistic performance of CRNFs using piassava fiber as reinforcement, the composites were produced in fiber volume fractions from 10 to 50%. Ballistic tests were carried out using 7.62 mm ammunition, where the evaluation of the ballistic test system was estimated by the penetration depth, which simulates consistency with the human body, in understanding certain needs of the NIJ 0101.06 standard. Test results showed that MASs using piassava fiber composites as a subsequent layer provide effective protection. This demonstrates that piassava fiber, which is a green material, can also be used effectively in armor systems.

CONCLUSIONS

According to the literary database available for consultation to date, it is observed that the use of CRNFs is currently growing. This interest may be directly related to the environmental aspect, since CRNFs are "green" materials and in part environmentally friendly, while they have vast possibilities for applications in various areas of knowledge, in particular replacing synthetic fibers.

It is understood that although natural fibers present possibilities for applications in composites of different matrices (polymeric, ceramic or metallic), the ones that stand out most are PMC. This survey can be related to the number of studies focused on these materials, which have shown promising properties. Thus, such characteristics result in an "awakening" on the part of some industrial sectors due to the applicability of CRNFs in their production lines.

It was also recognized that for the use of CRNFs to grow in the industrial environment, several drawbacks must be overcome, such as fiber degradation during the process, temperatures involved, excessive mechanical stress during processing and methodologies for obtaining least expensive fibers. Faced with these challenges, it is increasingly necessary to develop studies regarding the behavior presented by these materials, since in the near future CRNFs will be much more present in common applications.

It was observed that the properties of CRNFs are directly dependent on the individual characteristics presented by the reinforcement phase and matrix phase, as well as the interface. As a means of improving the properties presented, high investments are made in research, the main objective of which is to identify methodologies for the processing or surface treatment of fibers that provide better interaction with the matrix used.

Given the high number of works already published, it is concluded that CRNFs are a trend of application in different areas, where their main objective is to replace already common components with environmentally correct materials with equal or superior properties and presenting a viable production cost.

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