REVIEW

A Review of Recent Advances in Hybrid Natural Fiber Reinforced Polymer Composites

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ABSTRACT

Natural fiber reinforced polymer composites (NFRCs) have demonstrated great potential for many different applications in various industries due to their advantages compared to synthetic fiber-reinforced composites, such as low environmental impact and low cost. However, one of the drawbacks is that the NFRCs present relatively low mechanical properties and the absorption of humidity due to the hydrophilic characteristic of the natural fibre. One method to increase their performance is hybridization. Therefore, understanding the properties and potential of using multiple reinforcement's materials to develop hybrid composites is of great interest. This paper provides an overview of the recent advances in hybrid natural fiber reinforced polymer composites. First, the main factors that affect the performance of hybrid fiber-reinforced composites were briefly discussed. The effect of hybridization on the mechanical and thermal properties of hybrid composites reinforced with several types of natural fibers (i.e., sisal, jute, curauá, ramie, banana, etc.) or natural fibers combined with synthetic fibers is presented. Finally, the water absorption behaviour of hybrid fiber-reinforced composites is also discussed. It was concluded that the main challenges that need to be addressed in order to increase the use of natural-natural or natural-synthetic hybrid composites in industry are the poor adhesion between natural fibers and matrix, thermal stability and moisture absorption of natural fibers. Some of these challenges were addressed by recent development in fibers treatment and modification, and product innovation (hybridization).

KEYWORDS

Natural fiber composites (NFC); hybrid composites; mechanical properties; thermal properties; water absorption

1 Introduction

Several industrial sectors (e.g., the automotive and aeronautics), are continuously seeking development of lighter materials with increased resistance with the goal of improving system reliability and efficiency. Therefore, composite materials present an excellent solution to this demand, especially the fiber reinforced polymeric composites, due to their high strength and stiffness, as well as presenting low density [1]. Nowadays, the industry is looking for new characteristics of composite materials, such as renewability, low environmental impact and cost. Consequently, there is an increasing augment in research and innovation in the natural fiber reinforced composite (NFRC) materials, mainly due to the advantages of these materials compared to synthetic fibre-reinforced composites, such as low

environmental impact and low cost. However, one of the shortcomings is that the NFRCs present relatively low mechanical properties. One method to increase the mechanical performance of NFRCs to extend their applications is the hybridisation [2,3]. Hybrid composites use different reinforcement fibers, which can be arranged in a random, layer-by-layer or in-yarn manner [2]. The possible combinations of fibers in hybrid composites include synthetic–synthetic, natural–synthetic and natural–natural fiber types.

It is worth mentioning that the main natural fibers studied and applied in industry (i.e., jute, sisal, oil palm, kenaf, and flax) are well established on the global market with a well-defined production line. However, new promising types of natural fibers are being discovered and used on a smaller scale or are still being used only for research. It is the case of the buriti and curauá fibres, for example, that still need some improvements in their production line to be more commercially affordable and widespread used [4–6]. Another relevant factor in using natural fibers as reinforcement materials in composites is their eco-friendliness associated with lower energy consumption in their manufacturing process [7]. Nevertheless, its recyclable properties depend also on the type of matrices used in the composite production and the management of residuals produced by superficial treatments used to improve the adhesion between fiber/matrix [8].

Natural fiber reinforced polymer composites have demonstrated outstanding potential for many different applications in various industries such as automotive, domestic furniture, food packaging, and agricultural industry [7,9–11]. The growing importance of natural fiber reinforced composites is reflected by the increasing number of publications (e.g., reviews, patents, book chapters and books) during the recent years [12–17]. Although there are several reviews concerning the use of natural fibers in the production of natural hybrid composites, this paper provides an overview of the recent advances in both interlaminar and intralaminar hybrid natural fiber composites with natural-synthetic and natural-natural fiber combination in thermoset polymeric matrices. First, the main factors that affect the performance of hybrid fiber-reinforced composites were discussed. The effect of hybridization on the mechanical and thermal properties of hybrid composites reinforced with several types of natural fibers (i.e., sisal, jute, curauá, ramie, banana, etc.) or natural fibers combined with synthetic fibers is presented. Finally, the water absorption behaviour of hybrid fiber-reinforced composites is also discussed.

2 Main Factors that Affect the Performance of Hybrid Fiber-Reinforced Composites

The main factors that affect the performance of hybrid fiber-reinforced composites (types of fiber and resin, and the effect of fiber surface treatment), are briefly discussed in this section.

2.1 Type of Fiber

The natural fibers are promising renewable alternatives to the traditional human-made fibers (i.e., glass and carbon fibers) in composite materials. Their consumption is increasing in different industrial sectors, such as aeronautical, automotive, civil, and naval, with an average annual growth of 7.5% and an expected global market demand around of 35 million tons by 2022 [16].

The natural fibers can be divided into three main groups regarding their origin: mineral, animal and cellulose/lignocellulose (see Fig. 1). Mineral-based fibers were widely used in composite materials. However, their use presented many human health issues (carcinogenic parts that can be inhaled or ingested), nowadays being forbidden in many countries worldwide. The animal fibers present lower mechanical properties compared to the cellulose fibers, except for the silk fiber that presents high tensile strength. However, the silk fibers are quite expensive and are more used in the textile industries [1]. The animal fibers (i.e., protein fibers such as silk and wool) and the mineral fibers are not covered in this review.
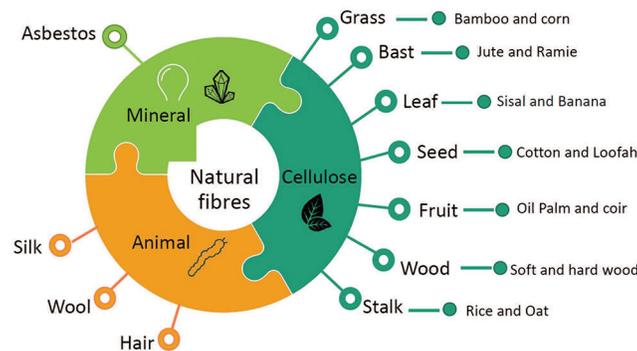
Figure 1: Schematic of natural fibers classification

The cellulose/lignocellulose fibers are the most used natural fibers due to their relatively low price and higher mechanical properties compared to other natural fibers. They can also be subdivided into different categories corresponding to the plant part that originated them.

The cellulose/lignocellulose fibers are primarily composed of cellulose (α–cellulose), hemicellulose and lignin [18]. They also present small quantities of waxes in their outer part. The chemical composition and cell structure of cellulose fibers are somewhat complicated [19,20]. Each fiber is basically a composite in which rigid cellulose microfibrils are embedded in a lignin and hemicellulose matrix. Moreover, the microfibrils are helically wound along the fiber axis to form hollow cells. For instance, the curauá, jute, and sisal fiber morphology are different with distinct lumen diameters (4, 6 and 8 μm, respectively) and cell wall thickness [10,21]. Besides, they also present different percentages of lignin, cellulose and hemicellulose in their configuration. It is worth mentioning that the amount of cellulose and lignin in the natural fibers depends on the environmental conditions during the plant cultivation, soil properties, irrigation, and extraction techniques (i.e., automatic harvesting or manual extractions) [22]. These characteristics changes the natural fiber's morphology, and consequently, their mechanical and thermal properties [23–27]. The main mechanical properties of the most used natural fibers in hybrid composite materials and their cellulose content are summarised in Tab. 1. The tensile strengths and Young's moduli of these natural fibers cover an extensive range of values with typical tensile strength in the range of 100 to 1000 MPa (except for the curauà fibers), which are generally lower than the properties of synthetic fibers (e.g., the tensile strength of E-glass fiber is about 2000–3500 MPa).

Generally, the fibers with higher cellulose contents present higher tensile strength and Young's modulus values (i.e., curauá, sisal and ramie). However, the high variability in fiber quality, which leads to high scattering in the material property values, is still a limitation, showing that many factors can influence their behaviour as the already cited environmental and processing conditions. Therefore, more studies about these parameters are necessary, especially in consideration to the fiber's conservation techniques. Another relevant issue is the lack of detailed information about single-fiber experimental testing procedures, making it difficult to compare the results obtained from different studies. Usually, these results are based on the sample's total cross-section, and its non-corrected measurement can significantly affect the tensile strength results, explaining the different values found in the literature [1].

Synthetic fibers like glass, carbon and aramid are being used in hybrid polymer-based composites mainly because of their high stiffness and strength properties [28]. However, the main drawbacks of using these fibers are their biodegradability, initial processing costs, recyclability, energy consumption, machine abrasion, health hazards, etc.

Table 1: Mechanical properties of several cellulose fibers [10,18,22,29–32]

Fiber	Density (g/cm³)	Length (mm)	Diameter (µm)	Elongation (%)	Tensile strength (MPa)	Young's modulus (GPa)	Cellulose content (% in weight)
Jute	1.23	0.8–6	5–25	1.5–3.1	187–773	20–55	45–63
Linen	1.38	10–65	5–38	1.2–3	343–1035	50–70	–
Sisal	1.20	0.8–8	7–47	1.9–3	507–855	9–22	50–73
Ramie	1.44	40–250	18–80	2–4	400–938	61.4–128	68.6–76.2
Hemp	1.35	5–55	10–51	1.6–4.5	580–1110	30–60	30–77
Coir	1.20	0.3–3	7–30	15–25	175	6	15–51.4
Kenaf	1.20	1.4–11	12–36	2.7–6.9	295–930	22–60	45–57
Banana	1.50	0.4–0.9	12–30	5–6	529–914	27–32	63
Curauá	0.20	1500	97	4.5–6	1250–3000	30–80	70–73
Pineapple	1.50	1–8	8–41	1–3	170–1627	60–82	81
Abaca	1.50	4.6–5.2	10–30	2.9	430–813	31.1–33.6	56–63
Bamboo	0.6–1.1	1.5–4	88–25	1.3–8	140–441	11–36	74
Nettle	1.51	5.5	20–80	1.7	650	38	53–73
Flax	1.5	5–900	–	2–4	300–800	30–60	30–77
Palm	0.7–1.5	0.59–142	150–500	17–25	150–500	3.24	30
Softwood	1.5	2.8–4.1	28–47	–	1000	18–40	–
Hardwood	1.2	2.67–3.98	31	–	–	37.9	–
Kapok	0.31–0.38	20–32	20	1.2	93.3	4	–

2.2 Surface Treatment of Natural Fibers

Surface treatment of natural fibers is usually performed to enhance the properties of natural fibers, before using them to manufacture a composite material. Fiber surface modification can improve the fiber-matrix interfacial bonding, roughness, wettability and also decrease the moisture absorption of the fibers [33–42]. As a result, better mechanical properties of the resulting composites can be achieved. In this work, the focus will be on natural fibers, since the treatment suffered by the fiber improves the performance of the hybrid composites. The main surface modification methods used for natural fibers (i.e., physical and chemical) are briefly presented in this section.

2.2.1 Physical Treatment Modifications

The main physical treatments used to modify the surface of natural fibers are as follows: plasma, corona, ultraviolet (UV), heat treatments, electron radiation and fiber beating [15,42–44]. These treatments improve the adhesion between the fiber and matrix by changing the surface proprieties of the fibers without changing their structural composition [7,36]. However, physical treatments are more expensive compared to chemical ones, mainly because of the equipment involved in the surface modification processes [7,15]. The principal effects of physical treatments on the natural fibers are summarised in Tab. 2.

2.2.2 Chemical Treatments

Chemical treatments are a common way of increasing the interface adhesion between the fiber-matrix by chemical bonding or mechanical interlocking at the interface and decreases the water absorption of the fibers [8,25]. This is achieved by using compounds that promote adhesion by chemically coupling the fibers to the matrix, such as: sodium hydroxide [25,45–52], silane [8,23,29], acetic acid [36,42,43,53], Benzoyl Chloride [43,54,55], maleated coupling agents [39,42,43], isocyanates [4,8], peroxide [31,56,57], stearic acid [58], etc. It was shown in the literature that the fiber modification approaches have different levels of efficacy.

In general, chemical approaches provide better improvements in properties compared to physical approaches. The improvement in mechanical and thermal properties depends on concentration of the chemical used and exposure time. Combination of treatments with two different chemicals presented better mechanical and thermal properties than the individual treatments in some cases.

Table 2: Principal physical treatments used to modify the surface of natural fibers

Surface treatment	Treatment effect	References
Plasma	Plasma treatment promotes the removal of contaminants from the fiber surface through ionising gases. Furthermore, this treatment improves the surface roughness by increasing interfacial adhesion between the fiber-matrix.	[4,42]
Corona	Corona treatment promotes surface fiber change by oxidation process and introduction of polar groups, which results in cleaning of the fiber surface and improving the interfacial adhesion between the fiber (hydrophilic) and matrix (hydrophobic).	[39,43]
Ultraviolet (UV)	UV treatment affects the polarity of the fiber improving wettability and adhesion between the matrix-fiber. In addition, several bonds are broken (C-C, C-O, C-F, C-Si and C-H) promoting the cleaning of the fibers.	[8,36]
Ultrasound	Ultrasound process promotes the cleaning of the surface of natural fibers in two ways: micro-jetting and micro-streaming.	[43]
Thermal	Thermal treatment heats the fiber between 100–200°C, affecting their physical and chemical property, cellulose crystallinity, decrease the water percentage and low temperature constituents.	[8,36]

Natural fibers that undergo chemical and physical surface modifications presented improved fiber-matrix interfacial adhesion, improved fiber roughness and wettability. In addition, most of the treatments help to minimise the hydrophilic nature and decrease the moisture absorption of natural fibers. The selection of the appropriate fiber modification technique will depend on the particular fiber/matrix used and the composite end-use application. It is essential to understand the interfacial properties and bonding mechanisms of fiber-matrix and this involves significant research efforts in order to maximise NFRCs and hybrid composite applications.

Tab. 3 presents the most common chemical treatments used to treat/modify the surface of natural fibers.

Table 3: Effect of chemical treatment of natural fibers

Chemical treatment	Treatment effect	References
Alkaline	Alkalisation or mercerisation treatment promotes cleaning of impurities, decreases hydrophilicity and fiber constituents. The treatment reduces the diameter and enhances the surface roughness of the fiber and increase the interfacial adhesion of the fiber-matrix.	[4,38,59]
Silane	Silane treatment promotes micropore coating on the natural fiber surface. Improves fiber-matrix interfacial adhesion. Pre-treatments are performed, mainly alkalisation, for cleaning existing constituents.	[8,23,29]

(Continued)

Table 3 (continued).

Chemical treatment	Treatment effect	References
Acetylation	Acetylation treatment uses acid catalysts (acetic anhydride and acetic acid) for the superficial plasticisation of the natural fiber, increasing interfacial bonding.	[36,43,53]
Benzoylation	Benzoylation treatment decreases the hydrophilicity of the natural fiber. Benzoyl Chloride is used to improve the interfacial adhesion between the fiber-matrix and the thermal stability of the fibers. Alkalisation pre-treatment is performed to activate the hydroxyl groups of the fiber.	[43,54,55]
Maleated coupling agents	This treatment is used to modify the fiber surface and matrix with the addition of a coupling agent. The maleic anhydride is grafted onto the polymers to increase the compatibility between the matrix and the coupling agent.	[43,60]
Isocyanate	The treatment is used as a coupling agent in natural fibers, where it can react with the hydroxyl groups found in the fiber, improving interfacial adhesion and increasing water resistance.	[4,8]

2.3 Type of Matrix

The mechanical properties (e.g., tensile, flexural, and impact) of composite materials are directly related to their fiber/matrix interface, where poor adhesion between these elements will result in an inefficient dispersion of loads [1]. Therefore, choosing the correct fiber/matrix combination is fundamental to obtain good composite mechanical and thermal properties. The matrix provides resistance to mechanical and chemical abrasion of the reinforcement fibers, keeps the fibers in the desired position, and is responsible for an efficient load transfer between them [61,62]. The most common matrices used to produce hybrid natural fiber composites are polymeric (thermoset or thermoplastic) mainly because they can be processed at low temperature and are lightweight [63].

Fig. 2 presents a diagram of classification of matrices with the main polymeric matrices types used to produce composites.
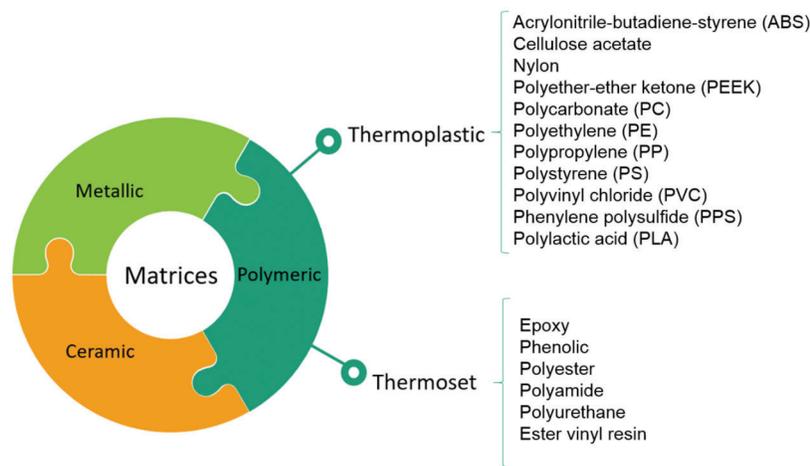

Figure 2: Schematic of classification of matrices with the main polymeric matrices types

Thermoplastic polymer matrices can be softened, melted and, also, recycled by the application of heat. However, the continuous reprocessing of these polymers can deteriorate their mechanical and thermal properties [64]. On the other hand, the thermoset matrices (epoxy, urethane, Vinyl Ester, phenolic, polyester, polyimide, polyurethane (PU)) can hardly be recycled or change the shape once the polymerisation (curing) is concluded. These matrices have different chemical structures and undergo different reactivities with the surface molecules of fibers in composites [65].

The main thermoset resins used for hybrid natural composites are epoxy and unsaturated polyesters [66,67]. Epoxy resins have high mechanical strength and environmental degradation resistance. In general, epoxy resins have excellent adhesion properties, and they are easy to cure and use. In addition, no volatile agents are formed during the curing process, which is an important advantage as compared to phenolic, polyester, and vinyl ester resins. Finally, the shrinkage associated with epoxy resins is lower compared to polyesters. However, one disadvantage is that epoxy resins are relatively brittle, which is detrimental to the interlaminar properties between the matrix and the fiber reinforcement. It is known that epoxy resin cures from liquid to solid through a catalytic chemical reaction, creating extremely strong bonds that cannot be easily reversed or reformed. Because of this, the performance of this matrix material is superior to other matrices; nevertheless their recycling remains challenging. To summarise, as the majority of natural fibers become unstable at temperatures higher than 200°C, the matrices that require high processing temperatures are not suitable to manufacture NFRCs or hybrid natural fiber composites.

3 Mechanical Properties of Hybrid Fiber-Reinforced Composites

Several researches were carried out in the literature to study the mechanical properties of hybrid composites reinforced with several types of natural fibers (i.e., sisal, jute, curauá, ramie, banana, etc.) or natural fibers combined with synthetic fibers [2,20,23,59,60,66,68–84]. The main objective of hybridisation is to balance the deficiency of one specific fiber to achieve sustainability, low cost, and improved performance of the material [2]. Generally, natural hybrid composites are produced by hybridising natural fibers with another either natural or synthetic fiber with superior properties (i.e., higher mechanical strength, chemical stability, nontoxicity, resistance to high temperatures, and thermal or acoustic insulation). Hybridisation of natural fibers with synthetic fibers (i.e., glass, carbon, Kevlar, basalt fibers) can significantly improve the mechanical performance of NFRCs and also their water resistance [20,71]. The glass fiber is the most used fiber in the hybridisation of NFRCs. Carbon and aramid fibers are also used in NFRC hybridisation but are limited by their high cost and difficult recyclability, thus reducing the main advantage of the hybrid NFRCs (sustainability). Furthermore, the carbon fiber provides stability and high mechanical properties to the hybrid composites but at a much higher cost [85]. For Kevlar, the main use found in literature is for higher impact resistance and in this case, the hybridisation is focused in reducing the number of aramid layers with minimal loss in impact resistance [86]. Finally, another common approach is the substitution of synthetic fibers with natural fibers. This is done with the increase of natural fiber fraction in the hybrid composite. The main purpose is to reduce costs and environmental impact of the composite, while minimizing mechanical property losses [85–87]. Hybrid composites of natural fibers with other natural fibers might not show comparable mechanical enhancements as those hybridised with synthetic fibers, but the natural-natural fibers hybrid composites will be superior in terms of sustainability.

The mechanical properties of fiber reinforced composites are mainly affected by fiber content and orientation [88], stacking sequence [86,89,90], hybrid ratio and the fiber-matrix interfacial strength [1–2,36]. For example, the tensile strength of hybrid composites is largely dominated by the reinforcing fiber properties, where the failure begins with the fiber with lower elongation. Considering iso-static loading of all fibers in the hybrid composite the fibers with lower strain at failure will fail first. This results in the failed fibers not participating in carrying the load. Therefore, the fibers with higher

elongation will have to carry the brunt of the load. Consequently, the composite should present a compromise between the high and low strain fibers [1,2]. Fig. 3 shows the hybrid effect of a 50:50 wt% LE/HE fiber composite (the hybrid effect is the apparent failure strain enhancement of the LE fiber in a hybrid composite compared to the failure strain of a LE fiber-reinforced non-hybrid composite).
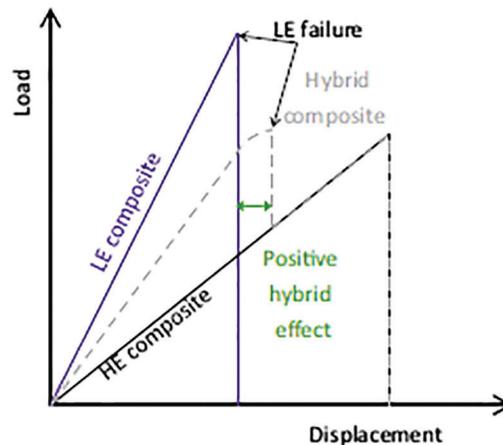

Figure 3: Illustration of the hybrid effect (the apparent failure strain enhancement of the LE fibers, under the assumption that relative volume fraction is 50/50 and that the hybrid composite is twice as thick as the reference composites: LE-low elongations; HE-High elongation). Reproduced with permission from [2]

The volume fraction of each type of fibers has an impact in how the failure will occur. For composites with high strain fiber volume, these fibers will dominate the stress distribution. The resulting stress/strain curve will present a higher secondary peak. If the fiber fraction is reversed, the composite's strength will be limited by the low strain fiber [2]. Another factor that can affect the mechanical properties of hybrid composites is the weight ratio, also known as volume or weight fraction of the reinforcing fiber constituents [1]. By varying the weight ratio/volume of the fibers it is possible to change the composites overall mechanical properties. Therefore, depending on the application requirements the hybridisation method provides flexibility in fiber selection for achieving the desired materials properties.

For the flexural properties the stacking sequence of the hybrid composite is highly relevant [91,92]. The tensile and compressive stresses are maximum in the bottom and top faces of a specimen under 3-point bending flexural load, respectively and they linearly decrease towards the neutral line (middle of the specimen). As such the fibers with higher mechanical properties should be placed in the outward most lamina in a composite. This is especially interesting when using synthetic/natural hybrid composites, since most synthetic fibers also present a hydrophobic characteristic, and thus will reduce water absorption [78,93]. The impact properties of natural hybrid composites was found to vary greatly in the literature. The impact resistance of hybrid composites is strongly linked to the fiber mechanical properties [94,95], layer stacking sequence [2] and the interfacial adhesion between the fiber and the matrix [23]. Some authors found that the stacking sequence has an insignificant effect on the impact strength, while the fiber orientation in the hybrid composite has a noticeable effect on the impact strength [91].

Tab. 4 summarises some recent studies on the effect of hybridisation on the mechanical properties of hybrid composites.

Table 4: Effect of hybridisation on the mechanical properties of hybrid composites

Composite (type of fiber and matrix)	Fabrication technique and test method	Mechanical properties	References
Sisal/jute/glass (Polyester-methyl ethyl ketone)	Hand-lay-up with compression moulding in a hydraulic press. Tensile, flexural, impact and interfacial shear strength tests	The 20% sisal, 20% jute and 60% of glass-fiber presented the best performance in tensile, flexural and impact properties when compared to the other hybrid composites.	[88]
Banana/glass/sisal (Epoxy resin-LY556)	Compression moulding. Tensile, flexural and impact tests	The hybrid banana/sisal/glass fiber composite presented highest tensile strength (glass fiber on the outer layers and in the middle). For flexural strength, the best configuration was found for the banana/sisal composite as a sandwich between layers of glass fiber.	[96]
Sisal/cotton (Isophthalic polyester)	Hand-lay-up. Tensile, flexural and impact tests	The optimal composite presented a percentage of 40% of cotton/sisal reinforcement. Sisal aligned in the load application direction presented better results when compared with the cotton ones.	[69]
Flax/glass (Phenolic)	Compression moulding. Tensile, mode I fracture toughness and short beam tests	For the mode I fracture toughness and the interlaminar shear strength the hybrid composites with higher flax content presented better results when compared with pure glass fiber composites.	[58]
Banana/Kenaf (Unsaturated polyester)	Hand-lay-up. Tensile, flexural and impact tests	The hybridisation process increased the mechanical properties of the single banana and kenaf composites. The superficial treatments improved the tensile, flexural and impact strengths around 10% of the hybrid composites when compared with the untreated condition.	[59]
Twisted kenaf/neem/glass fibers (Epoxy resin-LY556)	Vacuum assisted compression moulding. Tensile, flexural, impact and double shear tests.	The glass fiber + twisted neem (vertical) + twisted kenaf (inclined) + twisted neem (vertical) + glass fiber hybrid composites presented the best mechanical properties with a tensile ultimate strength around 70 MPa and absorbed impact energy equivalent to 12 J.	[97]
Curauá/glass (Polyester)	Compression moulding using a hydraulic press. Barcol hardness and impact tests.	The impact properties and composite hardness increased in the composites with higher percentages of glass fibers, due to better interfacial adhesion of these fibers with polyester resins.	[72]
Banana/jute (Epoxy)	Hand-lay-up. Tensile, flexural, impact and interfacial shear strength tests.	The hybridisation improved the mechanical properties of the studied composites, especially the impact properties. The maximum value of interlaminar shear strength was found for the banana/jute composite with 30 wt% of fiber loading.	[98]
Sisal/bamboo (Polyester resin–methyl ethyl ketone)	Hand-lay-up. Tensile, flexural and impact tests.	The mechanical properties of the hybrid composites increased in the composites with higher weight percentage of bamboo. The alkalinisation process also improved the tensile strength by 30% and the impact energy by 40%.	[71]
Echinoidea spike particles and kenaf (Neem oil blended epoxy)	Hand-lay-up. Tensile, flexural, impact test.	The Neem oil had little impact in the epoxy resin with a slight decline in mechanical properties. The addition of the kenaf fibers had a positive impact in mechanical properties. The addition of the hybrid kenaf- particles increased the mechanical properties of the composite.	[99]
Flax/TiO$_2$ (Epoxy)	Hand lay-up with compression moulding. Tensile, flexural, impact and short beam tests were carried out.	Improvements in tensile, flexural, impact and ILSS of hybrid composites were approx. 11%, 20%, 10% and 19% when compared to the unmodified samples.	[100]
Jute/nano-clay (Epoxy)	Hand lay-up followed by compression moulding. Tensile, flexural, impact testing.	The best material properties overall were found for the 5% NaOH-treated jute fiber with 5 wt% nano-clay content.	[101]

(Continued)

Table 4 (continued).

Composite (type of fiber and matrix)	Fabrication technique and test method	Mechanical properties	References
Pineapple/banana/sisal/ TiO$_2$ (Epoxy)	Compression moulding. Tensile, flexural, impact testing as well as vibrational analysis.	The highest mechanical property increase was observed for the filler content of 3% for all cases (improvements of approx. 20%).	[102]
Jute/Carbon (Epoxy)	Vacuum assisted resin infusion fabrication method. Tensile, flexural, impact testing as well as water absorption.	The highest mechanical properties were reported for the unidirectional fiber orientation compared to the angle and cross-ply hybrid composites. The flexural property varied significantly with stacking sequence. The highest value was reported for the C2J6C2 case. The impact property was not affected by the stacking sequence.	[91]
Jute/ramie, jute/sisal and jute/curauá (Epoxy)	Tensile, flexural and impact testing.	The hybridisation had a positive effect in tensile, flexural and impact strength for all studied composites when compared to the non-hybrid. The jute/sisal and jute/curauá presented the best overall mechanical properties (an increase of 68% and 77%, for tensile strength over the simple jute composite, respectively).	[23]
Flax/glass and jute/glass fibers (Epoxy)	Vacuum infusion method. Tensile, flexural, dynamic mechanical analyses.	Flexural strength increased when glass fiber was placed to the outer layers ("glass/flax/glass" or "glass/jute/ glass") with the increase in jute and flax fiber percentage.	[92]
Carbon, flax, and flax/ carbon hybrid (Epoxy)	Tensile, and flexural properties.	Flax/carbon fiber hybrid composites exhibited improvement in elongation at break compared to pure carbon composites. Addition of flax fibers decreased the flexural strength of carbon composites due to their inherently lower strength.	[103]

4 Thermal Properties of Hybrid Fiber-Reinforced Composites

The main techniques used for thermal analysis of hybrid epoxy/synthetic/natural fiber composites are as follows: Thermogravimetric Analysis (TGA), Differential Scanning Calorimetry (DSC), Thermomechanical Analysis (TMA) and Dynamic Mechanical Analysis (DMA). The main thermal properties of the composites provided by these techniques are: the crystallisation temperature (T_c), melting temperature (T_m), glass transition temperature (T_g), thermal expansion coefficient (CTE), viscoelastic behaviour and thermal stability (see Fig. 4). It was shown in the literature that several factors can affect the thermal properties of the composite materials, such as: fiber and matrix type, manufacture process, and type of loading [104–106]. In addition, the fiber orientation and volume and chemical treatment of the fibers will also impact the thermal properties of the composites [15].
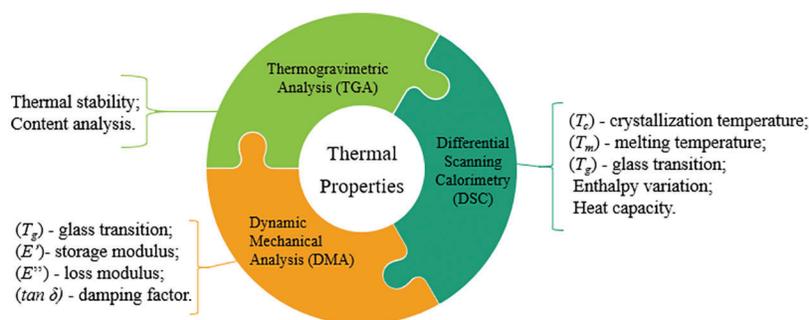

Figure 4: Schematic of the principal methods used to determine the thermal properties of composites with the main thermal properties of the bio composites provided by these techniques

TGA analysis consists of measuring the weight of a material sample, commonly either as a function of increasing temperature, or isothermally as a function of time, in an atmosphere of nitrogen, helium, air, or other gases [107,108]. Typical thermogravimetric parameters consist of moisture, volatile substances, loss on ignition or ash. Different temperatures and measurement times are applied in accordance with the matrix type of the composite sample [107–110]. Natural fiber based composites have the thermal stability measured from the decomposition of their constituents (hemicellulose, cellulose, lignin and others) [24,53,66,111–114]. The incorporation of synthetic fibers in natural composites will increase their thermal stability and shift the degradation point of the composites to the right, decreasing its thermal degradation [115]. The thermogravimetric behaviour of hybrid synthetic/natural composites depends on composite constituents (fibers and matrix). For example, similar natural fibers such as jute, sisal, wood and cotton have similar TG/DTG curves and thermal decomposition patterns. The derivative thermogravimetric test (DTG) shows the maximum rate of peak thermal decomposition, and fiber constituents are indicated by peaks in each degradation range. Lignocellulose fibers (i.e., jute, flax, wood, sisal, ramie, curauá and others) present three stages of degradation, the first stage is related to water loss by natural fiber (60°C–100°C). The second stage is related to the loss of the main constituents of the fibers: hemicellulose, cellulose, and lignin (200°C–500°C). Finally, the last stage of degradation is the formation of active coal as a form of residue [53,111–114].

The DSC determines the material transitions as a function of temperature and time. The endothermic (heat absorption) and exothermic (heat released) peaks and magnitudes indicate the thermal phase transformation of the composites [116,117]. Thermal data extracted from this analysis are: the glass-transition temperature (T_g), crystallisation temperature (T_c), fusion temperature (T_m), enthalpy variation and heat capacity [118]. The glass transition temperature (T_g) is the temperature band in which a thermoset polymer changes from rigid to a more flexible or 'rubbery' state. There are several factors that can affect the data collected by the DSC analysis, such as: sample size and shape, heat ramp and type of atmosphere. Several authors used the DSC technique to investigate the effect of hybridisation on the glass transition and the peaks (endothermic and exothermic) of hybrid epoxy/synthetic/natural composites [66,87,119–121].

The following data can be extracted from DMA: storage modulus (E'), loss modulus (E''), damping factor ($tan\ \delta = E''/E'$) and glass transition temperature (T_g) [118]. The storage modulus (E') is associated with the energy storage of the elastic characteristics of the material [24,105,122]. It decreases with increase in temperature and is associated with "stiffness" of the composites sample [105,122]. The loss modulus (E'') is associated with the energy dissipation promoted by the viscous part of the composite sample. This dissipation is related to the internal molecular friction of the molecular chains due to the following factors: morphological transformation and relaxation, morphology, and system heterogeneity [105,108]. The damping factor is defined by dividing the storage and loss modulus ($tan\ \delta = E''/E'$) and is associated with the internal mobility of the polymeric molecular chains, showing the influence of the fiber/matrix interactions [24,105,123]. A high $tan\ \delta$ value shows that the system is dissipating more energy than it is storing due to fiber/matrix interaction quality, while a low $tan\ \delta$ value shows that the polymeric chain has lower mobility indicating a good fiber/matrix interfacial interaction. To determine T_g using DMA, measurements of the complex modulus are typically made as temperature is increased at a constant heating rate. The thermal properties of the composites obtained from DMA depends on the physical or structural arrangement of phases such as interface, morphology and the nature of composite constituents. It was shown in the literature that the presence of additive fillers, fiber content, fiber orientation, the chemical treatment of the fibers and the mode of testing governed the dynamic mechanical properties of a composite material [105,124]. Several researchers used the DMA tests to study the influence of hybridisation on the thermal properties of the hybrid composites [24,92,119,125–129].

Tab. 5 summarises several recent studies on the effect of hybridisation on the thermal properties of hybrid composites.

Table 5: Effect of hybridisation on the thermal properties of hybrid composites obtained from TGA, DSC and DMA analysis

Composite	Method and parameters	Effect hybridisation on the thermal properties	References
Sugar palm/ Glass (Polyester)	**TGA**–The temperature was ramped from 30°C to 600°C with a heating rate of 20 °C/min, under a Nitrogen Atmosphere with 50 mL/min. The equipment used the GA Q500 from TA Instruments. **DMA**-Three-point Bending Test with frequency of 1 Hz. Equipment (DMA Q800 of TA Instruments). The temperature was ramped from 30 to 150°C with a heating rate of 5 °C/min. Rectangular specimens having size 60 mm × 10 mm × 3 mm were used.	**TGA**–the increase in fiberglass decreased the residue percentage. **DMA**-the hybridisation and the chemical treatment provided a higher storage and loss modulus, but smaller *tan δ*.	[130]
Cotton/Glass (Polyester)	**DMA**-Three-point Bending Test with Frequency of 1 Hz an Oscillation amplitude of 15 μm. Equipment used DMA 2980 Dynamic Mechanical Analyser. The temperature was ramped from room to 180°C with a heating rate of 3 °C/min. Rectangular specimens having size 60 mm × 10 mm × 3 mm were used.	**DMA**–T_g of hybrid composites increased compared to synthetic and natural fiber pure composite.	[131]
Bamboo/Kenaf (Epoxy)	**TMA**–Expansion or Compression test with minimal force of 0.02 N. The temperature was ramped −30°C to 100°C with a heating rate of 5 °C/min. **DMA**-Three-point bending Test with frequency of 1 Hz an oscillation amplitude of 10 μm and constant current force of 3 N. Rectangular specimens having size 45 mm × 10 mm × 3 mm were used. The temperature was ramped from 25°C to 180°C with a heating rate of 5 °C/min.	**TMA/DMA** The expansion coefficient of the hybrid composites (50% bamboo/50% kenaf) was lower when compared to the other cases. T_g and storage modulus results showed an increase in thermal properties.	[126]
Jute/oil palm (Epoxy)	**TGA**-The temperature was ramped from 30°C to 900°C with a heating rate of 20 °C/min, under a Nitrogen Atmosphere. Perkin Elmer thermal gravimetric analyser.	**TGA**–The increase of percentage of jute fiber improved the thermal stability of the composite.	[113]
Jute/Coir (Epoxy Novolac)	**TGA**–The temperature was ramped from 30°C to 700°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere with 30 mL/min. Equipment DTG-60 (Schimadzu). **DSC**-The temperature was ramped from 20°C to 400°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere with 50 mL/min. Equipment DSC Q10 (TA Instruments). **DMA**-Three-point bending Test with frequency of 10 Hz. Rectangular specimens having size 60 mm × 10 mm × 3 mm were used. The temperature was ramped from 30°C to 200°C with a heating rate of 5 °C/min. Equipment DMA Q800, TA.	**TGA**-The increase in coir fiber percentage increased the onset of degradation temperature of the hybrid composite. **DSC**-Hybrid composites presented higher enthalpy values than the pure resin. **DMA**-The hybrid composite (50% Jute fiber/50% coir fiber) presented the largest storage modulus and the lowest *tan δ* when compared to the other hybrid composites.	[132]

(Continued)

Table 5 (continued).

Composite	Method and parameters	Effect hybridisation on the thermal properties	References
Cocos nucifera sheath (CNS) /Kevlar (Epoxy)	**TGA**–The temperature was ramped from 30°C to 900°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere with 50 mL/min. Equipment Mettler Toledo DSC. **DSC**-The temperature was ramped from 30°C to 900°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere. Equipment Mettler Toledo DSC. **DMA**-Three-point bending test with frequency of 1 Hz. Rectangular specimens having size 60 mm × 12.5 mm × 3 mm. The temperature was ramped from room temperature to 200°C with a heating rate of 10 °C/min. Equipment DMA Q800, TA.	**TGA**-The 75%G/25%CNS hybrid presented the highest thermal stability when compared to the other hybrid composites. **DSC**-The DSC results were similar to the TGA. The 75%G/25%CNS hybrid composites presented lower endothermal peaks when compared to the other studied hybrid composites. **DMA**-Hybridisation with 25% CNS/75% K presented superior storage modulus value when compared to pure Kevlar composite.	[87]
Sugar Palm/ Glass (Epoxy)	**DMA-Dual Cantilever Test** with Oscillatory frequency of 1 Hz. Equipment used DMA Instrument Q800 V20.24 Build 43 Module DMA Multi-Frequency. The temperature was ramped from 25°C to 150°C with a heating rate of 5 °C/min.	**DMA**-The hybridisation and Benzoyl treatment increased the storage and loss modulus of the composites.	[133]
Banana/PALF/ Glass (Epoxy)	**TGA**-The temperature was ramped from 30°C to 600°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere at a flow rate of 20 mL/min. Samples of around 4.9–8.9 mg. The equipment used was TA TGA Q500. **DMA**-Three-point Bending Test with Frequency of 1 Hz. The temperature was ramped from 0–160°C with a heating rate of 3 °C/min.	**TGA**-The group with fiber percentage of 40% presented the best thermal stability within the studied composites. **DMA**-The 30 wt % banana-glass fiber hybrid composite presented the highest T_g when compared to the other cases studied.	[134]
Sisal/Glass (Epoxy)	**TGA**-The temperature was ramped from 30°C to 600°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere. Samples of approx. 20 mg. The equipment used was NETZSCH TG 209F3 Tarsus. **DSC**-The temperature was ramped from 30°C to 500°C with a heating rate of 20 °C/min, under a Nitrogen Atmosphere at a flow rate of 50 mL/min. Samples of around 20 mg. The equipment used was NETZSCH DSC 00F3 Maia.	**TGA**-The hybridisation increased the thermal stability when compared to the pure sisal. **DSC**-The natural/synthetic hybrid composite presented the lowest endothermic peak when compared to the other hybrid composites studied.	[66]
Kenaf/Pineapple leaf (Phenolic)	**TGA**–The temperature was ramped from 30°C to 700°C with a heating rate of 20 °C/min, under a Nitrogen Atmosphere. The equipment used was TGA Q 500 TA Instrument. **TMA**-The temperature was ramped from 30°C to 200°C with a heating rate of 5 °C/min, under a Nitrogen Atmosphere. Square specimens having size 5 mm × 5 mm × 3 mm^3 The equipment used was TA Instrument Q400. **DMA**-Three-point Bending Test. The temperature was ramped from 30–150°C with a heating rate of 5 °C/min. The equipment used was TA DMA Q800.	**TGA**-Hybridisation decreased the onset of degradation temperature of the composites. **TMA**-the composite with 30% Pineapple and 70% Kenaf showed high thermal resistance. **DMA**-The composite with 30% Pineapple and 70% Kenaf showed an increase in storage modulus.	[108]

(Continued)

Table 5 (continued).

Composite	Method and parameters	Effect hybridisation on the thermal properties	References
Jute/Sisal (Epoxy)	**TGA**–The temperature was ramped from 30°C to 800°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere with 20 mL/min. Samples of around 15–25 mg. Equipment Perkin Elmer TGA 4000. **DSC**-The temperature was ramped from 30°C to 400°C with a heating rate of 10 °C/min, under a Nitrogen Atmosphere at a flow rate of 20 mL/min. The equipment used was Perkin Elmer model DSC 4000. **DMA**-Three-point bending Test with frequency of 1 Hz. Rectangular specimens having size 50 mm × 13 mm × 3 mm. The temperature was ramped from 30 to 200°C. Equipment DMA 6100, Seiko.	**TGA**-The thermal stability augmented with the hybridisation and the increase of the jute fiber. **DSC**-All hybrid composites presented benefit in properties: T_g, T_c and T_d. **DMA**-The increment of jute fiber increased the storage modulus, loss modulus and T_g, of hybrid composites.	[119]
Jute/Carbon (Epoxy)	**TGA**–The temperature was ramped from 20°C to 800°C with a heating rate of 20 °C/min, under a Nitrogen Atmosphere. Samples of around 5–6 mg. Equipment SDT Q600 Model.	**TGA**-The effect of hybridisation increased the onset and end of the degradation temperatures of the composite.	[135]

To summarise, using natural fibers with low lignin content leads to better thermal performance of composites. The thermal stability of fibers can be enhanced by removing certain proportion of hemicelluloses and lignin constituents by different chemical treatments. The incorporation of synthetic fibers in natural fiber reinforced composites increases their thermal stability. The degradation of natural fibers is an important issue in the development of hybrid natural fiber reinforced composites in both manufacturing (curing, extrusion or injection moulding) and for the use of these materials in service.

5 Water Absorption Behaviour of Hybrid Fiber-Reinforced Composites

The affinity to absorb moisture by natural fibers and its composites, is widely known and reported in the available literature [12–13,25,66,136–138]. The absorption of moisture into a fiber-reinforced polymer composite leads to poor fiber-matrix interfacial adhesion, which will lead to an increasingly poor load-transferring efficiency from the matrix to the reinforcement fiber. In particular, for natural fibers, there is also the aggravating factor that water uptake within the fiber leads to the degradation of its physical, thermal and mechanical properties due to the leeching of water-soluble components [139].

Moisture diffusion in polymeric composites is governed by three different mechanisms [140]. The first involves diffusion of water molecules inside the micro gaps between polymer chains. The second involves capillary transport into the gaps and flaws at the interfaces between fiber and the matrix. This is a result of poor wetting and impregnation during the initial manufacturing stage. The third involves transport from microcracks in the matrix arising from the swelling of fibers (particularly in the case of natural fiber composites) [35] (see Fig. 5). According to these mechanisms, there are three types of diffusion behaviour of polymeric composites: Fickian diffusion model, anomalous or non-Fickian, and an intermediate case between Fickian and non-Fickian [140].

It was shown in the literature that moisture diffusion in hybrid composites depends on a variety of parameters: fiber volume fraction, fiber orientation and stacking sequence, fiber nature, voids, temperature, presence of fillers, fiber surface treatments and immersion time. For instance, higher immersion times, natural fiber volume fraction and voids increase the composite moisture uptake [13,141,142]. As well, at higher temperature of immersion, moisture uptake behaviour is accelerated and

the moisture saturation time is considerably shortened [1]. On the other hand, the fiber stacking sequence (e.g., placing the hydrophobic fiber fabrics on the outer layers), as well as the fiber orientation (e.g., 0° orientation of the natural fiber) was shown to lower the water uptake of the composites [143,144].
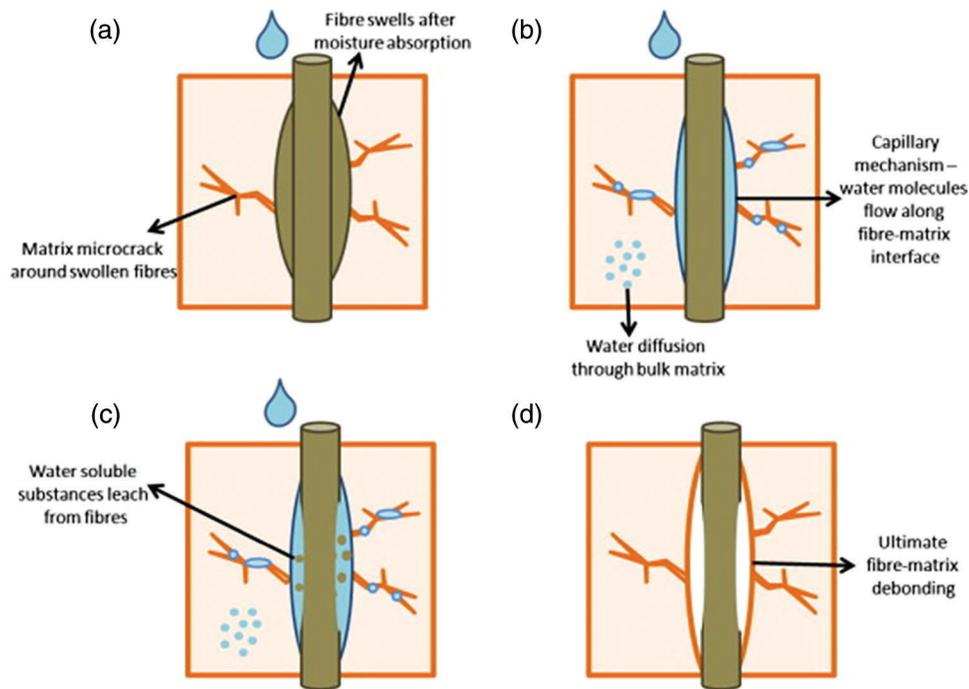

Figure 5: Effect of water on fiber–matrix interface: (a) Formation of micro-cracks due to fiber swelling; (b) water molecules diffuse in the bulk matrix and flow along the fiber-matrix interface (c) water soluble substance leaching; and (d) debonding of fiber-matrix interface. Reproduced with permission from [35]

One solution to reduce the water uptake in natural fiber composites is the fibers' surface treatments [31,34–42,145]. The main surface modification methods used for natural fibers (i.e., physical and chemical) were presented in Section 2.2 of this review. Another approach to decrease the water absorption in polymeric composites is to incorporate synthetic fillers which can fill the voids and better distribute the internal stresses in the composites [100,146–148]. For example, a number of researchers have investigated the effects of the incorporation of TiO_2 nanofiller and glass powder in the polymer matrix with the objective of delaying the effect of water absorption on composites [100,149].

Several studies have been carried out in the literature concerning the moisture absorption behaviour of hybrid composites, both natural-natural and natural-synthetic fibers reinforced composites [12–14,54,71,91,95,143,144,150–159]. Tab. 6 summarises some recent studies on the effect of hybridisation on the water absorption characteristics of hybrid composites. In general, the natural-synthetic hybridisation technique coupled with choosing an optimum fiber content, stacking sequence and fiber orientation, can drastically reduce the moisture content at saturation and decrease the diffusivity coefficient. Therefore, the moisture degradation and composite material property impact is reduced. The water uptake behaviour of natural-natural hybrid composites vary significantly as a function of reinforcement fiber, as different natural fibers present different contents of hydrophilic groups, porosity, crystallinity among other parameters, their water uptake behaviour changes. As such, the natural-natural hybridisation methods provide a viable technique so that a balance may be found in terms of cost/performance regarding its uses in moisture-rich environments.

Table 6: Effect of hybridisation on the water absorption behaviour of hybrid composites

Composite	Details of the process	Effect of hybridisation on the water absorption	References
Hemp/jute Hemp/ flax Hemp/jute/flax (Epoxy matrix)	Bi-directional woven mats. Hand lay-up compression technique.	Hemp/jute/epoxy, hemp/jute/flax/ epoxy and hemp/flax/epoxy absorbed 4.5%, 3% and 2.8% water, respectively.	[12]
Kenaf/Jute Kenaf/ Hemp Kenaf Jute Hemp (Epoxy)	Vacuum infusion technique. Interwoven hybrid fabrics.	Both kenaf/jute and kenaf/hemp hybrids absorbed less water than the single fiber composites. A reduction of moisture uptake of up to 64% was reported.	[13]
Kenaf/sisal Kenaf Sisal (Epoxy)	Plain woven kenaf and sisal fabrics. Hand lay-up compression moulding fabrication. Stacking sequence was varied.	Small variation was observed in moisture uptake as a function of hybridisation. The sisal/kenaf/ sisal case presented the highest water absorption (~3.5%).	[14]
Kenaf/jute (Epoxy)	Jute and kenaf fiber mats. Hand lay-up technique. Two stacking sequences were fabricated.	The water uptake behaviour varied significantly as a function of stacking sequence. The kenaf/ jute/kenaf (KJK) case presented the lowest water absorption (~3.8%).	[156]
Date Palm Leaf (DPL)/glass (Epoxy)	Hand lay-up fabrication. Alkaline treated short fibers. Glass fiber fraction was maintained while the DPL percentage was varied.	The rate of water absorption of the composites increased by adding more DPL fiber. Maximum water uptake was found for the 30 wt% of DPL.	[160]
Jute/carbon (Epoxy)	Plain woven jute fibers was used along with unidirectional carbon fabric. Vacuum assisted resin infusion fabrication method was applied. The stacking sequence and fiber orientation was varied.	Similar pattern of water absorption behaviour was observed for all cases or fiber orientation. The stacking sequence proved to be the most significant variable, with the synthetic layers on the outside of the natural core. The S4 ($C_2J_6C_2$) case absorbed the least amount of moisture.	[91]
Full flax fiber Full glass fiber Flax/glass fiber (Epoxy)	Vacuum assisted resin infusion method.	Water absorption for the flax–fiber composites was found to be 12 times higher than for the glass– fiber composites.	[151]

(Continued)

Table 6 (continued).

Composite	Details of the process	Effect of hybridisation on the water absorption	References
Flax/glass (Vinyl Ester)	Bi-directional flax and glass fiber fabrics were used. Hand lay-up and vacuum infusion fabrication.	Significant decrease in water absorption was observed as a function of hybridisation. The outer synthetic layers play a significant role in diminishing the moisture saturation content of hybrid composites.	[152]
Jute/glass/Carbon (Epoxy)	Plain weave bi-directional jute fabrics were used along with chopped glass mats and twill weave carbon fabrics. The hand lay-up process was used and the stacking sequence was varied.	Significantly lower absorption of moisture was reported for the inter-ply hybrid composites compared to the neat jute.	[143]
4 harness satin woven carbon fibers and flax fibers (Epoxy matrix)	Compression molding.	By covering the inner core of flax fiber/epoxy with an external layer of carbon fiber/epoxy the water absorption was kept to 4.8%–8.3%.	[144]
Flax fiber laminates Glass fiber Flax–glass composites (Epoxy)	Compression moulding Different stacking sequences.	The hybridisation of flax and glass fibers improved the moisture resistance of the flax fiber laminates by reducing the water absorption and the diffusion coefficient as well as the thickness swelling at room temperature.	[161]
Jute/glass fiber reinforced (Polyester)	Pultrusion. Different stacking sequences: JJ, GJJG, CGJJGC and SGJJGS.	The pure jute group displayed a moisture content at saturation of 47.65%, while the glass reinforced hybrid case (GJJG) presented a content of 13.31%.	[154]

6 Challenges, Market Trends and Perspectives

The global awareness of environmental issues is at an all-time high and NFRCs are nearing the point to become competitive with classical synthetic fiber-reinforced composites. This is leading advancements in nearly every industry as they seek the integration of biodegradable and more cost-efficient materials. However, there are still many challenges and a wide scope for improvement to extend their applications. The main challenges identified as potentially detrimental to market growth are supply logistics, moisture sensitivity and weak interfacial bond of NFRCs. In addition, the natural origin of these fibers, which result in variations in their properties produce large variations in properties, long-term stability and durability of NFRCs. Some of these challenges were addressed by recent development in fibers treatment

and modification, and process or product innovation (hybridization). Moreover, intense research efforts are in progress to address these limitations.

In this context, there are many exciting market trends going forward in many different industries. For example, tri-dimensional hybrid natural fiber reinforcement preforms have been used recently by sports cars manufacturers such as Porsche and even McLaren in Formula 1 [162]. This application involved the use of flax fibers in a specific 3D architecture to increase stiffness and toughness, lower weight and reduce debris in a crash. It has been successful used in F1 car seats, and the front crash absorbing structure (crash box). Similarly, many other car manufacturers such as Tesla and BMW have been investigating the use of this technique in structural components of the vehicle with promising results. These advances were made commercially available by companies such as Bcomp® and Ycom® [162,163]. Nowadays, Audi, Volkswagen, Toyota, Daimler-Benz, Volvo, Ford use NFRCs to produce components such as: seat back, interior door paneling, noise insulation panels, instrumental panel support, engine insulation, and internal engine cover. The continually growing demands for lightweight and fuel-efficient vehicles will further push the growth of NFRCs in the automotive market.

In the defence industry, natural fibers are used as an alternative in replacing Kevlar fibers. For instance, they were used in the Multilayered Armour System (MAS) due to their high impact energy absorption, low-weight, and efficiency in capturing splinters from the ceramic plate typically present in MAS [164]. The curauá based natural fibers reinforced composites, for example, are up-and-coming solutions for these applications as they are being intensely studied recently to be applied in military equipment mainly due to their excellent relationship between weight and strength [165,166].

Another booming area involving natural fibers is the application of natural reinforcements in additive manufactured parts. The advantages using additive manufacture to produce NFRCs and hybrids are the design flexibility to fabricate complex geometry parts or functionally graded composites with local-specific performance and functionality. The use of natural fibers can bring many advantages to single or dispensable production such as production of tailored, non-toxic, renewable, and recyclable parts. The advantages of natural fibers compared to synthetic fibers are especially relevant when considering their lower toll on both printer durability and production cost. However, using natural fibers and additive manufacture (AM) still presents significant challenges such as: composite filament preparation, porosity/void formation and fiber orientation and large amount of moisture. These issues can be solved by modifying the AM process parameters, hardware and feed stock quality. As the demand for more sustainable material options grows in the general global marketplace, natural fibers coupled with novel processing techniques (e.g., additive manufacturing) can lead to the advancement of many industries towards a smart eco-conscious future.

7 Conclusions

The main challenges that need to be addressed in order to increase the performance and consequently the use of NFRCs composites in industry are mainly the poor adhesion between natural fibers and matrix, fiber quality inconsistency, thermal stability and moisture absorption of natural fibers. Intense research efforts are continuously made and some of these challenges were addressed by recent advancements in fibers treatment and modification, exploration of new natural fibers and hybridization. The fiber modification techniques provide improved fibre-matrix interfacial adhesion, improved fibre roughness and wettability and depends on the particular fiber/matrix used and the composite application, while the hybridization methods provide flexibility in fibers selection for the materials properties according to the end-use application requirements. Exploration of new natural fibers, especially, natural fibers with higher strength and modulus, such as curaua or nano-scaled cellulosic fibers, offer a promising potential for expanding the application of these composites. In addition, can diversify fiber sources and reduce material cost. Although, hybridization of natural fiber composites with other natural fibers may not provide comparable

mechanical or thermal properties improvements to those hybridized with synthetic fibers, this approach may have noteworthy value in terms of sustainability. The hybridisation of natural fibers composites with other fibers characterised by low moisture absorption tendency is an effective method to improve the water absorption resistance of natural hybrid composites. Fiber's treatment and addition of fillers are also solutions to reduce moisture absorption of these composites. The application of natural and hybrid fibers composites, particularly in automotive, civil industry and sports is quite promising and will significantly increase in the future due to the significant pressure on reducing costs and increasing demand for material recyclability.

Conflicts of Interest: The authors declare that they have no conflicts of interest to report regarding the present study.

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