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Recent advances in development, characterization and joining of new sustainable materials

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Abstract. Nowadays, in order to increase the transport efficiency and reduce fuel consumption and emissions of contaminants, a reduction in weight associated with improved safety performance of the materials in use must be achieved. On the other hand, environmental concern has generated interest in research about new materials aligned with the principles of sustainability. Thus, this need for better performing and ecological structures has resulted in the development of a new variety of materials. Among these materials, currently, are the composites produced from resources of renewable sources. Natural fiber composites have recently attracted a great deal of attention by the industry due to their many attractive benefits (e.g., high strength-to-weight ratio, sustainable characteristics and low cost). Welding is highly impractical to use in these situations (i.e., thermoset polymer composites) and rivets or screws exhibit stress concentrations and offer a low fatigue resistance. Adhesive bonding is usually the preferred method since it allows for greater flexibility in design and is more efficient in mechanical and energy aspects. In this work, recent advances in development, characterization and joining of new sustainable materials, critical challenges and future perspectives are presented. The application of sustainable green composites may further increase in many structural and non-structural applications if their joining behavior is well-known and established.

Introduction

Global initiatives and agreements, such as the United Nations Sustainable Development Goals (SDGs) and the Paris Agreement, have highlighted the urgency of adopting sustainable practices in all aspects of human activity. Industries are increasingly recognizing that sustainable materials not only align with ethical and environmental objectives but also offer economic advantages, including cost savings, enhanced brand reputation, and access to emerging markets with environmentally conscious consumers. As a result, researchers, scientists, and engineers are actively engaged in developing innovative sustainable materials that exhibit improved performance characteristics while minimizing their environmental footprint.

Sustainable materials such as natural fiber composites (NFRCs) can be found in applications in many industrial sectors, such as: automotive, construction, marine industry, and sports, among others [1]. Moreover, the most promising applications of NFRCs are in the automotive industry. In general, they are used in non-structural car body parts, such as: door panels, package trays, hat racks, instrument panels, internal engine covers, sun visors, boot liners and oil air filters. However, they are evolving to be used in more structurally demanding parts such as seat backs, exterior underfloor paneling as well as anti-roll bars [2-4]. They can also be used in construction, and building applications like bricks, door panels, roofing sheets, furniture panels, etc.

The main natural fibers studied and used in the industry (e.g., jute, sisal, 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 studied [1, 5]. Both thermoplastic and thermoset polymers are used as matrices in natural fiber reinforced polymer composites [6]. There is an increased interest in the scientific community in using bio-based polymers in composites as combining these matrices with natural fibers produces “green composites” or “bio-composites”. An emerging trend is also found in the use of thermoplastic matrices of renewable sources, such as PLA, as a matrix material for NFRCs [7, 8]. This is crucial due to the fact that the classical thermoset matrix materials such as epoxy are dependent on fossil fuels.

As the global emphasis on sustainability grows, the demand for more sustainable (eco-friendly) materials with enhanced properties has intensified. This paper presents an overview of cutting-edge research and innovations in the development of more sustainable materials (NFRCs), their characterization methods, and the adhesive joining of these materials.

Development and Characterization of New Sustainable Materials

Natural-fiber-reinforced composites (NFRCs) vary greatly in their mechanical properties. Mechanical properties (e.g., tensile, flexural, and impact) are highly dependent on different factors such as fiber and matrix type, interfacial bonding between fiber and matrix, fiber dispersion orientation and processing, among others. By increasing their mechanical performance, the capabilities and applications of natural-fiber-reinforced composites will be expanded.

The most common mechanical tests used to characterize sustainable composites are the tensile, flexural and impact tests. Generally, the tensile strength of composites increases with fiber content up to a maximum value, then it tends to decrease. This is due to the higher mechanical properties of the fibers when compared to the matrices [2, 9]. Furthermore, it is well known that natural fibers contain moisture and when introduced into a hydrophobic matrix, the higher water absorption tendency of the natural fibers will lead to lower interfacial fiber/matrix adhesion and consequently lower mechanical properties of the NFRCs. One possibility to tackle this issue is the use of surface treatments [10]. The flexural properties of NFRCs are sensitive to a number of parameters, such as layup, ply angles, material type and stacking sequence [11]. This is due to the fact that the stresses increase from the neutral line and reach their maximum compressive and tensile stresses on the top and bottom of the test specimen, respectively. Impact resistance is a measure used to characterize the efficiency of the material during a rapid load event and is intrinsically linked with its toughness. The energy absorption may be derived via a penetration impact, residual properties after impact, and analysis of the damaged area after a non-penetration impact. Furthermore, the stacking sequence of the composite is a significant parameter when it comes to absorbed energy as it was observed that higher energy absorbing fiber layers on the outside of the composite enhances the overall impact resistance [12]. On the other hand, the interfacial quality of the fiber/matrix is highly significant as a low-quality interface will poorly transfer the loads and decrease absorbed energy. This issue can be improved by the hybridization with synthetic fibers.

Strategies for Improving the Performance of New Sustainable Materials

The main methods used to increase the performance of natural fiber-reinforced composites, such as: fiber treatment and modification, the use of filler materials (either on the fiber surface or into the matrix as the second reinforcing phase) and fiber hybridization) are briefly presented in this section.

Fiber Treatment and Modification

Surface treatment of natural fibers is usually performed to enhance their mechanical and thermal properties, before using them to manufacture a composite material. The fiber modification techniques provide improved fiber-matrix interfacial adhesion, improved fiber roughness and wettability and depends on the particular fiber/matrix used and the composite application [13]. It is fundamental to understand the interfacial properties and bonding mechanisms of fiber-matrix and this requires significant research efforts in order to maximize natural fiber composites applications [10, 13, 14].

Use of filler materials

Another method explored in the literature to improve the properties of natural fiber-reinforced composites is the use of filler materials (either on the fiber surface or into the matrix as the second reinforcing phase) [15-17]. Due to their high specific surface area, good chemical interaction with the hydrophobic matrix and crack arresting capabilities, a synergistic relationship is observed with NFRCs [18]. However, a careful determination of the fabrication and matrix/filler homogenization technique must be done in order to avoid filler agglomeration, which may lead to detrimental effects. The most commonly used synthetic nanofillers are: MWCNTs, metal oxides and titanium dioxide.

Neto et al. [17] investigated the effect of multi walled carbon nanotubes (MWCNTs) in the UV and water spray ageing process of natural fiber and hybrid composites. Jute and glass fiber plain weave bidirectional fabrics were used along with an epoxy resin matrix. The compression molding technique was used to fabricate the composite plates. The filler weight fraction was 0.6%. The mechanical properties were analyzed via tensile and flexural tests according to ASTM standards. The samples were aged at 500h and 1000h of exposure, respectively. They found that for the pure natural fiber composites an improvement in the mechanical properties was observed. However, for the hybrid composites, a slight decrease in tensile and flexural strength was observed. More recently, Neto et al. [19] studied the effect of the addition of the MWCNTs on the mechanical and thermal properties of curauá fiber reinforced composites. The MWCNTs were added either to the fiber surface (see Fig. 1) or into the resin matrix as the second reinforcing phase. They found that the incorporation of MWCNTs on the curauá fibers showed positive effects (an increase in properties was found for the MWCNT-modified fibers and their composites).

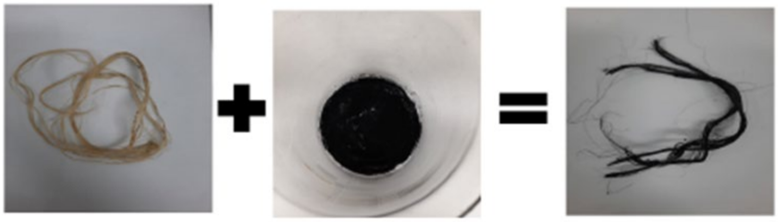


Figure 1. Schematic of the coating process of curauá fibers with MWCNT [19].

Fiber hybridization

One method to increase the mechanical performance of NFRCs to extend their applications is the fiber hybridization [1, 15, 20-22]. Generally, natural fiber reinforced hybrid composites are produced by hybridizing natural fibers with another either natural or synthetic fiber with superior properties (e.g., higher mechanical strength, chemical stability, nontoxicity, resistance to high temperatures, and thermal or acoustic insulation).

Fiber hybridization: use of intralaminar hybridization [21-23], use of interlaminar hybridization [24], use of 3D fiber-reinforcement architecture and use of multi-scale hybridization [15].

For example, for the case of intralaminar hybridization, Pereira et al. [23] used ramie, sisal and curauá fibers as natural fiber reinforcements for the hybrid composites based on sisal. They concluded that the mechanical properties are improved by the hybridization of sisal-based composites. On the other hand, they state that the hybridization did not significantly affect the thermal stability of the composites studied.

Interlaminarly hybridized composites of natural/synthetic fiber have been shown in the literature to present a non-linear enhancement of the material properties as the weight fraction of synthetic fibers increases. For example, Queiroz et al. [25], demonstrated that for hybrid composites reinforced with a symmetric synthetic glass envelope around a jute fabric core, the in-plane properties increased significantly as the number of glass layers increased from 2 to 3 (i.e., an approx. 35% enhancement in tensile strength). This is due to the much higher density of synthetic fibers present in a woven fabric compared to the in-plane intralaminar hybridization. On the other hand, Queiroz et al. [26] recently demonstrated that the effect of 3D reinforcement of hybrid natural composites (i.e., z-binder fibers stitched in the transverse direction through intralaminar 2D fabrics) is not significant in the in-plane properties. The highest variation observed in the composite tensile strength (when the 2D case is compared to the 3D) was of approx. 15%, while for most of the studied cases it was below 10%. Therefore, differently from the interlaminar reinforcement, significant variations will not be observed in the in-plane composite properties but rather in the transverse properties (strength and toughness).

3D Printing with Natural Fibers

Another area currently developing fast is the application of natural fiber as reinforcements in composites produced by additive manufacturing (AM or 3D printing) [27-32]. This technology allows for the fabrication of complex geometries without the need for expensive tooling and molds. It also allows for precise control over material distribution, opening up new possibilities for complex geometries and customizable structures [33, 34].

The primary materials utilized in Fused Deposition Modeling (FDM) are acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA). However, in their unmodified state, these materials exhibit relatively modest mechanical properties, characterized by typical bulk strengths ranging from 30 to 100 MPa and elastic modulus within the 1.3–3.6 GPa range [27]. Consequently, parts produced through FDM using these materials are not suitable for high-performance applications. To address this limitation, diverse methodologies have been investigated in the literature to enhance their properties. Examples include the incorporation of nanoparticles into the material matrix and the utilization of fibers or fabrics as reinforcements. These strategies aim to enhance the mechanical characteristics, thereby elevating the suitability of FDM-printed composites for applications demanding higher performance levels.

The research of our group in this area focuses on different key aspects: studying the effect of filament type and printing parameters on the mechanical properties of AM parts [28], the mechanical performance of AM parts reinforced with natural (jute and curauá) and synthetic (glass) fibers [30], developing natural fiber/filler-reinforced PLA and ABS composites through fused deposition modelling [31, 32] and exploring the mechanical performance of adhesive joints using various fiber reinforcements for AM parts [29].

For example, the use of natural fibers as filament reinforcement was investigated by Cavalcanti et al. [31] by using short curauá fibers with different lengths (3, 6, and 8 mm), and concentrations in terms of weight percentage (2, 3.5, and 5 wt. %) to fabricate polylactic acid (PLA) filaments which were subsequently used to fabricate composites via fused deposition modelling. They concluded that the curauá fiber-reinforced PLA composites may be a promising innovation to improve the performance of these materials, which might enable them to be used for new applications. However, some challenges remain to be solved in the fabrication of the reinforced filaments, such as difficult quality control due to voids and porosities, among others.

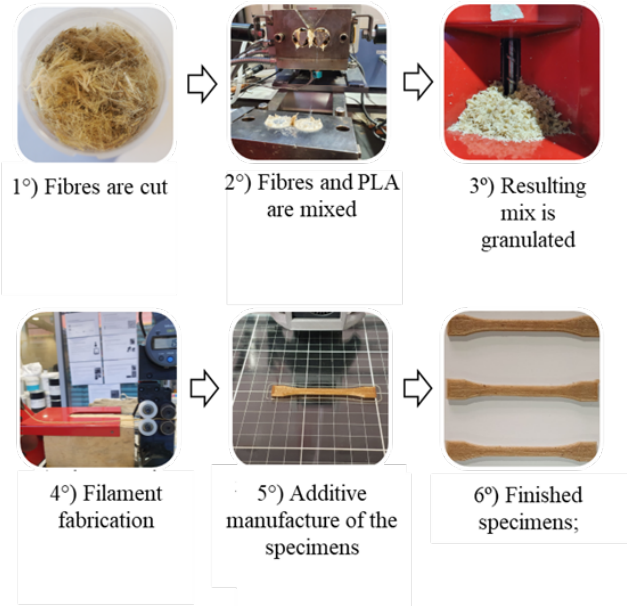


Figure 2. Schematic of the fabrication of the reinforced filaments [31].

In another study of our group [30] the mechanical properties of composite materials made by 3D printed (PLA and ABS) parts reinforced by different types of fibers was studied. Natural (jute and curauá) and synthetic (glass) fiber fabrics were laminated on the outer sides of the AM parts by using an epoxy resin via compression moulding. The reinforcement was applied to the AM parts as 1 and 2-layers for each fiber type. The mechanical characterization of the resulting composites was performed through tensile and flexural tests in accordance with their respective ASTM standards. This work provides an alternative to the more common reinforcement methods, by using a hybrid manufacturing method: 3D printing+ compression moulding (see Fig 2). The application of novel manufacturing methodologies allied with renewable biodegradable fibers can provide an interesting path forward for fabricating high-performance 3D printed thermoplastic composites.

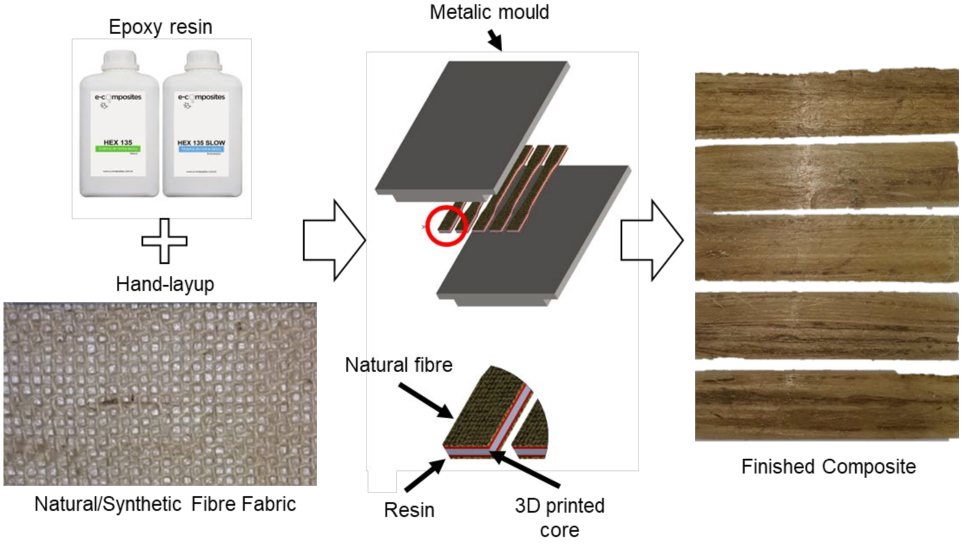


Figure 3. Schematic of the hybrid manufacturing method: 3D printing+ compression molding.

Adhesive Joining of New Sustainable Materials

The joining of natural fiber composites to larger structures, such as automotive interiors, is also of crucial importance [12, 35]. This field is still in its infancy and little research has been done. One of the main issues facing the joining of sustainable materials is that natural fiber reinforced composites present significantly lower mechanical properties when compared to their synthetic counterparts. This is a significant drawback, and it hinders the bonded joint performance of these materials. Therefore, it is crucial to enhance the mechanical properties of sustainable materials prior to bonding them. The reason the adherend material properties of composites are vital in a bonded joint is that due to the nature of the global geometry of the commonly used single lap joint (SLJ), an out-of-plane stress develops due to the eccentricity of the load path between the adherends (see Fig. 4). This peel stress is responsible for the crack initiation at the overlap edges where the stresses (both peel and shear) are highest. However, the propagation and path of the crack is controlled by other parameters, such as composite transverse strength and toughness. Furthermore, adherend stiffness will significantly impact its rotation tendency during the loading of the SLJ, either increasing or decreasing the peel stresses. This leads to the failure mode of delamination which is common for composite bonded joints and is characterized by the removal, partial or complete, of one or more layers of the composite. So far, the hybridization technique has been shown to be the most effective in enhancing the material properties of these more environmentally friendly materials, usually by employing a small weight fraction of synthetic fibers. This hybridization is done in the form of distinct architectural techniques, such as: intralaminar, interlaminar and 3D architectures. The intralaminar reinforcement architecture is defined by the application of two or more reinforcement fiber phases within the same fabric layer (see Fig. 5a). This architecture is more complex and labor intensive, but a highly tailored composite architecture can be achieved. On the other hand, the interlaminar reinforcement architecture is simpler, where each layer is comprised of a single fiber type, usually a fabric (see Fig. 5b). This arrangement has the advantage of being able to leverage the synthetic fiber's high water barrier properties to keep moisture from entering the composite [12, 36]. This is particularly relevant for natural fibers which have a high hydrophilic tendency.

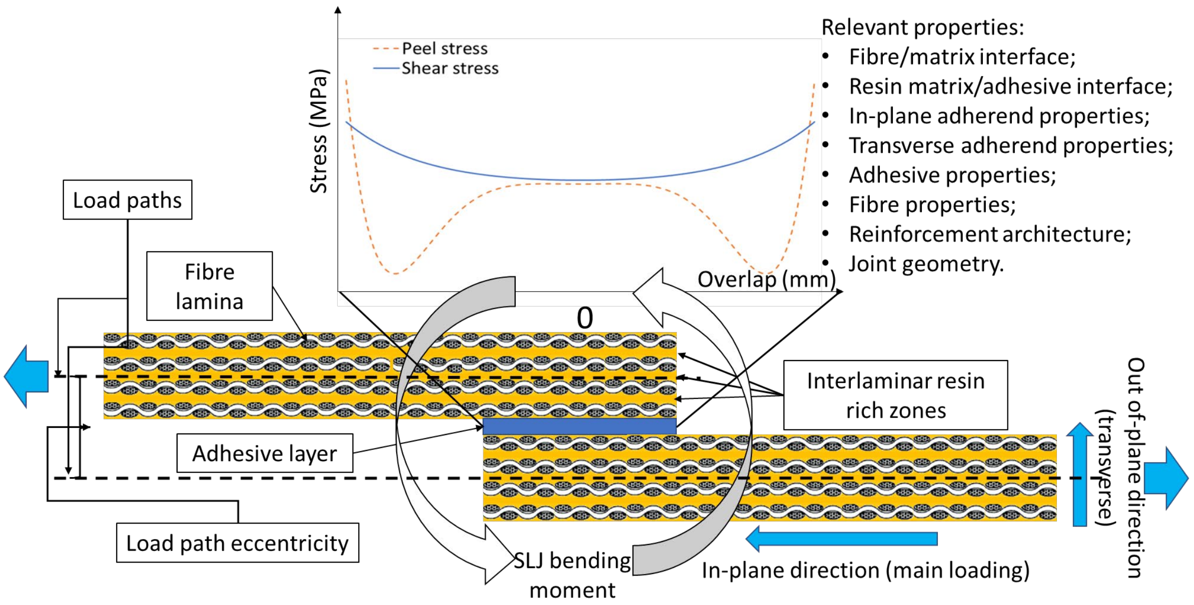
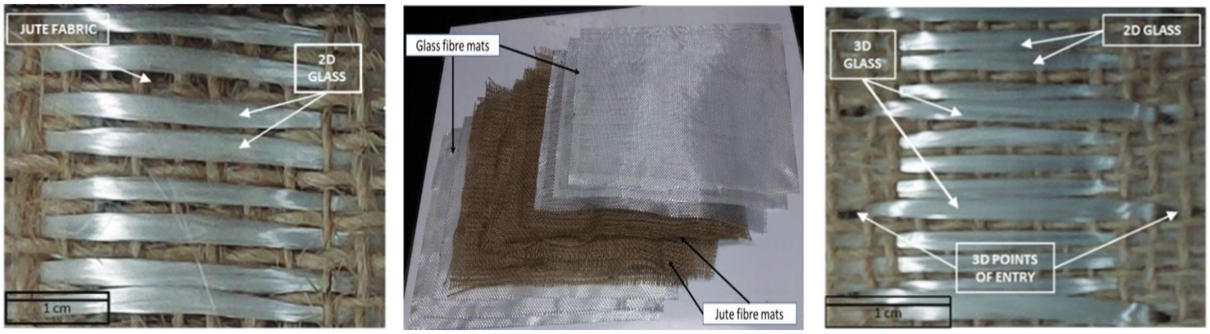


Figure 4. Schematic of composite bonded joint stress state and relevant parameters.



a) b) c)

Figure 5. Types of architectural reinforcement, a) intralaminar, b) interlaminar and c) 3D.

Another crucial material property of a composite to be bonded is the transverse strength and toughness. Due to the much lower transverse strength, the delamination failure is directly linked to the peel stresses exceeding the strength and a crack initiating and propagating in the interlaminar space. Therefore, the technique of 3D reinforcement comes into play to solve this issue by applying a z-binder fiber phase that binds the fabric layers in the transverse direction, significantly increasing the composite transverse strength and delamination toughness (see Fig. 5c).

Our group is responsible for the most relevant research efforts to date in the effect of natural fiber composite architectures in bonded joint performance [4, 12, 24-26]. The effect of intralaminar reinforcement in the bonded joint performance of hybrid composites was analyzed by Queiroz et al. [4]. Jute and sisal fabrics were used as a base fiber phase for intralaminar reinforcement via weaving with glass fiber to fabricate hybrid composites. Neat jute, sisal and glass fiber reinforced composites were also fabricated via compression molding. A bi-component epoxy matrix was used to fabricate the composites. Single lap joints, both similar and dissimilar, were fabricated and tested in quasi-static tension. An automotive grade epoxy adhesive as well as the epoxy matrix were used as adhesives. It was found that it is possible to reach up to approx. 77% of the bonded joint failure load of the neat glass composite (GFRP) via the glass fiber intralaminar hybridization of jute fabrics.

In a follow-up study by Queiroz et al. [25], the interlaminar reinforcement technique was used to improve the bonded joint performance of jute/glass fiber composites. Glass and jute fiber fabrics were used and the glass layers were placed on the outside of a jute fiber core of 5 layers. The number of exterior glass fiber layers varied from 2 to 3. The architecture was symmetric. An epoxy matrix was used along with a structural automotive bi-component epoxy adhesive. Single lap joints were fabricated and tested. They found that a hybrid interlaminar glass/jute composite with 3 glass layers was able to reach approx. 100% of the bonded joint failure load of a fully synthetic glass composite (GFRP). This was linked to the optimum balance of improved adherend material properties.

Finally, in a recent research effort, Queiroz et al. [26], investigated the effect of a novel 3D reinforcement architecture in the bonded joint performance of hybrid composites. Jute bidirectional fabrics were used as well as sisal, curauá, hemp and glass fibers in order to fabricate intralaminarly reinforced 2D fabrics. These fabrics were employed with the aid of a mold to fabricate 3D fiber-reinforced preforms by weaving a z-binder fiber through 5 2D fabrics. The composites were fabricated via the compression molding technique with an epoxy matrix. Single lap joints were fabricated with a modern automotive grade epoxy adhesive. It was found that the 3D reinforcement successfully enhanced the bonded joint performance of the novel sustainable composite materials for both the sisal and curauá specimens. This also marks the first time in the literature where a fully natural fiber reinforced composite joint matched approx. 100% the bonded joint performance of a fully synthetic joint (i.e., GFRP and CFRP).

Conclusion

The increasing importance of sustainable materials is a response to the pressing need to address global environmental challenges. This background sets the stage for a deeper exploration of recent advances in the development, characterization, and joining of new sustainable materials, as industries seek transformative solutions to create a more sustainable and resilient future. This paper reported the recent advances that have been made in this field. The main take away can be summarized as follows:

- The application of natural fiber composites, particularly in automotive, civil industry and sports is quite promising and will continually increase in the future due to the significant pressure on reducing costs and increasing demand for material recyclability.
- The future of natural fiber composites is likely to witness an increased focus on hybridization, combining different types of natural fibers or natural/synthetic fibers to create synergistic composites. This approach can enhance mechanical properties, durability, and tailor the materials for specific applications.
- The hybridization technique has clearly demonstrated its ability to improve the natural fiber composites adherend properties and the bonded joint efficiency at relatively low fabrication complexity. In addition, synthetic interlaminar hybridization also provides benefits by acting as a barrier to moisture, increasing the service life of natural/synthetic fiber reinforced composite structures. Finally, NFRC joints can be a viable replacement for synthetic composite joints at no load-bearing loss.

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