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REVIEW

# An overview of structural-functional-integrated composites based on the hierarchical microstructures of plant fibers

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## Abstract

Plant fiber-reinforced composites have raised great attention among materials scientists and engineers during the past decade. Most of the efforts were put on the interfacial modifications to improve the mechanical properties of the composites so that they could partly replace the currently largely used glass fiber-reinforced composites. The modifications were mainly focused on the surface treatment of plant fibers so that mechanical or chemical bonding between plant fibers and polymeric matrices could be set up. However, the unique hierarchical microstructures of plant fibers make the building up of multiscale interfaces possible so that more forces or energies would be needed to fracture the plant fiber-reinforced composites. The additional hollow structures could also bring benefits for sound absorption and damping properties. Therefore, this article reviewed R&D efforts to develop structural and functional-integrated plant fiber-reinforced composites by fully taking advantage of the hierarchical microstructures of plant fibers. Firstly, the unique hierarchical structures of plant fibers were revealed and hierarchical theoretical models for mechanical properties were discussed. Then, the modification, characterization, and evaluation of plant fibers in terms of their interfacial properties with polymeric matrices, especially by nanotechnologies with the consideration of their unique hierarchical microstructures, were reviewed. Finally, the design and manufacture of quasi-structures and structural-damping components using technologies that have been fully adapted to state-of-the-art industrial processes for use in critical applications, such as aircraft interiors, rail transportation vehicles, and constructions, were also introduced.

**Keywords** Plant fibers · Polymer-matrix composites (PMCs) · Mechanical properties · Structural-functional integration

## 1 Introduction

With the present state of composite technological development, plant fiber-reinforced composites (PFRCs) have raised great interest among materials scientists and engineers due to their potential mechanical properties and superior acoustic and damping properties caused by their unique hierarchical microstructure and chemical compositions [1–3]. Initially, PFRCs by using non-woven forms of plant fibers were developed in automotive sectors given their low density, low cost, and environmentally friendly characteristics to these parts [4]. Manufacturers, however, have since learned that these materials offer both structural and damping benefits [5–7]. This may render them an economical alternative to glass fiber-reinforced composites (GFRCs) for quasi-structural or structure-function-integrated applications in some critical sectors such as those of aircraft and ground transportation [8–11].

In the aviation industry, carbon fiber-reinforced composites (CFRPs) prevail as cutting-edge structural materials used for the development of primary and secondary structures, and GFRCs are typically used as secondary interior and exterior structures. However, new environmental regulations and societal concerns have triggered the searching for new products and processes that are functional and yet compatible with resource-related and environmental requirements [12, 13]. It is now envisaged that, in parallel with the further development of state-of-the-art CFRCs and GFRCs, the incorporation of bio-resources into

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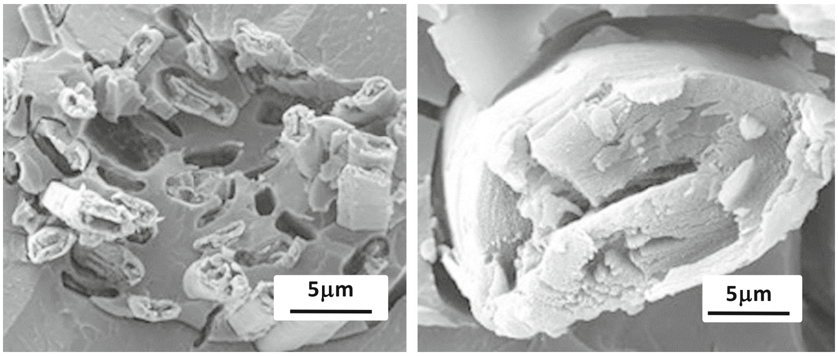


**Fig. 1** SEM micrographs of the cross section(s) of **a** several ramie fibers and **b** a single ramie fiber embedded in epoxy [23, 24]

composite materials may not only reduce further dependency on petroleum reserves but also contribute to the development of lightweight, noise- and vibration-reduction structures for aircraft. However, major barriers for their applications pertain to limited mechanical and thermophysical properties [14, 15], poor interfacial bonding between fibers and matrix resins [16–18], and poor resistance to moisture aging and flammability features compared to the state-of-the-art GFRCs [19, 20].

This paper reviewed the recent development of structure-function-integrated plant fiber-reinforced composites by fully taking advantage of the unique hierarchical microstructures of plant fibers. The efforts of a joint research consortium between Chinese universities and the Chinese Aviation Industry (AVIC) under the support of the national basic research program of China (973 program) were also gathered [21]. The fundamental features and the potential of PFRCs as environmentally friendly alternative materials for use in quasi-structural and function-integrated applications in aeronautic, rail transportation, and construction sectors were explored. An overview of this paper is presented with a special focus on the following topics and issues: interface characterization and modification of PFRCs, mechanical and damping properties of laminated composites, and case studies for their applications.

## 2 Uniqueness of plant fibers

Plant fibers, when used as reinforcing components of composites, are inexpensive, lightweight, and biodegradable and present unique mechanical, acoustic, and damping properties owing to their chemical and structural characteristics.

### 2.1 Hierarchical and lumen structures

#### 2.1.1 Microstructures

Compared to man-made fibers, plant fibers are non-uniform with irregular cross sections. They are regarded as a composite material with three components (cellulose, hemicelluloses, and lignin), in which the unidirectional cellulose microfibrils constitute the reinforcing elements in the matrix blend of hemicellulose and lignin [22]. Ramie, which is characterized by oval-shaped cross sections, serves as one of the typical plant fibers in this study (Fig. 1a, b). Ramie fiber, commonly known as China grass, is one of the oldest natural textile fibers grown primarily in China. As noted above, the surfaces of ramie fibers are naturally coarse and bumpy. A single ramie fiber is made up of a single hollow cell fiber, with average diameters between 10 and 50 μm. Thousands of nanosized fibrils compose the cell wall of the fiber.

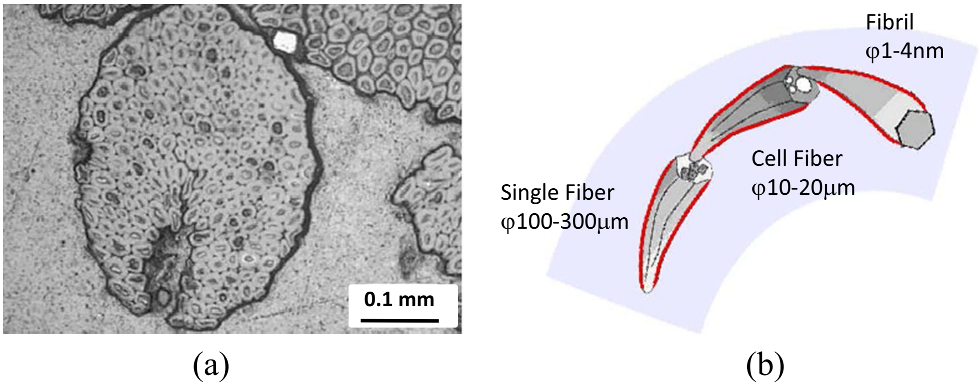
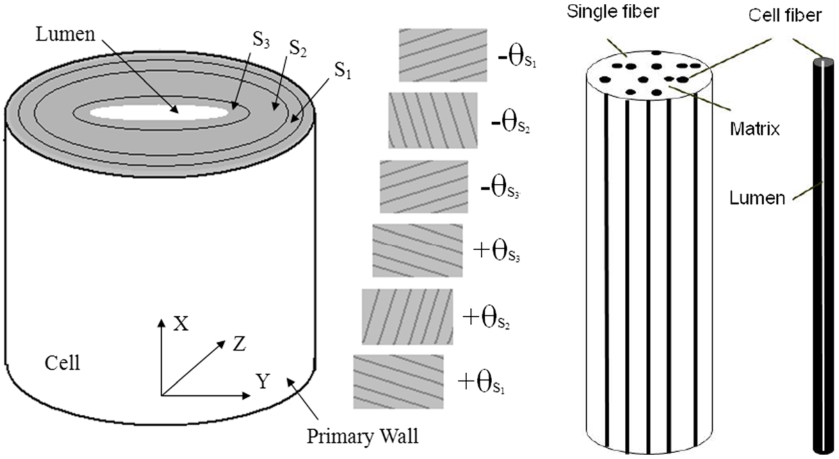


**Fig. 2** **a** Cross section of a single sisal fiber and **b** schematic drawing of the multiscaled hierarchical structure of sisal fiber

**Fig. 3** Hierarchical models for plant fibers **a** at the cell fiber level and **b** at the single plant fiber level



It should be noted that, except for ramie fiber, the rest of the plant fibers are all made up of a bundle of cell fibers [23].Taking sisal fiber as an example, from Fig. 2, it can be seen that a single sisal fiber is made up of a bundle of cell fibers with diameters of 10–20 μm. A cell fiber contains a lumen inside and its cell wall is made up of many fibrils, diameters ranging from 1 to 4 nm. Furthermore, the microfibril is composed of alternating crystalline and amorphous regions. The crystallite size is approximately 5–30 nm in a lateral direction and between 20 and 60 nm along the axis [25]. Therefore, it is clearly seen that plant fibers possess multiscaled hierarchical microstructures and the smallest structure is in nanosize.

### 2.1.2 Hierarchical models for plant fibers

The unique hierarchical structures of plant fibers brought new challenges for the mechanics and the mechanical properties of plant fibers by considering their unique structures have been studied by many researchers during the past decade [26–28].

**Elastic property** A multiscaled hierarchical theoretical model to calculate the elastic properties of a single plant fiber was proposed and compared with the experimental results [24]. At the cell fiber level, a laminated plate model proposed by Salmon et al. was employed, as shown in Fig. 3a [29]. A cell fiber was considered as a laminated plate of layers primary wall, S1, S2, and S3 of cellulose fibrils with different angles to the fiber axis. Thus, the cell fibers could be considered as anti-symmetric laminates by using the classical laminate theory for estimation of their elastic properties. The parameters used for the calculation, such as the elastic properties ($E_{11}$, $E_{22}$, $G_{12}$, $\gamma_{12}$) of the fibril and the matrix, the volume fraction of the fibrils, and the angle to the axis of the fibril (herein spiral angle), are given in Tables 1 and 2, respectively. The relative thickness of each layer was chosen as $P = 8\%$, $S1 = 8\%$, $S2 = 76\%$, and $S3 = 8\%$ [30].

**Table 1** Elastic properties of the constituents of plant fibers [8, 26, 27]

| Constituents | $E_{11}$ (kN/mm$^2$) | $E_{22}$ (kN/mm$^2$) | $G_{12}$ (kN/mm$^2$) | $\upsilon_{12}$ |
|---|---|---|---|---|
| Cellulose | 74–168 | 27.2 | 4.4 | 0.1 |
| Semi-cellulose | 8 | 4 | 2 | 0.2 |
| Lignin | 4 | 4 | 1.5 | 0.33 |
| Pectin | 1.6 | 1.6 | 0.62 | 0.3 |

Considering the existence of a lumen inside a cell fiber, a conversion coefficient, which was determined by the lumen ratio, was employed for the calculation. The lumen ratio was measured by image analysis of optical micrographs with the aid of the image analysis software Quantlab-MG. The calculated and converted elastic modulus of cell fibers were thus obtained and shown in Table 3.

The second scale model was set based on a single plant fiber and is shown in Fig. 3b. A single plant fiber could be regarded as a composite with parallel cell fibers reinforced in a pectin matrix. Thus, the Halpin-Tsai equation [31] was employed to calculate the elastic modulus of a single plant fiber as expressed by Eq. (1).

$$\frac{M_{\mathrm{c}}}{M_{\mathrm{m}}} = \frac{1+\xi\eta V_{\mathrm{f}}}{1-\eta V_{\mathrm{f}}} \qquad \eta = \frac{\frac{M_{\mathrm{f}}}{M_{\mathrm{m}}}-1}{\frac{M_{\mathrm{f}}}{M_{\mathrm{m}}}+\xi} \tag{1}$$

where ξ is a factor determined by the fiber geometry and can be calculated by Eq. (2). $M$ and $V$ are Young's modulus and volume fraction, respectively. The subscripts c, f, and m represent composite, fiber, and matrix, respectively.

$$\xi = 2L/D \tag{2}$$

where $L$ refers to the length of the fiber and $D$ is the thickness or diameter of the fiber. For a continuous reinforcement in the length direction, this factor $L/D$ approaches infinity and the Halpin-Tsai equation reduces to the rule of mixture (ROM).

**Table 2** Chemical composition and spiral angle of different cell fibers [9–11, 22, 26, 29]

| Fiber | Cellulose (%) | Hemicellulose (%) | Lignin (%) | Pectin (%) | Spiral angle (°) |
|---|---|---|---|---|---|
| Ramie | 68.6–76.2 | 0.6–0.7 | 13.1–16.7 | 1.9 | 8 |
| Jute | 61–71 | 13.6–20.6 | 12–13 | 0.4 | 8 |
| Flax | 71 | 2.2 | 18.6–20.6 | 2.3 | 10 |
| Kenaf | 60.8 | 20.3 | 11 | 3.2 | 6–10 |
| Hemp | 70.2–74.4 | 3.7–5.7 | 17.9–22.4 | 0.9 | 6 |
| Sisal | 67–78 | 10–14.2 | 8–11 | 10 | 20 |

In this study, the volume fraction of a single plant fiber was again obtained by analyzing the optical micrograph with the aid of Quantlab-MG. The longitudinal modulus of the cell fiber was derived from the abovementioned laminate plate model. A single fiber was considered as a bundle of continuous cell fiber-reinforced composite which means factor *L*/*D* approaches infinity. Therefore, identical estimation using either the Halpin-Tsai equation or ROM was obtained and listed in Table 4. A good agreement with the experimental results was observed.

**Strength** A fiber bundle model based on probabilistic fracture mechanics was generated to determine the tensile strength of plant fibers with hierarchical microstructures. Weibull distribution analysis on the tensile strength obtained from single fiber tensile tests was performed. Then, the probabilistic density function of a single plant fiber with countless cell fibers was determined. A modification on the tensile strength of plant fibers from countless cell fibers to a limited number of cell fibers was carried out. By derivation, the formula for the tensile strength of a single plant fiber was given by Eq. (3).

$$\overline{\sigma_b^N} = \alpha\gamma\sigma_{\mathrm{f}} \tag{3}$$

where

$$\alpha = \left[\beta^{\frac{1}{\beta}}\exp\left(\frac{1}{\beta}\right)\Gamma\left(1+\frac{1}{\beta}\right)\right]^{-1}$$

$$\gamma = 1 + 0.996N2^{-2/3}e^{\frac{2}{3\beta}}\beta^{-\frac{1}{3}}$$

$\sigma_{\mathrm{f}}$ denotes the tensile strength of a cell fiber and can be obtained based on the tensile strength of ramie fibers since a single ramie fiber itself is a cell fiber. $\beta$ is the shape parameter of the Weibull distribution for the strength of single cell fibers. $N$ denotes the number of cells within a single fiber. For a brittle fiber, the value of $\beta$ typically ranges between 2 and 4. Thus, for different values of $\beta$, strengths of different plant fibers were calculated and are shown in Table 5. The results were also compared with the experimental results, and a high level of agreement was found.

**Table 3** Elastic modulus of cell fibers [24]

| Fiber | Ramie | Jute | Kenaf | Sisal |
|---|---|---|---|---|
| Calculated elastic modulus (GPa) | 42.1–91.8 | 35.6–77.2 | 34.1–73.9 | 31.7–66.0 |
| Lumen ratio (%) | 16.7 | 24.3 | 28.6 | 39.2 |
| Conversion coefficient (%) | 83.3 | 75.7 | 71.4 | 60.8 |
| Converted elastic modulus (GPa) | 35.1–76.4 | 26.9–68.3 | 24.3–52.8 | 19.2–40.1 |

**Table 4** Elastic modulus of single plant fibers

| Fibers | Ramie | Jute | Kenaf | Sisal |
|---|---|---|---|---|
| Experimental values (GPa) | 14.05 | 29.30 | 29.89 | 9.56 |
| Calculated values (GPa) | 12.8–27.8 | 21.7–55.0 | 20.9–45.5 | 8.79–18.4 |

### 2.1.3 Acoustic properties

The hollow lumen structures of plant fibers also lead to higher damping and sound absorption properties over man-made fibers which made structural-functional-integrated structures possible. Take acoustic properties as an example; the sound absorption coefficients of plant fibers measured with the aid of an impedance tube are much superior than those of man-made fibers, like glass or carbon fibers as shown in Fig. 4 [32].

## 2.2 Poor interfacial bonding between plant fibers and polymers

It is widely acknowledged that the inherent polar and hydrophilic nature of lignocellulose fibers and nonpolar hydrophobic characteristics of most polymeric resins lead to poor interfacial bonding between plant fibers and matrix resins, impairing mechanical properties of the composite. Silane, permanganate, and alkali are standard chemicals used to pre-treat plant fibers to improve interfacial properties [33–36]. Chemical treatments with $KMnO_4$, NaOH, and 3-aminopropyltriethoxy silane gradually rendered the surface of plant fibers scaly, rough, and bumpy [37]. Interfacial shear strength was thus improved considerably by these pre-treatments as shown in Fig. 5, with silane treatment generating the highest values for the fiber types tested due to the chemical bonds set up between plant fibers and the polymer matrix [38].

**Table 5** Comparison between tensile strengths derived from fiber bundle model and experimental tests

| Fiber | Ramie | Kenaf | Jute | Sisal |
|---|---|---|---|---|
| Theoretical (MPa) | 938 | 477–573 | 500–603 | 412–511 |
| Experimental (MPa) | 938 | – | 393–773 | 511–635 |

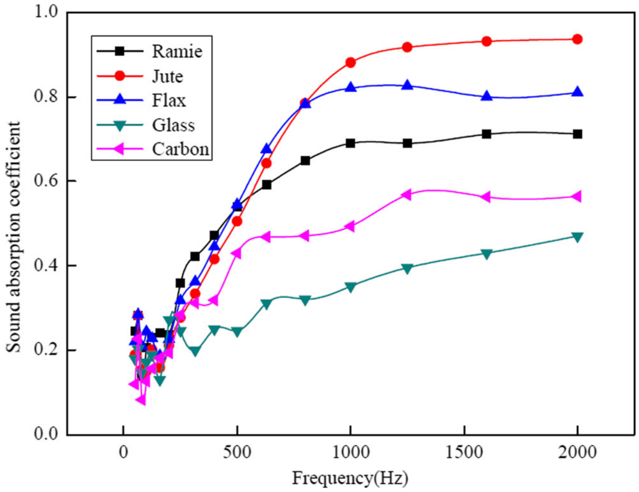


**Fig. 4** Sound absorption coefficients of plant fibers, glass fibers, and carbon fibers

By taking advantage of the nanoscaled structure of fibrils of plant fibers, carbon nanotubes (CNTs) were grafted onto the surfaces of flax yarns [39]. The CNTs used were carboxyl-functionalized multiwall carbon nanotubes, and the –COOH content was 2.56 wt% which may form strong hydrogen bonds between the modified CNTs and the fibers. It can be seen from Fig. 6 that CNTs were uniformly dispersed and randomly oriented on the surface of the fibers. It was also noted that some of the CNTs were inserted into the flax fibers with their ends left outside. The stiff CNTs can be easily inserted into the soft cell fibers and also between the twisted cell fibers. Therefore, the IFSS of CNT-coated flax yarn-reinforced epoxy composites were considerably higher than that of the control group, which is 55 MPa for the yarns coated with 1.0 wt% CNTs.

Figure 7a–d shows the optical microscope images of the matrix holes being left after the flax yarns were pulled out. A clear transition of the fracture mode was observed. More fibrils were attached to the boundary of the hole for the CNT-coated flax yarns. From the observation of the pulled-out flax yarns, hierarchical fractures were clearly seen from the fiber-coated by CNTs (Fig. 7f, g). The fracture mode for pulled-out flax yarns changed from direct debonding to fibrillation of the flax fibers. It can be realized that there are multiscaled interfaces between flax yarns and epoxy resin, which are between the fiber and matrix and between fiber cell walls. Since CNTs can reinforce the interface between the flax yarn and epoxy resin to a relatively high level, fibrillation could occur leading to the peeling and splitting of cell walls, which eventually resulted in an improved IFSS.

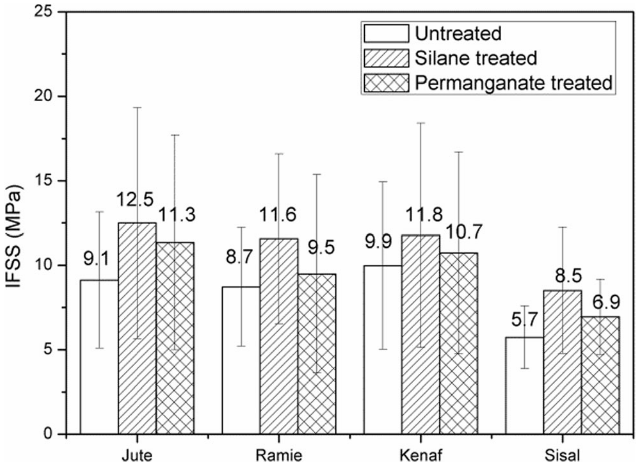


**Fig. 5** Interfacial shear strengths of different plant fiber-reinforced polyester composites via the two surface treatments by the single fiber pull-out test [38]

From the above discussion, it can be concluded that the fracture modes of the plant fiber-reinforced composites possessed hierarchical characteristics, from meso to micro and even to nano scale, which are very different with man-made fiber-reinforced composites (Fig. 8). Therefore, fully taking advantage of the hierarchical fracture modes of plant fiber-reinforced composite may lead to different desired mechanical or even functional properties.

Even though the improvement of the interfacial properties between CNT-modified plant fibers and polymer matrices was achieved by taking advantage of the hierarchical microstructures of plant fibers, the dispersion of these nanomaterials at the interface of the composites needs to be optimized so that even bigger improvement could be expected. Using bio-based nanocellulose to replace CNTs to modify the interfacial properties of plant fiber-reinforced composites may be also a good attempt to achieve high performances.

## 3 Enhancement of mechanical properties of plant fiber-reinforced composites via hierarchical fractures

### 3.1 Enhancement of mechanical properties of plant fibers

Attempts have recently been made to explore the use of nanoparticles for the modification of plant fibers by taking advantage of their hierarchical microstructures to rectify the limitations of enhancing the mechanical properties of PFRC by chemical methods.

In this study, zirconia dioxide nanoparticles were designed to graft onto flax fibers. Hydrous zirconia nanoparticles were synthesized via $ZrOCl_2$ solution hydrolysis and were grafted onto the surfaces of the fibers through hydrogen bonds between abundant hydroxyl groups on flax fibers and $ZrO_2 \cdot nH_2O$. HCl and ammonium were added to adjust pH values and in turn the size of synthesized nanoparticles [40]. Flax yarns were surface-modified by hydrous zirconia nanoparticles through a simple chemical process involving the hydrolysis of zirconium oxychloride solution under mechanical

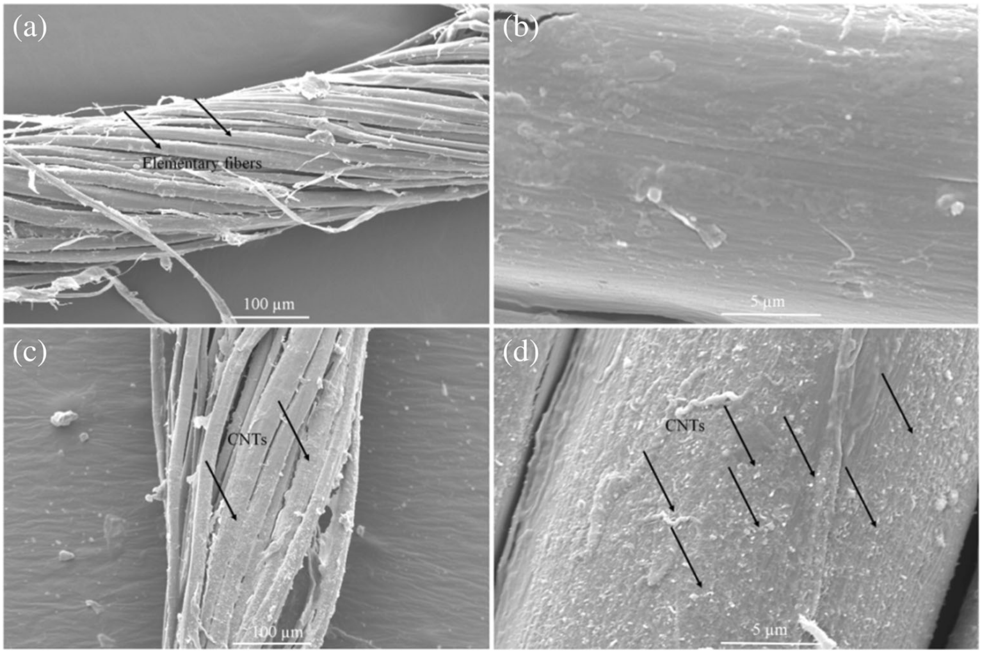


**Fig. 6** Surface morphologies of flax yarn at different magnifications. **a** and **b** untreated, **c** and **d** 1.0 wt% CNTs [39]

stirring and ultra-sonication conditions. From SEM observations, grafting assisted with stirring or sonification resulted in more homogenous distribution of nanoparticles on the surfaces of the fibers.

Table 6 compares tensile strength, Young's modulus, and elongation at the break of the untreated and grafted fibers obtained from a single fiber tensile test. It is clear that $ZrO_2 \cdot nH_2O$ grafting effectively increased tensile strength due to the removal of fiber surface defects [41]. The maximum increase of tensile strength can be as high as 81%. However, the tensile modulus of flax fibers, presented in Table 6, was not influenced by the treatment. This is reasonable as zirconia nanoparticle grafting was mainly found on the surfaces of the fibers. While the modulus was mainly dependent on cellulose

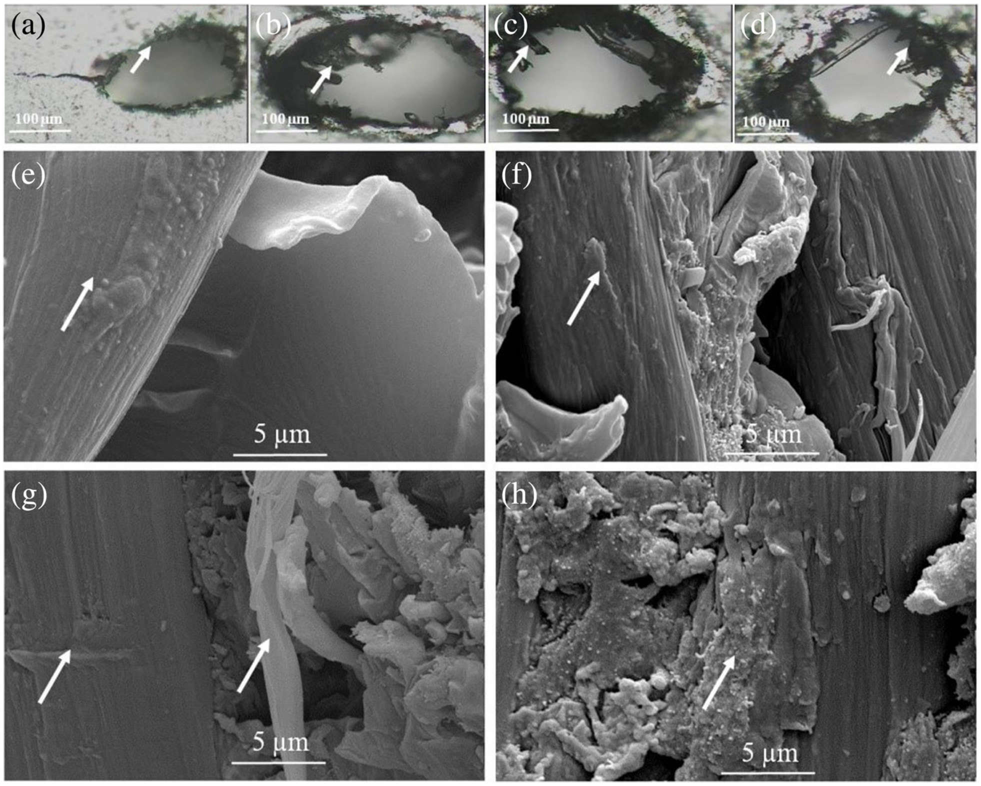


**Fig. 7** Morphologies of both epoxy matrix and flax yarn after single yarn pull-out tests. **a** and **e** untreated, **b** and **f** 0.5 wt%, **c** and **g** 1.0 wt%, and **d** and **h** 2.0 wt% CNTs [39]

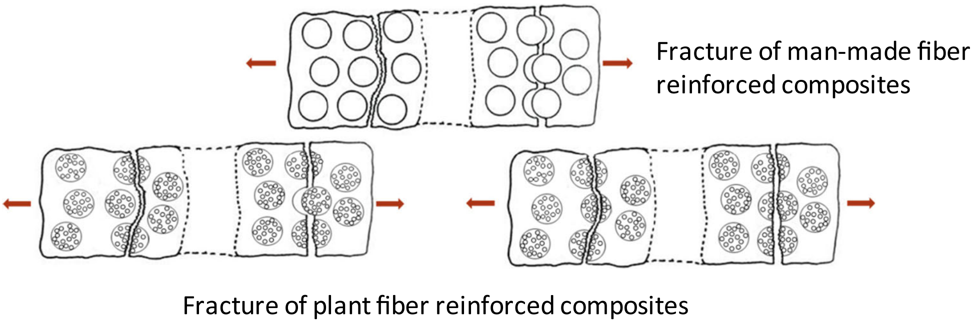


**Fig. 8** Comparisons of the fracture modes of PFRCs and man-made fiber-reinforced composites

fibrils in the cell wall, which was not affected by the present treatment as expected.

Enhancing the mechanical properties of plant fibers by coating carbon nanotubes onto plant fibers was also conducted [42]. Figure 9 shows the changes of Young's modulus and tensile strength versus diameters of coated and uncoated flax yarns obtained by single fiber tensile tests. It can be seen that coating CNTs onto flax yarns increased their Young's modulus. The flax yarns coated by 1.0 wt% CNTs possessed nearly the double value of modulus compared to that of the control group due to the existence of the CNT component which possesses high modulus. However, the strength of flax yarns was not noticeably affected. The unique yarn structure formed by the twisted short discontinuous single fibers might be the reason. Slippage between short single fibers by the untwisting of the yarns was the main failure mode of the yarns. As a result, CNTs did not play a role in the failure of the yarns.

From the above studies, it can be seen that both the strength and modulus of plant fibers could be greatly affected by the incorporation of the nanomaterials. However, manufacturing plant fibers or yarns specifically used for the reinforcement of composite materials is very necessary for achieving high-performance plant fibers.

## 3.2 Enhancement of mechanical properties of the composites

By taking advantage of the unique hierarchical microstructures of plant fibers, enhancement of the mechanical properties of PFRCs was attempted by incorporating carbon nanotubes into the multiscaled structures of PFRCs. Three ways of adding multiwall carbon nanotubes (MWCNTs) to the composite laminates were attempted, which were mixing CNTs with the resin [43], interleaving buckypaper (BP) between fabric layers [39], and coating CNTs onto flax fibers using a "soaking or spray-drying" process [42] as shown in Fig. 10.

**Table 6** Tensile properties of untreated and grafted flax fibers

| | Tensile strength (MPa) | Young's modulus (MPa) | Elongation at break (%) |
|---|---|---|---|
| Untreated fiber | 399.7 | 30.4 | 1.56 |
| Fiber grafted with non-stirring | 512.9 | 32.8 | 1.82 |
| Fiber grafted with stirring | 450.3 | 29.2 | 1.43 |
| Fiber grafted with sonication | 723.4 | 33.6 | 1.69 |

The addition of 0.6 wt% of MWCNTs by mixing them with the matrix was found to enhance interlaminar shear strength (ILSS), flexural strength, and flexural modulus by roughly 38, 34, and 37%, respectively, as shown in Fig. 11. Observations of the fractured surfaces of the composites from Fig. 12 indicated that the unique hierarchical microstructure of plant fibers might bring a new mechanism for the improvement of the fracture toughness. Ramie fibrils and CNTs entangled with each other at the fractured interface, which at least partly contributed to the increased mechanical properties of PFRCs.

Carboxyl-functionalized carbon nanotubes (CNTs-COOH) were also successfully coated onto flax fibers using a "soaking or spraying-drying" process by taking advantage of the unique chemical compositions of plant fibers. The maximum enhancements for interlaminar shear strength (ILSS), mode I interlaminar fracture toughness, were 26 and 31%, respectively, as shown in Fig. 13. It can be seen from the Fourier transform infrared (FTIR) spectrum (Fig. 14) that with the increasing of the CNT content, the intensity of the peaks around 3340 $\mathrm{cm}^{-1}$ (O–H stretching) kept on decreasing, indicating that the amount of the hydroxyl groups of the fibers was reduced by an amount related to the contents of CNTs. It is suggested that hydrogen bonds between the hydroxyl groups of flax fibers and carboxyl groups of CNTs were formed which could strongly bind CNTs to the fibers. Microscopic analysis also showed many fibrillations during the delamination process of flax fiber-reinforced composite laminates due to the existence of CNTs as shown in Fig. 15. The hierarchical microstructures of flax fibers induced new mechanisms for enhancing the mechanical properties of flax fiber-reinforced composites.

Carbon nanotube buckypaper (BP), which is a network structure formed by numerous carbon nanotubes, was also developed and interleaved between layers of flax fiber-reinforced composites (FFRCs). The double cantilever beam (DCB) test results showed that the mode I interlaminar fracture toughness of the FFRC laminate was greatly improved,

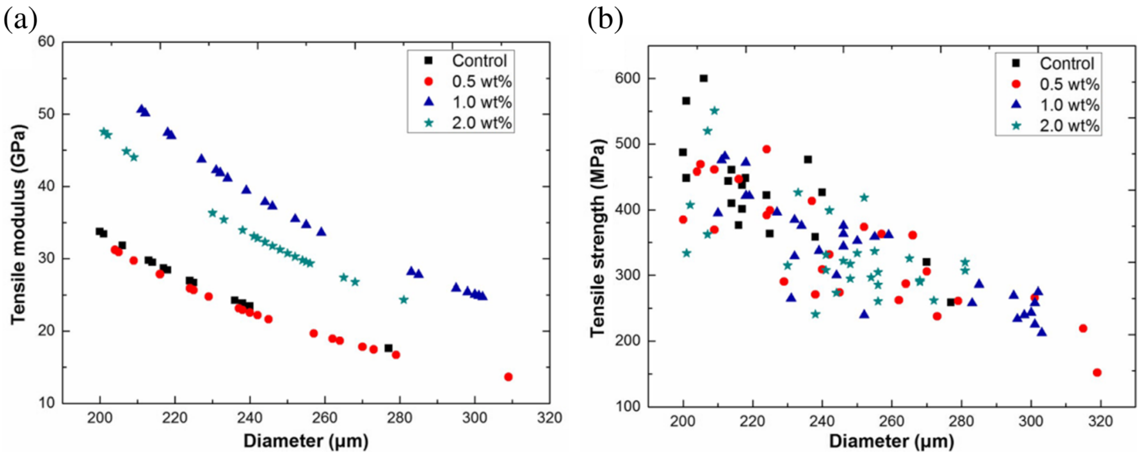


**Fig. 9** Tensile properties of flax yarns coated with different CNT contents. **a** modulus and **b** strength [39]

which was as high as about 30%. Fractured surfaces of the composites revealed the synergistic effects between the BP and the hierarchical structure of flax fibers. The toughening mechanism is summarized in Fig. 16 which included the interlocking of CNTs caused by the penetration of CNTs into flax fibers, the fibrillation of flax fibers due to the enhanced interfacial properties between flax fiber and polymeric matrix, and the toughening of the matrix. Additionally, tensile tests confirmed that the introduction of BP interleaves had no influence on the fiber-dominated in-plane properties.

It can be seen that even though using CNTs can improve the mechanical properties of plant fiber-reinforced composites by taking advantage of the unique hierarchical microstructures of plant fibers, a feasible way to do the treatment is highly required. Using nanocellulose to replace CNTs is also a good way to realize a totally green reinforcement material. Manufacturing and providing nanocellulose-treated plant fibers/yarns or fabrics will be a milestone for plant fiber-reinforced composite industries.

## 4 Damping properties of plant fiber-reinforced composites

Energy absorption by mechanical damping is highly relevant in that vibrations can be dampened in mechanical systems.

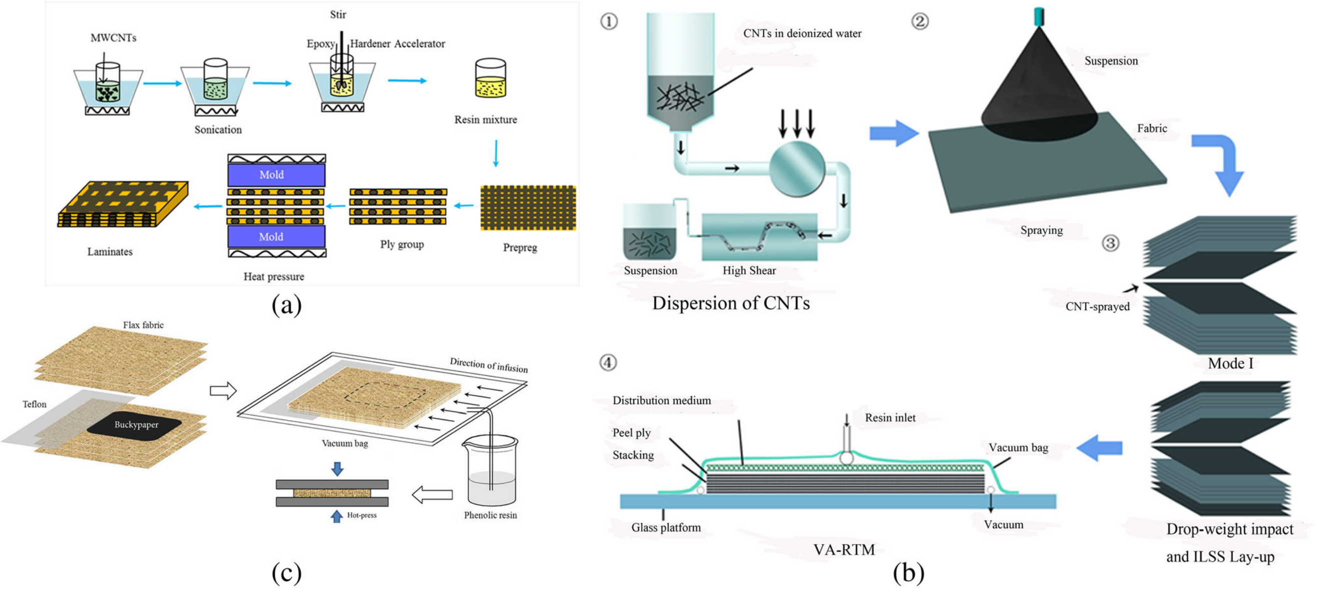


**Fig. 10** Flowcharts for making MWCNT-modified ramie/epoxy composites. **a** adding CNTs to the matrix [43], **b** coating CNTs to the fibers [39], and **c** interleaving buckypaper [42]

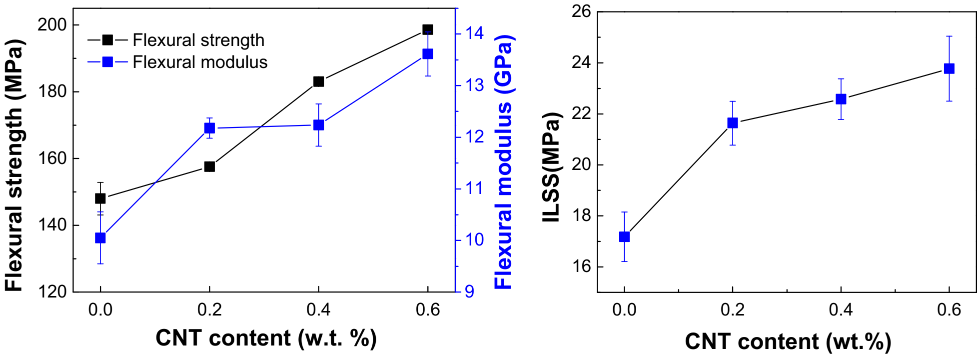


**Fig. 11** Flexural properties and ILSS of ramie fiber/epoxy composites as a function of CNT content

High damping materials are used to reduce vibration in aircrafts and other machinery, thus extending the service life of components and reducing weight and noise. Structural materials generally exhibit high modulus but low damping properties. Rubbery materials do not exhibit structural properties, and they are thus mainly used for damping purposes. Materials that present both high damping and stiffness properties are desired. Fiber-reinforced composites are non-homogeneous materials, and thus major candidate sources for composite damping would include the following: the viscoelastic nature of matrix and/or fiber materials, damping as a result of interphase properties, frictional damping as a result of delamination or slippage in unbonded regions between fiber and matrix interfaces, etc. In comparison with man-made fibers, the hierarchical and porous microstructure of plant fibers may generate a larger number of damping sources.

In a preliminary study, damping behavior of ramie and jute fiber-reinforced composite laminates was tested under free and forced vibration conditions, respectively. It was observed that the loss factor ($\eta$) of ramie fiber-reinforced composites (RFRCs) was 0.0129 at free vibration condition, which is over 30% higher than that of jute fiber-reinforced composites (JFRCs). Vibration behavior of a multilayer composite laminate can also be controlled via hybridization through using alternative symmetrical or asymmetrical plies [44]. Figure 17 shows typical vibration responses of glass-, carbon-, glass/ramie-, and carbon/ramie-reinforced composites. As shown, the vibration property denoted by loss factor $\eta$ for GFRCs (0.0042) and CFRCs (0.0018) was clearly much lower than that for RFRCs (0.0129). Given this difference, which is basically associated with the intrinsic stiffness of the fibers, it was predicted their hybrids could behave in a controlled manner: the more the ramie plies, the greater the damping response of hybrid laminates as shown in Fig. 17. Moreover, the hybrid ratio of the specific fiber types and the symmetrical layer stacking sequence also significantly affected the resulting vibration behaviors. In general, arranging ramie layers symmetrically on both surfaces of the laminate was found to be more effective in terms of damping than covering both surfaces with glass or carbon. The quantitative difference in $\eta$ appears to be remarkable for the surface configurations.

In correspondence with vibration behaviors, mechanical properties were also evaluated for the laminates, and the results are summarized in Table 7. All the properties of the laminates varied depending on the fiber types and stacking configurations used. Ramie-, glass-, and carbon fiber-reinforced laminates set the upper and lower boundaries. Besides the elongation at break, the properties of the hybrid laminates varied in between.

Loss tangent tan$\delta$, a damping measure, serves as the ratio of the imaginary component to the real component of the complex modulus $E^*$. The angle $\delta$ is the phase angle between

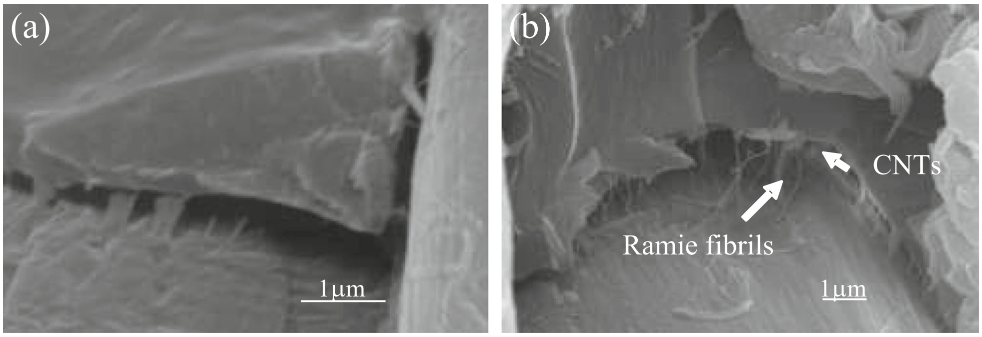


**Fig. 12** Delaminated interfaces for ramie/epoxy composite specimens **a** without and **b** with 0.6% CNTs [43]

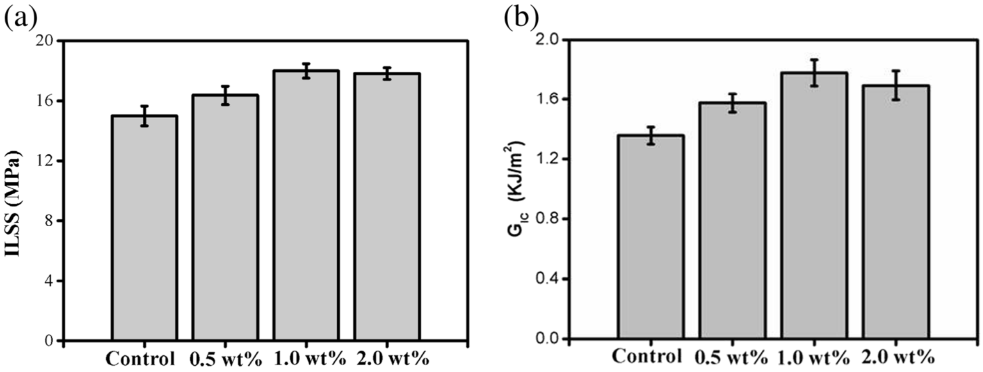


**Fig. 13** **a** ILSS and **b** mode I interlaminar fracture toughness for flax/epoxy composite laminates with different CNT contents

stress and strain sinusoids. Tan*δ* is proportional to the energy loss per cycle within the framework of linear viscoelasticity. Figure 18 shows the storage modulus *E*′ and tan*δ* values for the composite laminates examined as a function of temperature. The curves fluctuated considerably, thus creating numerous opportunities for the definition and design of structural damping properties by hybridization of carbon or glass fibers with plant fibers.

The product of |*E**| and tan*δ* serves as a figure of merit for structural damping materials [45]. Figure 19 presents a stiffness-loss map (|*E**| vs. tan*δ*) of common materials. The upper-right region above the dashed line in the diagram presents structural damping materials to pursue. Composite laminates designed and tested in this study are clearly close to the structural damping materials region.

Similar to the mechanisms of damping properties, sound absorption properties of plant fibers are all superior to those of glass or carbon fibers. Therefore, the sound absorption property of the composite can also be designed by hybridization of plant fibers with glass or carbon fibers. Figure 20 shows the variation of sound absorption coefficients and sound transmission losses of the composites by changing the stacking sequences of different fabric layers.

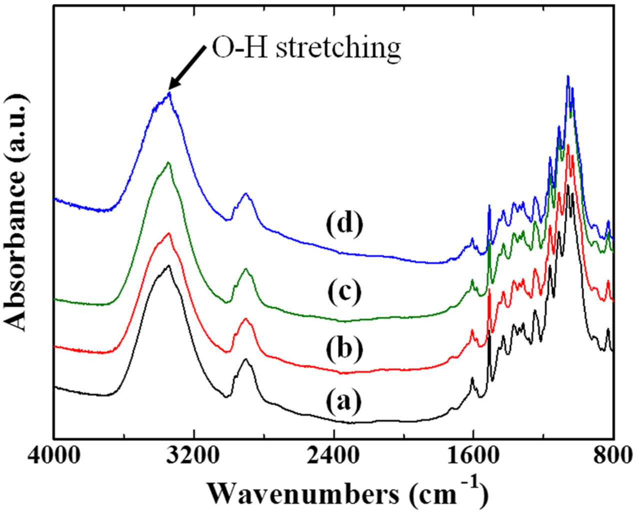


**Fig. 14** FTIR spectra of flax yarns coated with different CNT contents. a uncoated, b 0.5 wt%, c 1.0 wt%, and d 2.0 wt% [39]

## 5 Development of quasi-structural and function-integrated composite parts

The abovementioned fundamental results were applied in making quasi-structural or function-integrated composite

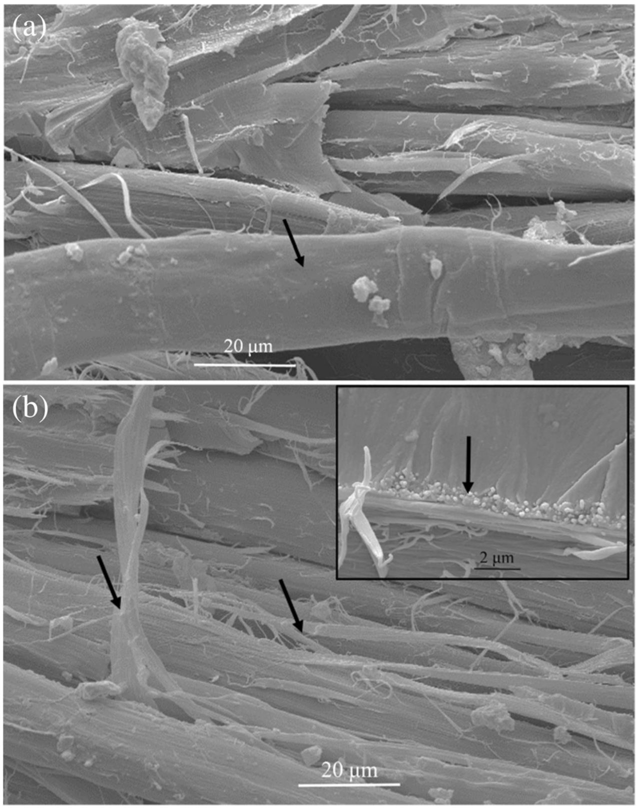


**Fig. 15** Fractured surfaces of flax fiber-reinforced composite laminates after being tested by DCB tests. **a** uncoated and **b** 1.0 wt% CNTs (fiber fibrillation)

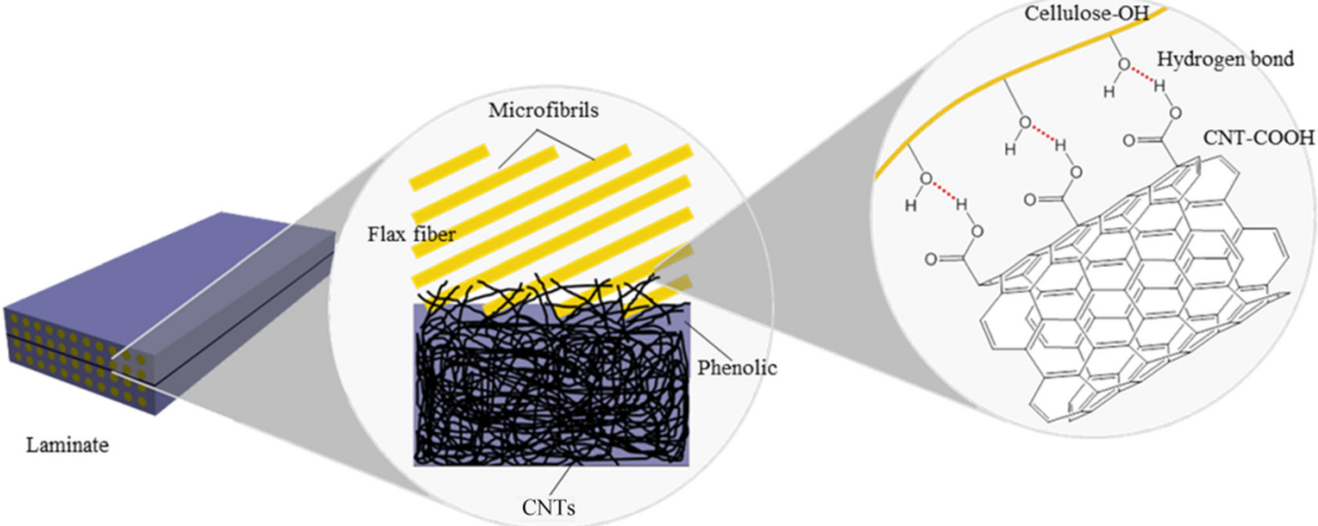


**Fig. 16** Schematic drawing of toughening mechanisms of FFRCs by BP interleaving [42]

structures. A part which can be used in commercial aircraft interior as shown in Fig. 21a was made using plain-woven ramie fabrics, phenolic resin, and an aramid honeycomb core. Strong conforming capacities of the panel along an extremely curved contour were well demonstrated. The ramie fiber-reinforced phenolic composite showed acceptable stiffness and strength in relation to its GFRC counterpart. More specifically, ILSS and tensile modulus were even slightly higher than those for the GFRCs. A manufacturing benefit of the RFRC parts cured in an autoclave pertains to their full compatibility with state-of-the-art industrial production processes. The flammability study also generated acceptable results for aviation interior applications.

Ramie fibers were also used for reinforcing and sound absorption purposes, though in a hybrid fashion with glass, in so-called acoustic panels of propeller aircrafts. The matrix resin used in this case was an intermediate-temperature cured epoxy. Figure 21b shows the manufacturing process and the final panels. Relatively lower mechanical properties of the RFRC laminates in this case were compensated via hybridization with glass fibers. In the application, noise-reduction benefits of the ramie fibers were fulfilled, generating a hybrid design that endowed the structure with additional damping properties.

Most prior R&D studies on plant fiber-reinforced composites and associated trial applications were mainly focused on mechanical and functional issues. Besides their moderate

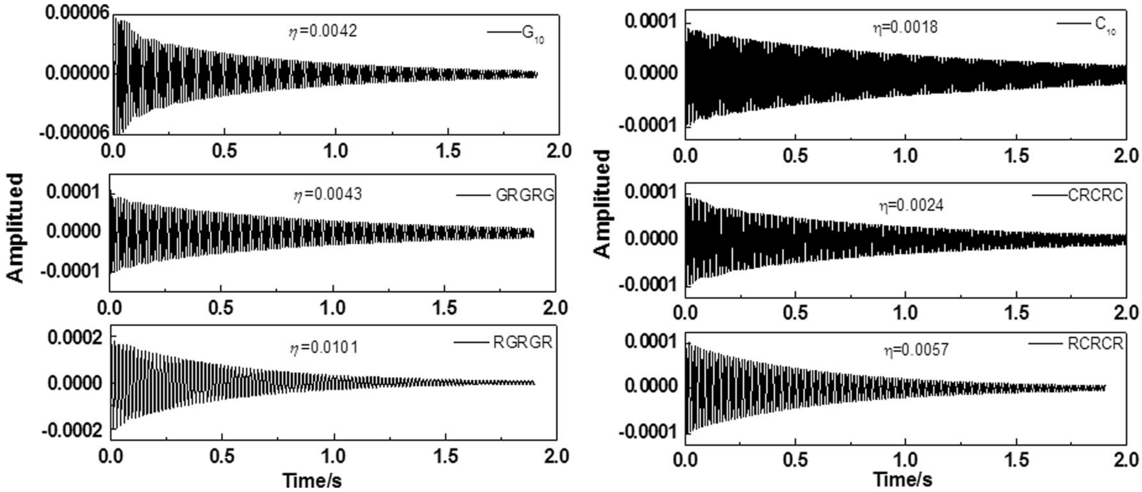


**Fig. 17** Typical vibration responses of glass (G10, 10-ply glass laminate), carbon (C10, 10-ply carbon laminate), glass/ramie, and carbon/ramie hybrid-reinforced composites (For these hybrid composites, each G, R, and C denote two-ply glass, ramie, and carbon layers arranged in a symmetrically alternating manner, respectively) [44]

**Table 7** Comparison of the physical and mechanical properties of the composite laminates [44]

| | Density (g/cm$^3$) | Loss factor | Tensile strength (MPa) | Tensile modulus (GPa) | Interlaminar shear strength (MPa) | Elongation at break (%) | Impact toughness (kJ/m$^2$) |
|---|---|---|---|---|---|---|---|
| R10 | 1.35 | 0.0129 | 68 | 11 | 21 | 0.6 | 24.7 |
| G10 | 1.73 | 0.0042 | 481 | 20 | 71 | 2.4 | 220.7 |
| GRGRG | 1.59 | 0.0043 | 322 | 15 | 59 | 2.2 | 138.4 |
| RGRGR | 1.51 | 0.0101 | 278 | 10 | 36 | 2.84 | 116.4 |
| C10 | 1.53 | 0.0018 | 888 | 61 | 88 | 1.3 | 104.4 |
| CRCRC | 1.49 | 0.0024 | 596 | 40.2 | 50 | 1.5 | 86.2 |
| RCRCR | 1.40 | 0.0057 | 382 | 21.3 | 32 | 1.1 | 55.0 |

mechanical and unique functional properties, ramie fibers are one of the premium plant fibers used in the clothing industry. They are highly durable, white in color, and have a silky finish, enabling their use for decoration purposes. It is a favored textile material for the construction of apparel and home fashions. Such features are promising, as ramie can be used to produce white or colorful composites, unlike known carbon composites that are black in color or aramid fiber composites, which are characteristically yellow.

In a proof-of-concept study, colored ramie fabrics were applied as a decorative face layer on composite laminates to produce a decorative composite. An interior side panel was then manufactured via vacuum infusion with an unsaturated polyester resin used for a rail car (Fig. 22a), whereby dry decorative ramie fabrics were accordingly positioned on the surface of ramie/glass hybrid laminates. For decorative purposes, the more transparent the matrix resin, the better the look of the final product. The structure-decoration-integrated panel was easily fabricated to adapt to the product specifications. The decorative face layer simultaneously served as a structural component that could bear loads, while the traditional decorative surface functions only decoratively. In addition to these advantages, ramie fabrics used in this application also served other functions, e.g., noise absorption and vibration damping.

The addition of decorations to the structure is clearly beneficial. It not only adds esthetic appeal to the composite structure but also simplifies the manufacturing process in terms of materials, labor, and time, and thus reduces costs. A similar part for the interior cabin structure of the world's largest seaplane made in China clearly highlighted the material's esthetic appeal (Fig. 22b).

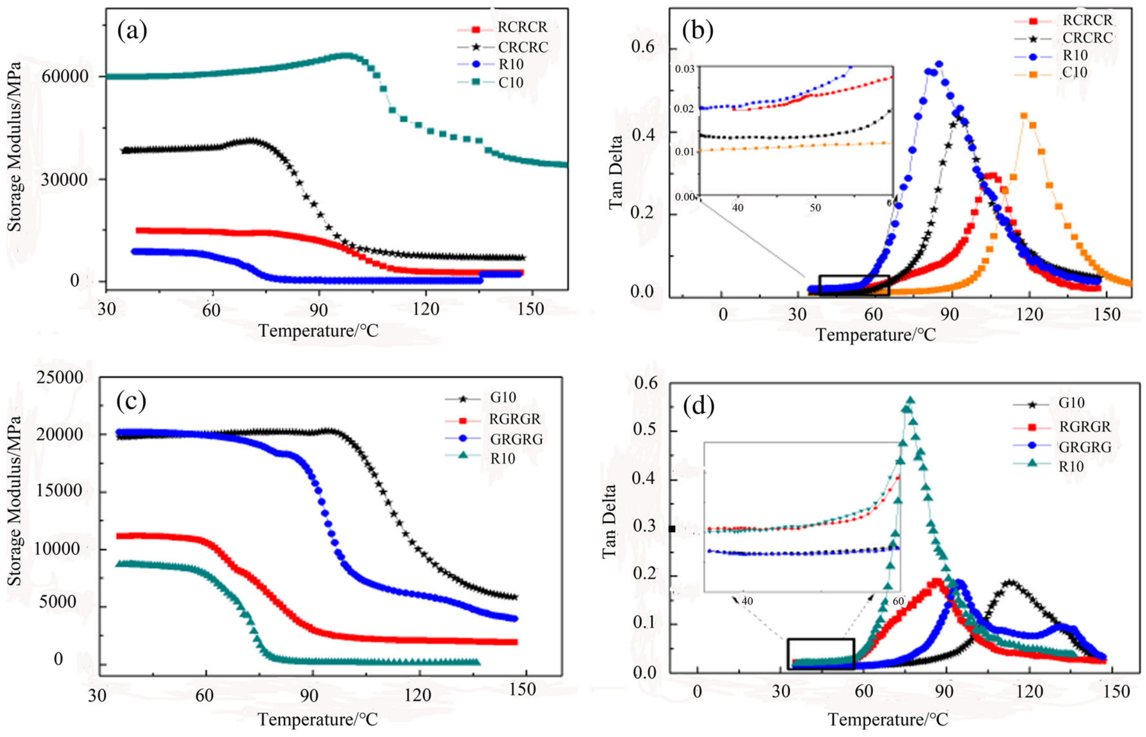


**Fig. 18** Comparisons of **a** loss factor and **b** storage modulus of the laminates [44]

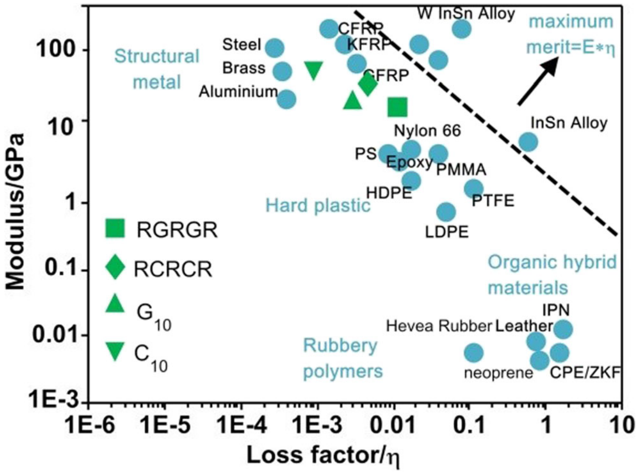


**Fig. 19** Plot of storage modulus vs. loss factor for different materials where areas of the maximum figures of merit and composites studied are schematically illustrated [44]

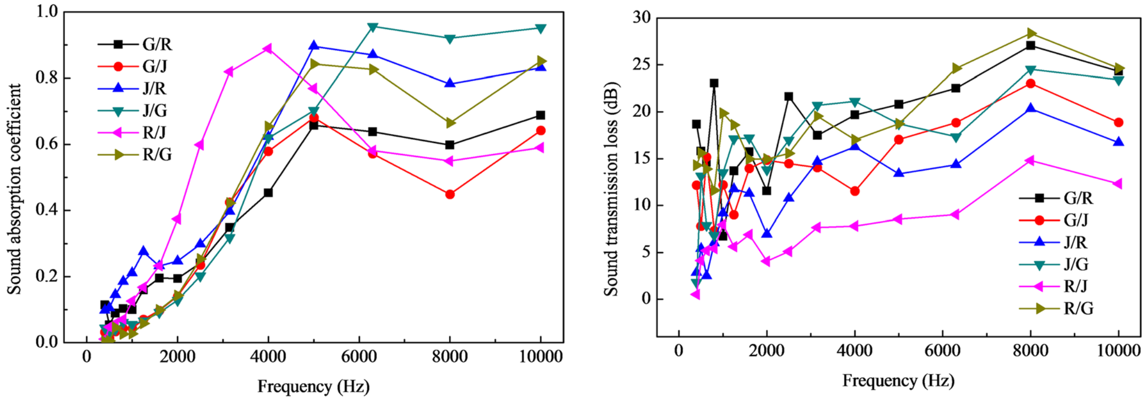
It is noteworthy that the use of multifunctional decorative composites for interior and quasi-structural applications is by no means restricted to ramie textiles. Such composites can also be applied to other plant fiber textiles with similar features such as flax, hemp, etc. Structure-decoration integration methods can clearly support the production of identical or even more complex composite parts while simultaneously presenting mechanical and structural damping advantages in various applications.

Within the realm of civil engineering, some PFRCs can likely compete with GFRCs in terms of absolute and specific tensile stiffness and specific strength [46]. PFRCs have been reported to be used as construct beams and roof panels using hemp, flax, and plant oil-based resin [47, 48]. Misnon et al. critically reviewed the uses of plant textile materials for engineering composite applications and found that PFRCs can only be used in low- to medium-load-bearing applications [49]. PFRCs have also been used to strengthen existing concrete structures that generally adopt GFRCs or CFRCs. Xian et al. used a plain-woven flax fabric with room temperature cured epoxy to confine concrete cylinders (Fig. 23) [50]. The compressive results showed that flax fabric-reinforced composite improved ultimate strain and stress of the cylinders by more than ten- and twofold, respectively, depending on the fabric layers applied. Coefficients of FFRC confinement on ultimate compressive stress were slightly higher than those of basalt fiber-reinforced composites, which have been applied for purposes of comparison. Similarly, Uedae et al. experimentally examined shear beam strengthening processes using a jacketing sheet of flax fibers [51]. The ultimate beam shear strength was reported to increase from 22 to 72% after one to three layers of the sheet were jacketed. All the tested specimens failed by shear immediately after the flax fiber sheet fractured along the major shear crack. Other studies have recently identified ways of strengthening civil engineering structures such as columns [52] and masonry walls [53, 54] using various plant fibers.

## 6 Conclusions and future research

From the review of the recent research development of plant fiber-reinforced composites, it is found that chemical and physical modification of the plant fibers as well as the interlayer modification by taking advantage of their unique hierarchical microstructures in the composite laminates is fundamental and critical for the mechanical properties of this kind of green composites. The hierarchical microstructure also endows plant fibers with superior damping and acoustic properties over man-made fibers. It is recommended that the development of structure-function-integrated PFRCs appears to be more important and feasible than that of mechanically “high-performance” biocomposites.

The main conclusions from this review paper are the following: (1) Mechanical properties of continuous plant fibers were improved by the nanomaterials by taking advantage of their hierarchical microstructures. (2) Modification of plant



(a) (b)

**Fig. 20** Sound absorption coefficients and sound transmission loss of the composites (G, R, and J denote glass, ramie, and jute fibers)

**Fig. 21** Demonstration applications for PFRCs. **a** commercial aircraft interior part and **b** an acoustic panel for propeller aircraft

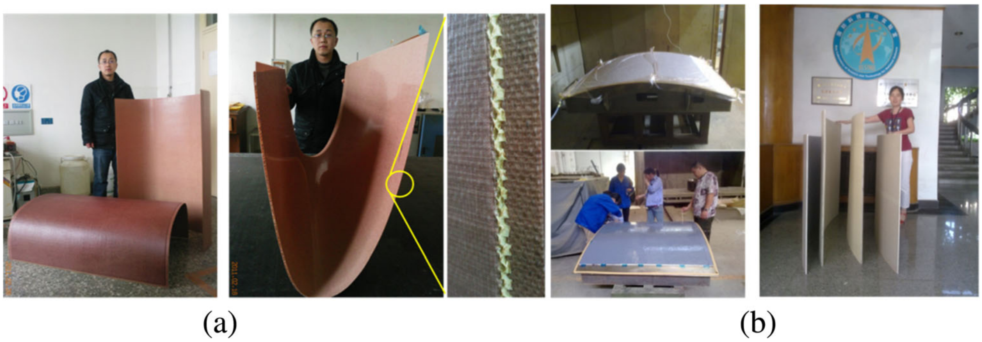
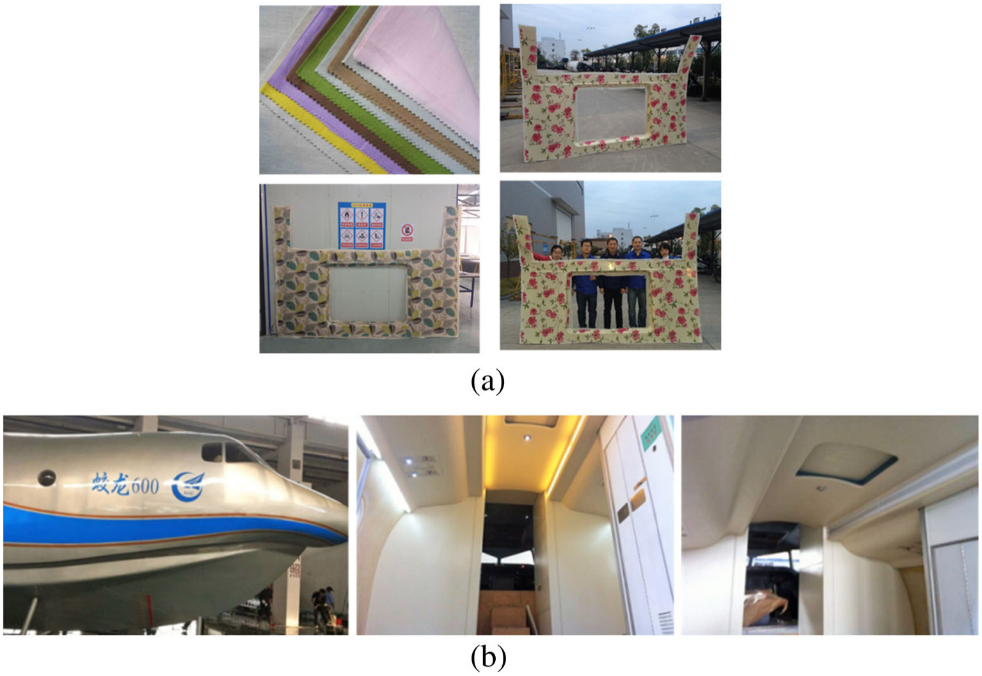
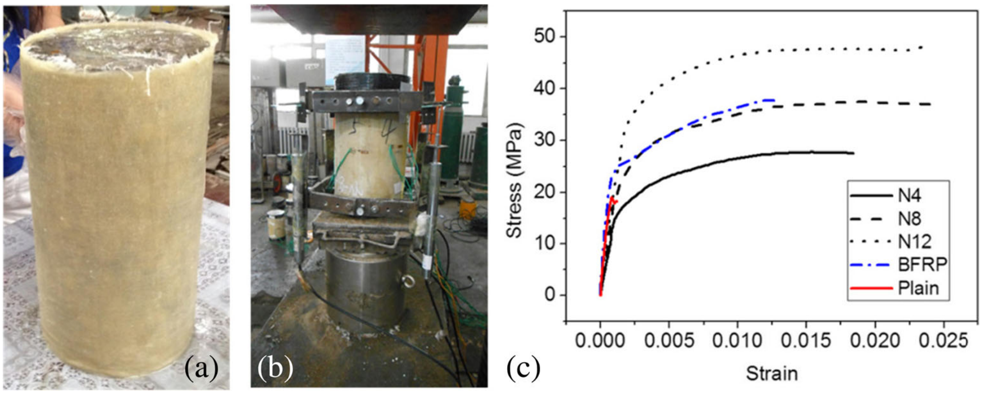


**Fig. 22** Proof-of-concept demonstration of **a** a decorative, quasi-structural composite side panel of a railcar and **b** the interior of a seaplane



fiber-reinforced composites by nanomaterials simultaneously improved the interfacial shear strength between the fiber and matrix, the interlaminar shear strength, and fracture toughness by causing hierarchical multiscaled fracture modes. (3) Plant fiber-reinforced composites possessed superior acoustic and structural damping properties due to their hierarchical and hollow structures. Properly designed and manufactured plant fiber/glass fiber hybrid composites showed great application potentials by owning both structural damping properties and relatively high stiffness. (4) Fundamental studies on

**Fig. 23** **a** Concrete cylinder (150 mm × 300 mm) wrapped by flax fiber fabrics, **b** compressive test set-up, and **c** compressive stress-strain curves of the concrete cylinders (plain: unconfined cylinders; BFRP: cylinders confined by one layer of unidirectional basalt fiber fabric; N4, N8, or N12: cylinders confined by 4, 8, or 12 layers of flax fabric, respectively)



enhancing mechanical properties and designing functional properties were successfully applied to some industrial applications. Quasi-structural and multifunctional PFRC components used for aircraft, railway transportation vehicle, and civil structures were designed and manufactured using technologies fully adapted to standard industrial processes.

Even though structure-function-integrated plant fiber-reinforced composite structures were developed and demonstrated in certain applications with the aid of the successful transformation of the fundamental researches by considering the unique hierarchical microstructures of plant fibers, the realization of the large-scale application of this environmentally friendly material and the partial replacement of the currently widely used man-made glass fiber-reinforced composites are still for more than enough. Even though typical cases pertaining to structural damping and decorative quasi-structural composites have been presented, in order to capitalize on the full capacities of biocomposites, material service performance, manufacturability, and affordability factors must be studied further.

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