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Review

Hybrid Fiber-Reinforced Biocomposites for Marine Applications: A Review

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Abstract: Highly efficient fiber-reinforced composites find extensive application in diverse industries. Yet, conventional fiber-reinforced composites have significant environmental impacts during both manufacturing and disposal. Environmentally friendly fiber-reinforced composites have garnered significant attention within the framework of sustainable development. Utilizing natural fibers in place of synthetic fibers and progressively decreasing the use of synthetic fibers are the main approaches to achieving a balance between economic progress and environmental quality. Attention is increasingly being drawn to natural fiber-reinforced biocomposites that exhibit outstanding environmental performance, exceptional physical and mechanical capabilities, and biological features. The lightweight and high-strength characteristics of these biocomposites enable them to significantly decrease the weight of structures, making them increasingly popular in many industries. The objective of this review is to evaluate the effectiveness of hybrid fiber-reinforced biocomposites in marine applications, specifically examining their mechanical characteristics, resistance to seawater, and ability to absorb moisture, all while advocating for sustainable material methodologies. To achieve this objective, the paper delineates the distinction between synthetic and natural fibers, examines the benefits of hybrid fiber-reinforced biocomposite materials, and addresses the obstacles and effective approaches in their production and application in seawater. Considering the review analysis, it can be inferred that the use of fiber-reinforced biocomposites in maritime applications shows significant potential and has abundant untapped growth prospects in the future years.

Keywords: natural fiber; synthetic fiber; marine application; hygroscopicity; water absorption; nanoparticle; composite material

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1. Introduction

Fiber-reinforced composites (FRCs) are sophisticated structural materials often composed of two or more constituents, such as a matrix and reinforcements, which possess distinct physical, chemical, and mechanical characteristics [1,2]. The many components are combined into a novel intricate framework by highly sophisticated production methods [3,4]. The chemical composition of composite materials also influences their characteristics, therefore addressing the constraints of homogenous materials [5]. The applications of these technologies are extensive and include several sectors like as construction, automobiles, aviation, and watercraft [6].

Synthetic fibers, such as glass, carbon, and aramid fiber, have historically served as primary reinforcements to improve composites and achieve substantial benefits [7,8]. In addition, FRC properties are improved by modifying manufacturing processes and optimizing treatments [9]. Nevertheless, the manufacturing and disposal of synthetic fibers have substantial environmental consequences due to their dependence on finite resources, intensive energy consumption, and exacerbation of the environmental load [10,11]. In response to increasing consciousness of environmental preservation and sustainable development, there is a strong need to substitute unsustainable materials in different industrial sectors with sustainable alternatives [12]. Furthermore, the high cost of synthetic textiles, especially carbon fibers, tends to stimulate the need for more cost-effective and environmentally friendly substitutes [13].

Experimental investigations were carried out by researchers in the field of composite fabrication [14]. Investigations revealed that plant fibers exhibited exceptional properties in composites and substantial potential to enhance the thermostability and mechanical properties of composites [15]. The use of natural fibers as reinforcements in polymer composites presents numerous advantages as potential alternatives to synthetic fibers, particularly in enhancing the mechanical properties of the matrix [16]. Furthermore, natural fiber-reinforced composites exhibit exceptional economic efficiency and environmental friendliness, rendering them highly suitable for a wide range of applications [17]. By incorporating plant fibers as partial or complete substitutes for synthetic fibers, the production of novel functional composite materials can effectively mitigate the environmental consequences linked to synthetic fiber composites [18–21]. Compared with synthetic fibers, using natural fibers can effectively reduce environmental pollution and conserve nonrenewable resources. In addition, fiber-reinforced composites made from natural fibers have low density and superior mechanical properties, which makes these green biomaterials safe and environmentally friendly [22].

Plant fiber, sometimes referred to as cellulose fiber, can be obtained from a variety of plants such as cotton, flax, coir, kenaf, sisal, pineapple, banana, hemp, oil palm, and others. Fibers derived from cellulose plants can be categorized into several groups based on the location of extraction, including seed, leaf, fruit, and bast fibers [23]. In their capacity as renewable resources and sustainable materials, these fibers possess unique properties that make them suitable alternatives to synthetic fibers like glass, carbon, and aramid fibers [24,25]. One possible application of these cellulose fibers is the production of biodegradable biocomposites. Incorporating synthetic and plant fibers can facilitate the advancement of the composite material sector. In their study, Khalid et al. [26] delineate the progression of composite materials into three clearly defined phases, as depicted in Figure 1. The first stage of composite materials development is transitioning to the second stage, characterized by advanced composite materials, then to the third stage, namely green composite materials.

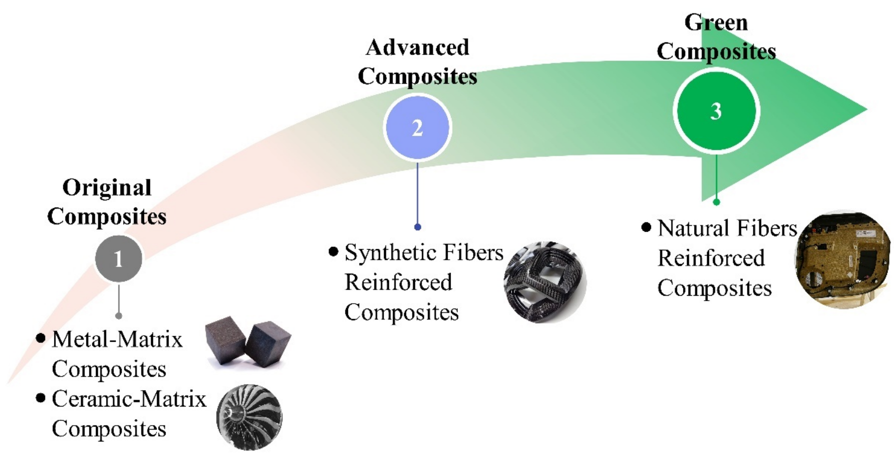


Figure 1. Development of composite materials.

Nevertheless, there are several challenges that must be resolved when using plant fibers for synthesizing biocomposites. These challenges include the susceptibility of plant fibers to moisture and the instability of fiber characteristics, together with the inherent incompatibility between hydrophilic fibers and hydrophobic matrices [27]. Successful resolution of these problems is crucial for the use of biocomposites in maritime applications.

Around 71 percent of the Earth's surface is occupied by the world's seas, which are crucial and essential regions for human production and biodiversity. The prudent harvesting and efficient use of marine resources are highly important for the survival of humanity and the advancement of society [28]. Human activities encompass a wide range of industrial sectors in the majority of oceans, such as navigation, fishing, resource exploitation, and offshore wind operations [29]. Applications in the marine environment pose greater challenges for all materials in comparison to those in the terrestrial environment. Marine structures are exceptionally vulnerable to more intense physical, chemical, and biological forces. Under marine conditions, materials are prone to corrosion and degradation caused by splashing, flooding, and the pressured infiltration of seawater solutions [30]. Particularly, the elevated salinity and humidity in the maritime environment expedite this process, significantly diminishing the longevity and safety of marine structures [31,32]. The vital solution to these problems is in the development of sophisticated functional materials capable of withstanding the severe conditions of the maritime environment.

The FRCs were specifically developed to address the issue of corrosion in conventional materials and have been extensively employed in the shipbuilding sector since their inception [33]. In 1942, Ray Greene constructed a ship using glass fiber-reinforced polyester, hence initiating the integration of fiber composites into the marine sector. A few decades later, carbon fiber and aramid fiber were introduced into the shipbuilding business [34]. The exceptional physical and chemical characteristics of these synthetic fiber-reinforced composites have led to their rapid development in marine engineering [35]. Nevertheless, concerns about the ecological consequences of synthetic fibers have motivated researchers to investigate potential substitutes. By virtue of their renewability and biodegradability, plant fibers serve as a significant substitute for synthetic fibers, therefore mitigating the environmental consequences associated with the latter. Prior investigations have demonstrated that composites reinforced with plant fibers have exceptional mechanical properties [36]. These biocomposites can be utilized in the construction of boat hulls and other water-related equipment [12].

Advanced biocomposites possess exceptional characteristics and find extensive use in all sectors [1,37]. The exceptional quality of biocomposites is driving the increasing market demand. The projected market share of materials derived from renewable raw materials is anticipated to increase to 25% by 2030, surpassing the 18% recorded in 2020 [25]. Furthermore, it is projected that two-thirds of the global industries will eventually be capable of shifting to models based on renewable resources. Furthermore, the wood-plastic composites market is projected to have significant expansion with a substantial compound yearly growth rate of 8.9% from 2021 to 2030. It is anticipated that the market valuation would reach $12.6 billion by 2030 [38]. With the ongoing increase in market demand, biocomposites are anticipated to undergo a period of swift advancement. Expected development focus will be on advanced biocomposites specifically designed for maritime applications.

The aim of this review is to assess the efficacy of hybrid fiber-reinforced biocomposites in marine applications, particularly by analyzing their mechanical properties, resistance to seawater, and moisture absorption capacity, while promoting sustainable material practices. This study investigates different fiber reinforcements and evaluates the performance of FRCs reinforced with them. Furthermore, this review also analyzes the effects of hygroscopicity and seawater aging on these composites and evaluates the enhancements obtained by alkali treatments and nanoparticle incorporation. The present review offers readers a thorough analysis of FRCs and their considerable potential for marine applications, therefore making a valuable contribution to technical innovation and progress in this domain.

2. Types and Properties of Fiber Reinforcements

Composite materials are formed by combining more than two separate materials that have different properties. These components do not dissolve or blend together, but instead interact in a unique way that gives composite materials their exclusive features [39]. The FRC is a prevalent advanced composite material extensively used in several industries such as aerospace, maritime, automotive, construction and infrastructure, medical devices, and sporting event equipment. By incorporating high-strength, low-density fibers as the primary components during the manufacturing process, FRCs exhibit superior strength, stiffness, modulus, and reduced densities compared to traditional composites. Furthermore, composites provide a greater range of combinations compared to metals, polymers, and ceramics, and can be readily customized to achieve any desired characteristic [1]. The fiber reinforcements usually utilized in FRC can be classified into the following two types based on their intrinsic characteristics: synthetic fiber and natural fiber.

2.1. Synthetic Fibers in FRCs

Synthetic fibers provide exceptional reinforcements for the production of composite materials, exhibiting robust stability and great mechanical properties [40,41]. Carbon fibers consistently exhibit superior mechanical strength, chemical stability, and moisture and temperature resistance compared to natural fibers, resulting in increased attention towards them [14,42]. Carbon fiber not only has tensile strength, but also has high thermal and electrical conductivity, respectively, 900–1100 W/mK and 106 S/m [43,44]. Carbon fiber has a wide range of applications in the field of aviation [45]. By employing these dependable synthetic fibers, FRC can be customized with diverse characteristics to correspond to the needs of different situations. Furthermore, the utilization of sophisticated production techniques, such as vacuum bagging, vacuum-infused resins, and extrusion technologies, enhances the precision of composite manufacturing and raises the overall quality of the products. Indeed, synthetic fibers, as their name suggests, are artificially produced and do not arise from natural sources. Certain synthetic fibers are derived from petroleum and necessitate a substantial amount of energy for their production. Industrial manufacturing and recycling of synthetic fiber materials and their byproducts have a detrimental effect on the environment [46].

The synthetic fiber production process generally encompasses a sequence of stages, which include the preparation of raw materials, chemical polymerization, spinning, stretching, winding, and packaging. According to Rajak et al. [40], synthetic fibers can be classified into the following three main groups: organic fibers, inorganic fibers, and others. These categories are further expanded based on their origin, as illustrated in Figure 2. Nylon fiber and aramid fiber are the predominant organic synthetic fibers now in use, whereas glass fiber and carbon fiber are the most often used inorganic synthetic fibers.

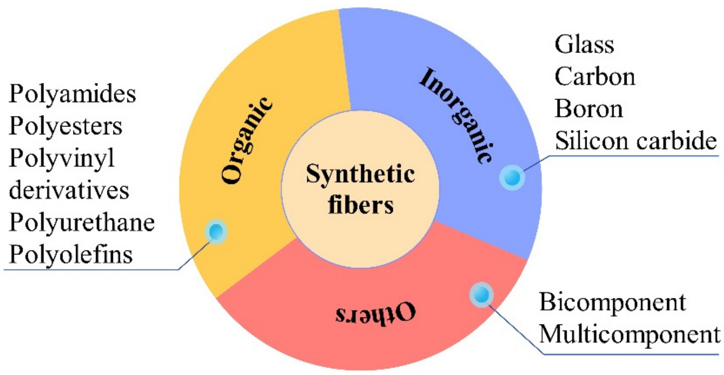


Figure 2. Classification of synthetic fibers based on organic, inorganic, and other fibers.

The properties of synthetic fibers are shown in Table 1. Owing to their distinctive characteristics, these fibers find use in many domains of life and industry. Furthermore, nylon fibers exhibit exceptional toughness, flexibility, and lifespan. Nylon fibers also exhibit a higher degree of softness in comparison to other synthetic fibers that are frequently employed in the textile and home goods sectors. The broad fabrication of sutures, catheters, dentures, and other medical equipment is attributed to the biocompatible nature, good chemical stability, and changeable mechanical properties of nylon [47]. Aramid fiber is a durable synthetic fiber that demonstrates great qualities such as high-temperature stability, impact stability, lightweight, and other unique features, which contribute to its outstanding energy absorption performance [48]. The density of aramid fibers is merely 55% of that of glass fibers, although its strength is equivalent to that of carbon fibers, far more than that of glass fibers, and exhibits a notable level of toughness. Aramid fibers have widespread application in the aviation industry, military industries, and the manufacturing of protective equipment [49].

Table 1. Physical and mechanical properties of common synthetic fibers.

Materials	Density (g/cm^3)	Elongation at Break (%)	Tensile Strength (MPa)	Young's Modulus (GPa)	Ref.
Nylon 6	1.13–1.15	16–19	600–1050	4–5	[47]
Nylon 66	1.06–1.08	41–59	36.1–45.1	0.939–1.17	[47]
Aramid	1.4	3.3–3.7	3000–3150	63–67	[17]
Aramid	1.44	-	3620 ± 68	131 ± 8	[50]
Carbon	1.4	1.4~1.8	4000	230–240	[17]
Carbon	1.65	-	3190 ± 56	228	[50]
E-glass	2.5	2.5	2000–3500	70	[17]
E-glass	2.55	2.7	1950	72	[51]

Fibers of synthetic origin have exceptional properties for the production of FRCs. Nevertheless, the dependence on finite resources, excessive energy usage, and environmental contamination during the whole lifespan of synthetic fibers have led individuals to contemplate the use of environmentally renewable fiber materials as substitutes for synthetic ones. Natural fiber is a prominent replacement with distinct inherent advantages.

2.2. Natural Fibers in FRCs

The ecological issues resulting from waste and the significant benefits of renewable and biodegradable natural fibers have motivated researchers to focus on the advancement of ecologically sustainable composites reinforced with natural fibers [52]. Natural fibers can enhance the mechanical characteristics of FRCs in comparison to synthetic fibers, thereby making the composite biodegradable and environmentally benign [53,54].

Natural fibers are abundant and sustainable resources on Earth, fundamentally important in many sectors such as textiles, paper, and packaging. Humans have accumulated vast expertise in the extraction and utilization of natural fibers. The sources of natural fibers encompass a wide range, including animals, plants, and minerals [55]. Within the realm of natural fibers, plant fibers distinguish themselves by their remarkable benefits in terms of cost-efficiency and ecological sustainability as compared to other types.

Plant fibers are commonly known as cellulose fibers due to their high cellulose content. The application of these fibers has attracted considerable attention due to their eco-friendly nature, physical properties, biodegradation characteristics, simple production process, and low energy consumption [56]. The fibers can be extracted from different locations of plants, such as straw, leaf, fruit, bast, seed, among others, to meet various demands using advanced physical, chemical, and biological extraction technologies, such as the mechanical extraction method, manual extraction, and retting process [55]. These particular fibers are the most suitable candidates for replacement of their synthetic counterparts due to their favorable environmental attributes and mechanical properties [6,57,58].

Cellulose fibers can be extracted from diverse locations on plant bodies. According to the name of these areas, cellulose fibers can be further subdivided into seed fibers (cotton, luffa, and kapok), leaf fibers (sisal, pineapple, abaca, agave, and banana), bast fiber (jute, flax, ramie, hemp, kenaf, and nettle), fruit fiber (oil palm, coir, and areca), and stem fiber (wood, bamboo, grass, barley, straw, wheat straw, and bagasse) [59]. The major chemical elements of cellulose fibers contain cellulose, hemicellulose, lignin, wax, pectin, and other impurities [59–61]. However, the mechanical properties of cellulose fibers exhibit significant variation based on their precise chemical composition and the type of plant from which they are extracted, as shown in Table 2. Compared with synthetic fibers, we can find that the density of cellulose fibers is lower than that of glass fibers and comparable to that of aramid and carbon fibers; the elongation at break of cellulose fiber is higher than that of carbon fiber and exceeds that of aramid and glass fiber. The characterization of cellulose fibers with low density is crucial for the fabrication of lightweight composite materials.

Table 2. Physical and mechanical properties, and chemical composition of common plant fibers [17,59–63].

Materials	Density (g/cm^3)	Elongation at Break (%)	Tensile Strength (MPa)	Young's Modulus (GPa)	Cellulose wt.%	Hemicellulose wt.%	Lignin wt.%	Wax wt.%	Pectin wt.%
bamboo	1.5	3	575	27	26–43	30	21–31	-	-
coir	1.2	15~30	175~220	4~6	36–43	0.15–0.25	41–45	-	3–4
cotton	1.51	3–10	400	12	85–90	5.7	-	0.6	-
flax	1.4	1.6	1400	70	71	18.6–20.6	2.2	1.5	-
kenaf	1.2	1.6	930	53	72	20.	9	-	-
jute	1.46	1.5~1.8	393–773	26.5	61–71	14–20	12–13		0.5
PALF	1.32	2.4	413~1627	60~82	55–68	15–20	8–12	-	-
sisal	1.43~1.5	2~7	350~700	9~22	67–78	10–14.2	8–11	2	1
hemp	1.47	2.38	690	70	68	15	10	0.8	-

Cellulose fiber is a complex multilayer structure composed of a thin primary wall and three secondary wall layers, as shown in Figure 3. The primary wall is the first layer formed during cell growth and encases the internal secondary walls [56]. The mechanical properties of the cellulose fiber are determined by the secondary wall (S2), which accounts for approximately 80% of the total wall thickness [64].

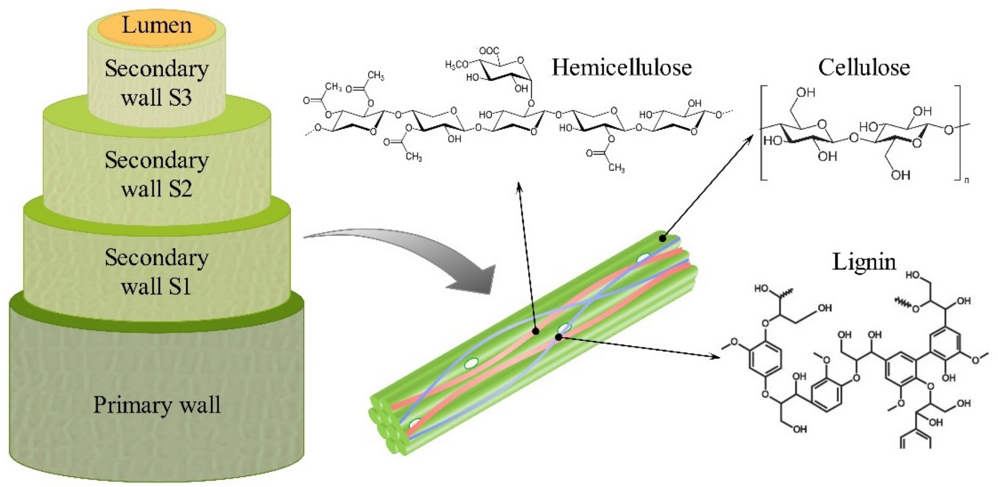


Figure 3. Structural composition of the cellulose fiber.

However, the mechanical properties and structure of cellulose fibers are subject to several factors, including species, climate, geographical location of growth, degree of maturity, and age of the plant, due to the intrinsic characteristics of their natural growth processes. The quality of these fibers is also influenced by the methods employed in their extraction and the technology utilized in their processing. To eliminate unfavorable effects when using cellulose fibers, physical or chemical treatments are employed to remove surface impurities from fiber surfaces. Previous research results have shown that natural fibers can be used to produce composite materials that wholly or partly replace synthetic fibers, and these new composite materials have good mechanical properties and capabilities to meet the requirements of utilization [62,65].

3. Performance of Hybrid Fiber-Reinforced Biocomposites

The growing emphasis on environmental protection and sustainable development contributes to innovation in eco-friendly materials, prompting researchers to shift their research and development focus to biocomposites [59]. To meet the requirements of functional applications, materials must be biodegradable with sufficient mechanical properties. Cellulose fibers are ideally suited for manufacturing biocomposites [65]. The reason is that these fibers have a wide variety, high yield, low cost, excellent mechanical properties, and minimal environmental impact. Although cellulose fibers demonstrate excellent mechanical properties, they still fall short of providing the same level of stability and durability as synthetic fibers, such as carbon, glass, and aramid fiber. Just using natural fibers as reinforcements limits the performance of FRCs. In addition, the hydrophilic nature of cellulose fibers also influences the properties of FRCs [66]. Combining plant and synthetic fibers or using two kinds of plant fibers to create hybrid fiber-reinforced composites can mitigate these adverse issues. In addition, rational hybridization techniques can be applied to produce green and sustainable biocomposites. Furthermore, the hybridization of multiple reinforcements has synergistic effects on the dynamic mechanical properties [67].

Ensuring that hybrid composites have the necessary physical and mechanical characteristics requires suitable ratios to create high-strength, long-durable, and low-density composite materials. The performance of the composites varies with different raw material ratios, enabling better customization to meet specific design requirements. Khalid et al. [26] used woven E-glass fiber and jute fiber to create 5-layer hybrid composite laminates, and the tensile test result showed that the non-hybrid laminate GGGGG displayed the highest tensile strength, reaching 87 MPa. The tensile strength decreased as glass fiber layers were replaced with jute fiber layers, with the tensile strengths of GGJGG, GJGJG, and GJJJG laminates measured at 83, 54, and 43, respectively (see Figure 4). The tensile strength declined when jute layers were incorporated. The replacement of a single glass fiber layer with jute fibers in the core location resulted in a reduction of only 4.6% in tensile strength. However, the GJGJG laminate led to a significant decrease in tensile strength of 37.93% compared to the GGGGG form. The main reason for this result is that the mechanical properties of jute fiber are lower than glass fiber. Although the strength of FRCs was reduced with the replacement of synthetic fibers by plant fibers compared to the pure synthetic fiber-reinforced composite, the elongation of FRCs at break is improved, and the ductility of FRCs increases as the percentage of plant fibers increases.

The stacking sequence of fiber fabric layers with different properties in the composites affects the properties of the composites, especially in flexural strength, interlaminar shear strength (ILSS), fatigue properties, and so on [68,69]. Apart from that, the number of layers also influences the mechanical properties. Das et al. [70] used jute fiber and glass fiber as reinforcement to fabricate unsaturated polyester resin composite laminates. After tensile and flexural strength tests, they found that stacking sequence and number of layers influence laminate strength, as shown in Table 3.

The number of accumulated layers has an obvious effect on the thickness of the specimen. Incorporating synthetic fibers into the laminate positively impacts both tensile strength and modulus, as demonstrated in Figure 5. The primary reasons for this result are as follows: firstly, synthetic fibers exhibit superior strength to natural fibers; secondly, the fiber-matrix adhesion in glass fiber/polyester composites is more effective than in jute/polyester composites, so pulling glass fibers from polyester substrates and breaking them requires greater force than natural fibers. The stacking sequence is a critical factor in determining the flexural strength and modulus of laminates, with these properties being significantly enhanced when glass fibers are positioned on the outer layers, a result consistent with previous findings by Sanjay and Yogesha [71].

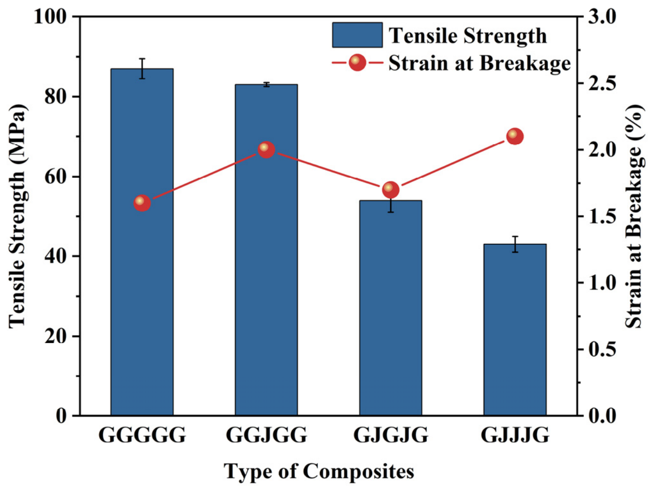


Figure 4. Tensile strength and fracture strain of E-glass fiber (G) and jute fiber (J) hybrid composite laminates.

Table 3. Tensile and flexural strength and modulus of various stacking sequences.

Stacking Sequence	Number of Layers	Thickness (mm)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Flexural Strength (MPa)	Flexural Modulus (GPa)
JJJJJ	5	1.95 ± 0.07	64.6	3.48	127.2	3.52
JG	2	0.71 ± 0.02	91.3	4.89	166.6	5.54
JGJ	3	1.66 ± 0.07	76.7	2.58	114.4	2.99
GJG	3	1.43 ± 0.03	123.1	4.30	313.0	9.14
JGJG	4	1.11 ± 0.08	132.8	5.42	173.6	6.71
JGJGJ	5	1.62 ± 0.03	125.8	3.68	163.3	4.41
GJGJG	5	1.80 ± 0.07	137.6	4.62	252.4	10.59
GGGGG	5	2.17 ± 0.09	159.1	3.79	340.4	9.36

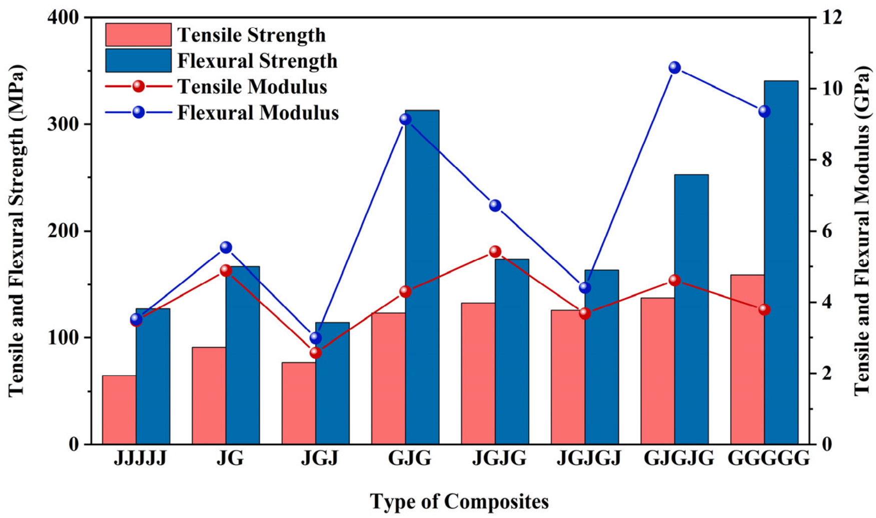
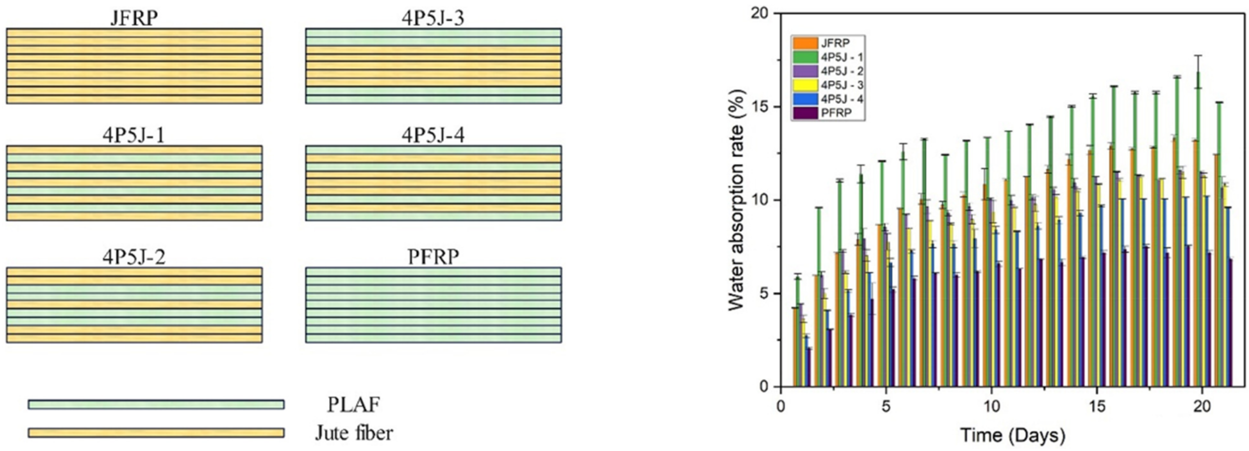


Figure 5. Tensile strength, flexural strength and modulus of E-glass fiber (G) and jute fiber (J) composite laminates.

Researchers also employed two types of natural fiber materials to fabricate hybrid biocomposites to increase mechanical properties and reduce moisture absorption [72]. Baigh et al. [73] created hybrid biocomposites reinforced with pineapple leaf fiber (PALF) and jute fiber, as illustrated in Figure 6a. The moisture absorption results revealed that the mass of all composites increased daily. The weight gain was most significant during the first few days, but, after six days, the water absorption capacity declined, reaching a plateau stage. The weight gain peaked on the 20th day, as shown in Figure 6b. The researchers found that the hybrid composites, particularly 4P5J-2, 4P5J-3, and 4P5J-4, significantly reduced moisture absorption by 16.70%, 14.84%, and 29.50%, respectively, compared to 12.44% JFRP water absorption after 21 days. However, the moisture absorption test results for 4P5J-1 were contrary to those of other hybrid composites. Although hybridization increases water absorption, optimizing the hybridization sequence and improving the interface between the fiber layers and the substrate can help mitigate the issue of hygroscopicity. Praveena [72] also demonstrated that reasonably hybridized plant fibers can increase the mechanical properties of composites and reduce moisture absorption.



(a) (b)

Figure 6. Hybrid natural fiber-reinforced composites uptake water test: (**a**) Stacking sequence and nomenclature of biocomposite laminate; (**b**) Moisture absorption rate after immersion in water at indoor temperature (Reproduced with permission from [73], Elsevier, 2023).

4. Hygroscopic Behavior of Plant Fibers in Biocomposites

The hygroscopic behavior of plant fibers is related to fiber structure and its chemistry; cellulose, hemicellulose, and lignin are all extremely hygroscopic [74]. The inherent chemical composition of cellulose fibers leads to moisture absorption from the environment. Additionally, the rate of moisture absorption of materials is affected by environmental conditions, and high temperatures can accelerate the process of moisture absorption [75]. The hygroscopic behavior of fiber composite biomaterials is positively related to the percentage of plant fibers in the material [76]. The hydrophilic nature of these fibers can significantly impact the mechanical performance of biocomposites [77,78]. The moisture can weaken or disrupt the effectiveness of the fiber-matrix interface [79]. Addressing the issue of interfacial bonding between the fibers and the matrix is paramount when keeping the mechanical properties of biocomposites in wet environments. Numerous elements influence hygroscopic qualities, such as the kind of matrix and fiber, the surrounding temperature, and the period of exposition [72,80].

Traditionally, natural fibers have more sensitive hygroscopicity than synthetic fibers in composites due to the difference in chemical composition. Hygroscopic behavior is one of the important indexes affecting all kinds of composite materials [81–84], especially in water absorption tendency composites [85]. The water absorption of biocomposite materials typically exhibits an initial rapid uptake, which gradually slows down over time [73]. Generally, when water molecules penetrate the biocomposite, the natural fibers absorb moisture and expand. Fiber expansion generates micro-stress at the fiber-matrix interface, forming cracks in the biocomposite, further affecting the integrity of the materials. As a result, the strength and modulus of the composite diminish, along with the material's durability and other properties; this process is shown in Figure 7. As the soaking time increases, water-soluble substances will separate from the material, and fungus will grow, further accelerating degradation [86]. However, some experiments showed that the properties of the material are enhanced after absorbing water; for example, Muñoz and García-Manrique [87] made flax/epoxy composite samples to test the tensile strength in wet (with 768 h water immersed) and dry conditions. After testing, they found that the wet samples displayed higher tensile strength than the dry ones. For samples reinforced with 40 wt% flax, the tensile strain increases by 51%, while for samples with 55 wt% flax, the strain increases by 27%. The expanded fiber can fill the gaps between the fiber and the matrix material caused by shrinkage and deformation, thereby enhancing the binding ability [88], as shown in Figure 7a,b.

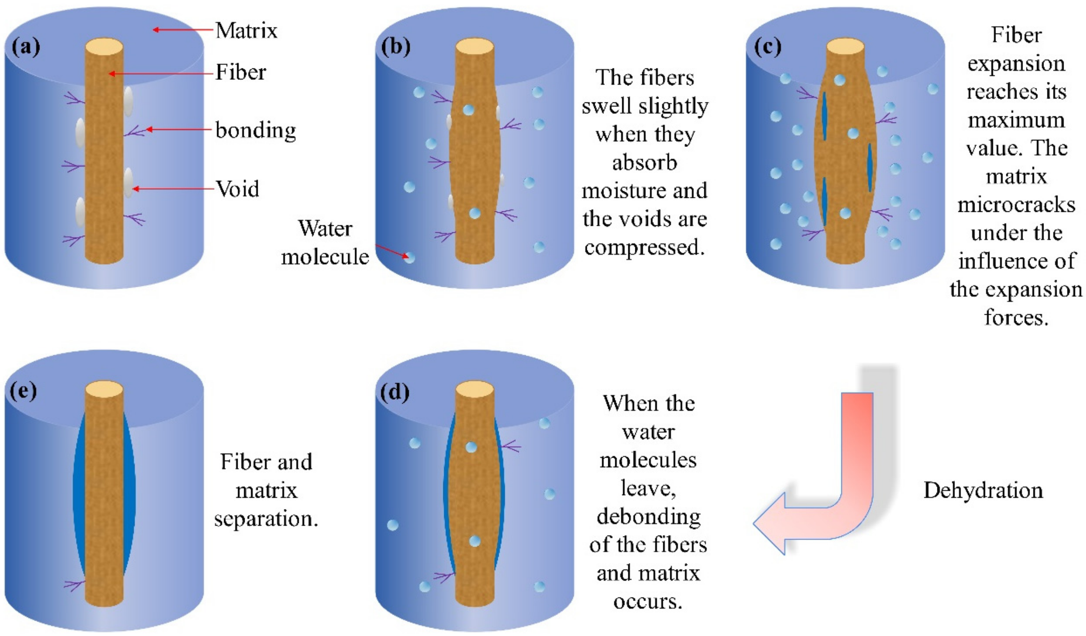


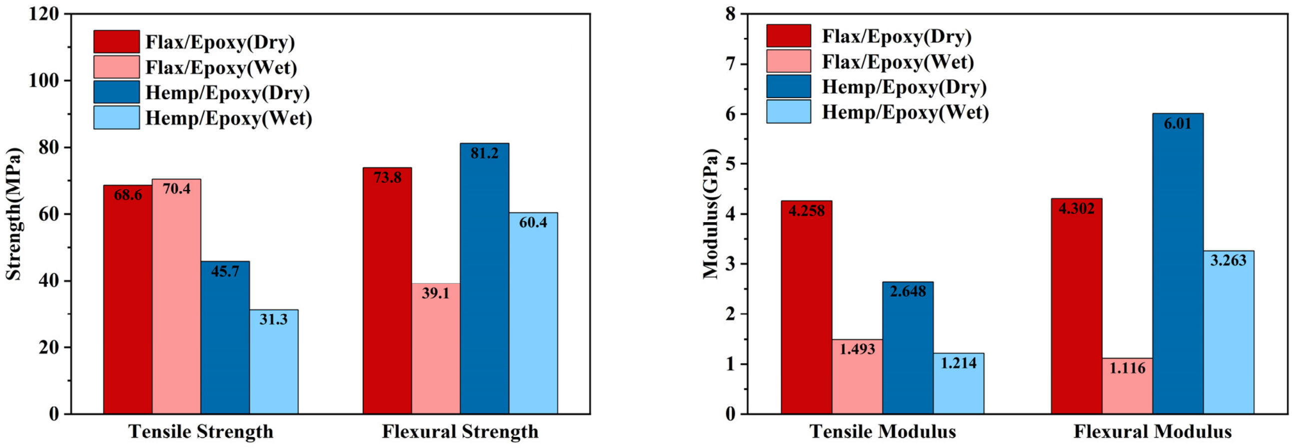
Figure 7. The influence of water in the interface between fiber and matrix: (**a**) Initial state before water absorption; (**b**) Moisture absorption process; (**c**) Saturation state; (**d**) Drying process; (**e**) Dry state after moisture absorption.

After absorbing moisture, biocomposites can have adverse effects, such as extraction of soluble components, degradation of mechanical properties and material structure, and alteration of dimension and quality [89]. The water absorption of composites can be affected by various factors, including the type of fiber, the matrix material, their respective volume proportions, and the preparation method [90,91]. Effectively limiting and managing water absorption is crucial throughout the manufacturing process and service life of biocomposites. Pre-fabrication thermal treatment is necessary to remove moisture from natural fibers, enhancing surface adhesion between the fibers and polymers [4,92].

5. Seawater Aging of Biocomposites

The seawater aging test refers to the experimental method of exposing composite materials to seawater for a long period of time and analyzing the changes in material characteristics [93,94]. Seawater is a solution of complex composition with high salinity values [95]. The hygroscopic nature of plant fibers can lead to challenges for their composites in marine applications. Seawater contains various ions, such as Na^{+}, Cl^{-}, Mg^{2+}, SO_4^{2-}, Ca^{2+}, K^{+}, CO_3^{2+}, etc. [96]. The chemical interactions between seawater and composite materials reduce material stability, promote the dissolution of water-soluble components, and negatively impact durability [97]. The constant moisture and harsh seawater environmental conditions significantly accelerate material corrosion and degradation, posing serious challenges to the safety and durability of marine engineering structures [98]. In addition, seawater engineering also suffers from physical effects, such as wave impact and solid floating objects impact [99]. Seawater aging cycles can promote the imperfectly crystalline components of the structure to dissolve, allowing the cellulose fibril to rearrange and improving the hygroscopic properties of the composite [100]. The deterioration of FRCs can be attributed to several factors, including the expansion and plasticization of the material, the disparity in expansion coefficients between components, and the reduction in cross-linking density during the aging process [101]. The bonding between the fiber and matrix may break down, further degrading structural integrity [102]. In addition, the breaking threshold of FRCs decreases with increasing exposure temperature and time [95,103]. With increasing interest in extending the application of cellulose FRCs in this field, researchers have devoted greater attention to this study area [104,105]. Therefore, after prolonged exposed to seawater, FRCs will suffer dimensional changes and deterioration of mechanical properties, such as fiber swelling, material volume expansion, deterioration of the fiber-matrix interface, intrusion of harmful ions in seawater, and degradation of natural fibers.

Exposure to the seawater environment reduces the mechanical properties of FRCs and increases elongation at fracture, reflecting a decrease in brittleness and an enhancement in ductility [106,107]. Velasco-Parra et al. use the vacuum infusion method to fabricate jute/epoxy composites. After the aging test, the results indicated that the tensile strength and modulus decreased by 1.49% and 4.11%, and the elongation increased by 6.67% at the break [108]. In addition, Haramina et al. [109] conducted aging tests on flax/epoxy and hemp/epoxy biocomposites, finding that flax/epoxy's elongation at break increased from 4% to 8% and hemp/epoxy's elongation from 2.5% to 3.5%, corresponding to increases of 100% and 40%, respectively. Furthermore, they also found that different fibers with the same weight fraction in composites can exhibit varying seawater uptake performances. Water absorption tests showed that flax/epoxy composites demonstrated lower water uptake compared to hemp/epoxy composites, with a maximum water absorption rate of 7.5% for flax/epoxy and 9.8% for hemp/epoxy, and the mechanical properties (Figure 8).



(a) (b)

Figure 8. Mechanical properties of Flax/Epoxy and Hemp/Epoxy: (**a**) Strength changes under wet and dry conditions; (**b**) Modulus changes under wet and dry conditions.

However, the short period of seawater aging positively affects the strength of FRCs. Antunes et al. [110] evaluated the strength of glass fiber/epoxy composite cylinders exposed to seawater at 80 °C for 7 to 28 days, and the results indicated that the strength of cylinders aged for 7 days exceeded that of unaged samples, and the strength decreased with time after 7 days. This finding suggested that limited seawater aging time may enhance the cross-linking density of the matrix, further increasing the resistance strength of FRCs.

Hybridizing synthetic fiber in natural FRCs and altering the stacking sequence are key methods for enhancing the physical and chemical properties of biocomposites [111,112]. Calabrese et al. assessed the durability and mechanical stability of the flax/epoxy laminate and the hybrid glass and flax/epoxy laminate for application in the marine environment. The results indicated that adding glass fibers to flax laminate increased flexural strength by 90% and modulus by 128%, and external glass fiber can protect the internal flax fiber to enhance the durability of flax/epoxy laminate in marine environments [113].

Fiber-reinforced biocomposites show a tendency to decrease mechanical properties after long-term seawater aging [111,114]. To adapt to the requirements of seawater environments, the primary objective is to address how to maintain or improve the original mechanical properties of composites in such environments. Applying biocomposites in seawater environments requires reducing the diffusion coefficient of seawater molecules in composites and increasing the durability of materials [115]. Furthermore, improving resistance to microbial degradation, salt fog aging, and UV aging is crucial when using these biocomposites in the marine environment [105,116]. Various methods, including the use of corrosion-resistant reinforcements, optimization of matrix materials, chemical and physical pre-treatment of fibers, and the incorporation of appropriate nanoparticles, have been explored and applied to enhance the mechanical properties and resistance of biocomposites [20,117–119]. These measures can significantly enhance the interfacial bonding between reinforcing fibers and the matrix, improving the overall material performance.

6. Effect of NaOH Treatment and Nanoparticles on Biocomposites

6.1. Effect of NaOH Treatment on Biocomposites

Sodium hydroxide (NaOH) treatment, also known as alkali treatment or mercerizing, is an effective chemical method of cleaning fiber fabrics and can remove impurities from the surface of natural fibers, such as waxes and lignin, thereby increasing the roughness of the fiber surface and improving adhesion with polymer. The structure of cellulose fibers is complex and multi-component, containing not only cellulose but also impurities that will affect the bonding with polymers [120]. Before using natural fibers, it is important to remove the detrimental impurities [121]. Although various physical and chemical methods can achieve this result [122,123], treating with a sodium hydroxide solution is one of the preferred modification methods because of its effectiveness, simplicity of operation, and low cost [15,55,124]. Alkali treatment can modify the surface of the cellulose fibers and remove the impurities on the surface of fibers [125]. The fiber surface becomes clean and rough after treatment, so the interface bonding between fiber and matrix can be strengthened through the interlock formed after the alkalization process [126]. In addition, mercerization can improve the crystallinity and stability of cellulose fibers [127]. After alkali treatment, Bernardes et al. [128] reported that pineapple crown fibers' cellulose content increased from 18.93 to 57.00%, fiber diameter decreased from 6.1 to 4.3 μm, and the fiber's crystallinity index rose from 53 to 62%. Furthermore, hydroxyl on the natural fibers can be eliminated by NaOH treatment, which reduces moisture absorption when exposed to wet conditions, and the reaction occurs as shown in the below function [66]:

$$\mathrm{Fiber} - \mathrm{OH} + \mathrm{NaOH} \rightarrow \mathrm{Fiber} - \mathrm{O} - \mathrm{Na}^{+} + \mathrm{H_2O} \tag{1}$$

The alkali treatment enhances the affinity between the fibers and the polymer, increasing biocomposite's mechanical properties and water resistance [36]. Treating kenaf fibers with an 8% NaOH solution for 4 h effectively removes hemicellulose and lignin, improves the adhesion between the fibers and the epoxy matrix, increases the flexural strength of the biocomposites by 52%, and enhances the flexural modulus by 46% [124]. Furthermore, similar results were confirmed by Ganesan et al., who tested jute/epoxy composites fabricated with jute fibers treated with 5% NaOH and untreated ones. They found that the mechanical properties of the alkali-treated fiber-reinforced composites increased significantly, with tensile strength, flexural strength, and impact strength improving by 68.9%, 79.1%, and 4.4%, respectively [129]. In the study [130] of the water absorption of sisal and epoxy composites, researchers found that the water absorption of biocomposites made from fibers treated with 5% NaOH was greatly reduced. The treated fiber-reinforced composites absorbed 41% less water than the untreated composites after 600 h of immersion in water at 50 °C. To treat natural fibers with NaOH solution, the relationship between alkali solution concentration and soaking time must be well-controlled to obtain the best treatment results [131], as shown in Table 4. Excessive alkali treatment can cause the fiber texture to become thin and fragile, deteriorating the composite material's mechanical properties [132].

Table 4. Effect of NaOH treatment on biocomposites.

Type of Fiber/Matrix	Treatment Time	NaOH Concentration	Tensile Strength Change	Flexural Strength Change	Water Absorption Change	Ref.
kenaf and glass/epoxy	4 h	8%	-	↑ 52%	-	[124]
jute/epoxy	45 min	5%	↑ 68.9%	↑ 79.1%	-	[129]
jute/epoxy	90 min	6%	↑ 37.47%	↑ 71.46%	-	[127]
jute/epoxy	24 h	5%	↑ 36%	↑ 14.63%	↓ 48.45	[133]
sisal/epoxy	2 h	5%	↑ 39.7%	-	↓ 41%	[131]
bamboo/epoxy	30 min	5%	↑ 50%	↑ 7.36%	↓ 23.81%	[134]
flax/epoxy	30 min	5%	↑ 20.74%	↑ 13.86%	↓ 15.38%	[134]
flax/polypropylene	2 h	5%	↑ 22.8%	-	↓ 22.48%	[135]
PALF/phenolic	3 h	5%	↑ 79.32%	↑ 82.27%	-	[136]
PALF/polypropylene	4 h	5%	↑ 12.01%	↑ 7.98%	-	[137]
kenaf/polypropylene	4 h	5%	↑ 15.35%	↑ 12.82%	-	[137]
kenaf/epoxy	24 h	6%	↓ 34.7%	-	-	[132]

In the table: ↑ indicates an increase in the respective property; ↓ indicates a decrease in the respective property.

6.2. Effects of Nanofillers on Biocomposites

Several factors, including voids within the material, weak matrix-reinforcement bonding, and manufacturing defects, undermine the integrity and comprehensive properties of FRCs [138]. Improving the quality of product processing and incorporating new active ingredients are effective ways to enhance the durability of materials, and adding nanoparticles or nanocomponents to composites is considered an effective strategy [139,140]. Nanoparticles are ultra-fine particles with diameters ranging from 1 to 100 nanometers, and judicious use of them in composites can bring positive effects [141,142]. The specific surface area of nanofillers plays a crucial role in influencing the properties of nanocomposites [143]. Uniform dispersion of nanoparticles in composites can significantly improve the mechanical properties of FRCs [144]. Fine nanoparticles can reach the interfacial sites of fiber and matrix and enhance linking, which improves binding properties. At the same time, the fracture toughness and other mechanical strength of nanocomposites are improved [145]. In addition, nanofillers can fill out the micro holes in the matrix and reinforce the bond between fibers and matrix, playing a positive function on the area between polymers and fibers, whether natural or synthetic [146,147]. Moreover, nanoparticles in composites form chemical bonds and physical interactions with the matrix, reducing the formation and propagation of cracks in the matrix [148]. Using them can significantly improve composites' seawater resistance and flame retardancy [149–151]. Nanoparticle-reinforced fiber composites are the subject of considerable attention due to their remarkable stability, superior mechanical properties, and exceptional capacity to withstand various environmental conditions [152]. Furthermore, different nano-additives can bring different functional properties, modifying the resistance of composites to fit for various environments; for example, proper selection and amounts of nanoparticle fillers can enhance the biocomposite's UV resistance, making it suitable for outdoor applications [153]. The property effects of some synthetic/natural fiber-reinforced composites modified by nanofillers from previous literature are listed in Table 5. Obviously, the addition of nanoparticles in the range of 0.1–2.0% can play a greater role in improving the properties of the composites. There are vast differences in the results when using the same nanoparticle with different ratios to modify fiber-reinforced composites. Furthermore, using the same type and proportion of nanofiller to treat biocomposites, which are made of different materials, produces significantly different results. Additionally, fine and well-dispersed nanoparticles are more favorable for the modification of biocomposite properties [154,155].

Table 5. Effects of nanofillers on biocomposites.

Fiber/Matrix	Filler/Amount	Changes in Properties	Ref.
flax/epoxy	TiO_2/0.7%	↑ 10.95% tensile strength ↑ 20.05% flexural strength ↑ 10.45% impact strength ↑ 18.81% interlaminar shear strength ↓ 31.66% water diffusion coefficients	[156]
flax/epoxy	TiO_2/0.6%	↑ 16.03% tensile strength ↑ 24.66% flexural strength ↑ 16.45% interlaminar shear strength ↓ 43.06% water diffusion coefficients	[157]
PLAF/epoxy	Nano clay/1.5%	↑ 33.33% tensile strength ↑ 14.74% flexural strength	[158]
palm leaf stalk and glass/unsaturated polyester	Nano clay/1.5%	↑ 12.56% tensile strength ↓ 36.92% water absorption (24 h) ↓ 17.72% water absorption (21 days)	[21]
kenaf/epoxy	SiO_2/2%	↑ 20.61% tensile strength ↑ 23.71% compressive strength ↑ 22.88% impact strength	[159]
flax/polypropylene	SiO_2/10%	↓ 23.63% water absorption	[160]
glass/epoxy	Al_2O_3/0.1%	↑ 12% flexural strength ↑ 17% interlaminar shear strength ↓ 17% water diffusion coefficients	[117]

In the table: ↑ indicates an increase in the respective property; ↓ indicates a decrease in the respective property.

7. Marine Application

One of the primary motivations behind the development of FRCs is to modify the seawater corrosion of traditional metal materials. The FRCs, as advanced materials, are ubiquitous in the marine industry, such as boats, competitive sports equipment, and waterpark installations, owing to their lightweight nature and exceptional mechanical properties [161]. Their applications extend to underwater facilities, underwater investigation, military sectors, and numerous other fields [99,162], as shown in Figure 9. Biocomposites have important uses in the fields of water purification and wastewater treatment [163,164], and researchers found that a chitosan-based biocomposite performs well in this field [165]. However, most of the FRCs used in the marine industry rely on synthetic fibers as reinforcement materials, which are not environmentally friendly. Most synthetic fibers are derived from non-renewable resources, such as petroleum. In addition, these synthetic fibers are resistant to degradation in natural environments. Once dispersed in the natural environment, these fine materials are challenging to collect and persist for extended durations, causing long-term environmental pollution [10]. Employing renewable and biodegradable materials can reduce or mitigate the adverse effects of synthetic fibers from the source. Moreover, compared to synthetic fibers, natural fibers have a lower carbon footprint, benefiting from lower energy use in the production process [166]. Natural fiber-reinforced biocomposites are naturally degradable and less toxic, reducing adverse impacts on marine life as well as human health [167].

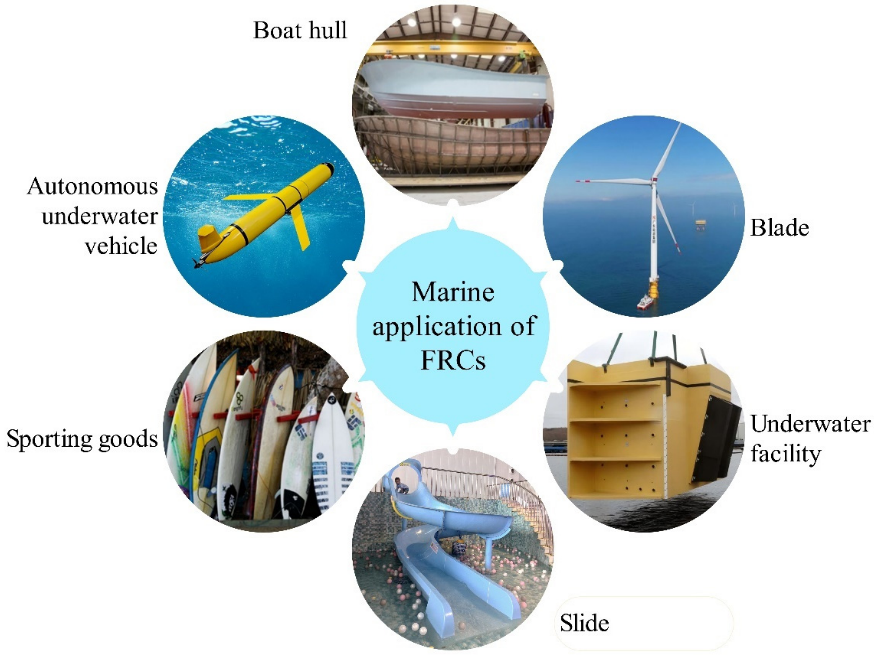


Figure 9. The application of FRPs in marine fields.

Despite the rapid adoption and development of FRCs across several marine industries, their usage remains relatively recent compared to the long history of wood and metal materials in this field. For example, in the ship hull construction field, wood dominated as the primary material for thousands of years [168]; metal materials began to be employed in the 19th century for building larger vessels [12]; and by the mid-20th century, FRCs reinforced with glass and carbon fiber were successfully applied in this field, leading to widespread acceptance and rapid development [34]. The FRC components can be factory fabricated and assembled together in a bolted structure [169–171]. Further research is needed to explore and enhance the marine applications of FRCs, particularly in improving durability and water repellency, resistance to seawater environmental conditions, and developing more sustainable alternatives.

Ships are the primary application of FRCs in the marine environment, and they must be able to overcome harsh environments during their service life. After Ray Greene created a fiberglass sailing boat, the FRCs entered a rapid development period in marine applications. By 1957, Ray Greene had manufactured 175 of the 25-foot Sparkman & Stephens design [34]. In addition, HMS Wilton, a mine-countermeasures vessel measuring 60 m, was applied in naval in 1973. This boat was the first to be made using a glass fiber-reinforced composite for naval shipbuilding [33]. More and more ships were made using these materials. However, with the increasing abandonment of synthetic fiber-reinforced vessels and the associated environmental risks stemming from these composites, the disposal of such hulls has emerged as a critical and pressing issue [172]. Using biocomposites in hull structures offer a sustainable solution, effectively mitigating environmental challenges while ensuring reliable operational performance. A previous article aimed to provide biocomposite reliability in marine environments and useful information to boat designers [173]. They made a racing sailboat using flax/epoxy and balsa wood materials by vacuum resin infusion method, and the boat structure displayed excellent mechanical properties and durability. Other biocomposite boats were created, as shown in Table 6. Furthermore, biocomposites have many advantages, including simple fabrication, easy-obtained material, low carbon emission, nontoxicity, and recyclability. Using new biocomposite materials has a positive impact on the maritime industry.

Table 6. The applications of fiber-reinforced biocomposites on boat hull.

Year	Fiber/Matrix	Marks	Ref.
2009	Bamboo/epoxy	A 6 m boat hull was built using the vacuum bagging method.	[174]
2010	Glass and sugar palm/ unsaturated polyester	The sugar palm fiber was used to replace glass fiber to create a hybrid fiber-reinforced biocomposite boat, and 50% glass usage was reduced in this work.	[175]
2010	Carbon and Flax/polyester	A 6.5 m racing yacht, Araldite, was built with 50% flax and carbon fibers and launched in 2010.	[167]
2011	Flax/PLA	Manufacturing a canoe with 4.4 m length by vacuum forming then autoclave.	[176]
2013	Flax/polyester	A 7 m trimaran, Gwalaz, was created with flax fiber without synthetic fibers, and the boat was launched in 2013.	[167]
2013	Jute/polyester	Tara Tari built a boat, Gold of Bengal, made entirely of natural fiber-reinforced composite, and is the first boat created by jute fiber without any synthetic fibers.	[12]
2021	Bamboo/Bio Resins	A yacht, named FLOKI 6.5, was designed. The boat was made of bio-based and recyclable raw materials and displayed a uniquely sustainable capability.	[12,177]

Aside from maritime vessels, fiber-reinforced biocomposites hold significant potential for various applications in future marine exploration. The sea is a vital resource repository upon which humanity depends for survival, providing food, energy, and minerals. Human efforts to exploit and utilize the ocean's resources have been continuous throughout history [178,179]. Autonomous underwater vehicles (AUVs) are crucial equipment for surveying marine environments, with applications spanning scientific research and military operations [180,181]. Employing biocomposites in AUVs presents potential advantages, including environmental protection demands and a lightweight structure that enhances underwater endurance and maneuverability. These factors make AUVs more suitable for exploring marine resources and conducting marine environmental research [182,183]. However, reports on using ecological composite materials in this field remain limited. Further fundamental research is needed to develop biocomposites, focusing on areas, such as selecting and processing raw materials, advancing composite fabrication techniques, and enhancing mechanical properties and durability. Overcoming these challenges to modify the environmental problem remains a key objective.

8. Conclusions

The FRCs are a sophisticated composite material characterized by lightweight qualities and superior mechanical attributes, extensively utilized across multiple domains in terms of transport, engineering, sport, etc. Nevertheless, conventional reinforcements in FRCs consist of synthetic fibers, including carbon, glass, and aramid, which negatively impact the environment. Plant fibers possess a distinct advantage as a substitute for manufactured fibers. Nonetheless, numerous issues must be addressed when utilizing plant fibers, including the reduction of water absorbency and the enhancement of binding capacity between the fibers and the matrix to improve the environmental durability of biocomposites.

This review addresses the challenges encountered and prevalent countermeasures for the implementation of plant fiber-reinforced biocomposites in marine environments. The hygroscopic characteristics of plant fibers cause volumetric expansion upon contact with water, influencing the mechanical properties of the composites. Consequently, mitigating the detrimental impacts of water in marine environments is the foremost challenge for material applications in this domain, and mitigating water absorption in plant fibers and augmenting fiber-matrix adhesion are the primary methods to enhance mechanical characteristics. The alkaline treatment process is an effective method for modifying plant fibers and improving fiber-matrix bonding. Similarly, oxide nanoparticles have a positive effect on the performance enhancement of biocomposites by improving the mechanical properties of the composites and reducing the water absorption, etc. Moreover, a hybrid fiber-reinforced biocomposite is an appropriate material for maritime applications, combining the benefits of both synthetic and natural fibers. These technologies enhance biocomposites, enabling materials to maximize performance to meet the requirements of more demanding and intricate application environments. Additionally, these improvements in fabrication techniques have significantly enhanced the performance and water resistance of biocomposites, offering promising prospects for their application in marine environments.

Current research indicates that plant fibers possess significant potential for maritime applications owing to their inherent characteristics and superior mechanical qualities. Nevertheless, studies on bio-based fiber-reinforced composites in maritime applications remain limited. Therefore, to promote the utilization and implementation of plant fiber-reinforced composites in marine environments, the following research areas need to be explored to:

1. Enhance the mechanical properties and durability of biocomposites in the dry and wet states;
2. Increase the proportion of renewable plant fibers in biocomposites and reduce the ratio of synthetic fibers used;
3. Improve biocomposites' environmental resistance, including against UV and underwater biopollution;
4. Optimize manufacturing processes to reduce costs and facilitate industrial applications.

In short, following the continuous development of manufacturing technology, bio-based fiber-reinforced composites will have a wider prospect in the marine field.

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