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*Review Article*

# Nontraditional Natural Filler-Based Biocomposites for Sustainable Structures

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In recent years, there has been a growing awareness and demand for global sustainability, as well as a mandate for the use of renewable and environmentally sustainable materials and processes. Due to which, massive efforts are being made to develop and nurture the next generation of composite materials that are energy efficient, environmentally friendly, and biodegradable. Light weight, lower coefficient of thermal expansion, and comparable tensile strength exhibited by natural fibers render them the choice for use in several industrial products and applications over the last decade. Natural fibers as the reinforcing entity are pitted against their synthetic variants primarily because of the superior aspects like biodegradability and excellent strength-to-weight ratio. This article presents the review on various nonconventional natural fibers such as tamarind seed and shell, Luffa cylindrica, groundnut shell, coconut coir, papaya bast, okra, and Ashoka tree seed. The flow of the chapter includes the introduction, extraction methodologies, and fabrication, and investigations of mechanical properties, applications, and sustainability are dealt in detail for nontraditional natural fibers. The okra fibers possess greater tensile strength of up to 262.8 MPa in comparison with other fibers, while the Ashoka tree seed fibers are known to possess a maximum flexural strength of up to 125 MPa. Further, these fibers are used as reinforcements in potential applications in interiors and automobile and aircraft panels and wood-based particle board composites owing to the increase in tensile and flexural strengths of composites.

## 1. Introduction

Development and use of natural fiber-based composite materials are steadily increasing due to the superior properties offered by these fibers and their abundant availability [1]. In the recent past, various types of plant and fruit fibers are explored and studied. The natural fibers, which are extracted from various plants, mineral sources, and animals, seem to have better specific properties and weight saving potential. The properties of plant fibers are dependent on the age of plant, extraction technique adopted, and interaction with the environment [2, 3]. Nevertheless, natural fiber composites find applications in automobiles, naval, household items, and chemical industries [4].

The plant fibers such as banana [5, 6], jute [7, 8], kenaf [9, 10], abaca [11, 12], pineapple [13], sisal [14, 15], agave [16], flax [17], hemp [18], cotton [19], and roselle [20] are the traditionally used natural fibers for making the composites. Various thermoplastic [21, 22] and thermosetting [23, 24] resins are used as matrix material. Most of the studies are related to the mechanical characterization of these novel composites. The quality of these composites depends on the mode of adhesion and the strength of the constituent elements. Few researchers have demonstrated that the surface treatment of the naturally available fibers enhances the adhesion between the constituents, leading to improved properties owing to a better load transfer mechanism. Animal fibers such as wool obtained from sheep, goat hair, horsehair, and yak are explored by researchers [25]. The fiber extraction methods and fabrication of composites are dealt in these articles [26].

Due to the desired properties exhibited by these composites, researchers have taken steps to explore the possibilities of extracting and using these new materials for several engineering applications [27] and in particular low load-bearing structures [28]. Hence, in the present study, the processing methodology, characterization methods, and their consequences are elaborated for newer natural fibers and animal fiber composites. In this paper, a comprehensive review of the newly explored natural fibers such as tamarind, Ashoka tree, fish scale, and groundnut shell which are extracted and studied for their mechanical and thermal properties is dealt in detail. The review articles presented the mechanical properties and applications of natural fibers such as jute, banana, kenaf, and sisal for the multiutilities [29] and structural applications [30]. However, the findings on the nontraditional natural fiber-reinforced biopolymer composites are incipient to utilize them for structural applications. In this regard, the present chapter provides a detailed review of the use of nontraditional natural fibers for advanced applications, particularly related to load-bearing low weight structural applications. Figure 1 gives a schematic of some of the nontraditional natural fiber sources that can be utilized for the synthesis of biocomposites for structural applications.

These natural fiber sources provide an additional advantage of high strength-to-low weight ratios. The densities of some of the conventional natural fibers in comparison with the nonconventional fibers are herewith given in Figure 2.

From the graphs of variation of densities for different natural fibers, it is herewith evident that the nonconventional natural fiber sources like gold cane, vakka, jackfruit stem fiber, and waste boom grass have relatively lower densities. However, they provide higher strength, thus providing composites with high strength with relatively lesser weights.

Jagadeesan et al. [31] have accomplished research on sesame oil cake biomass waste-derived cellulose microfillers reinforced with basalt/banana fiber-based hybrid polymeric composite for lightweight applications and have ascertained the need for nontraditional filler materials for natural fiber-reinforced biocomposites. Further, Rantheesh et al. [32] have worked on the characterization of novel microcellulose from *Azadirachta indica* A. Juss agroindustrial residual waste oil cake for futuristic applications. The higher cellulose content (73.53%), better crystallinity (66.23%), lower density (1.59 g/cm$^3$), considerable thermal stability (335.71°C), kinetic activation energy (83.06 kJ/mol), particle size (17.93 $\mu$m), and good surface roughness (47.004 nm) make neem cake cellulose

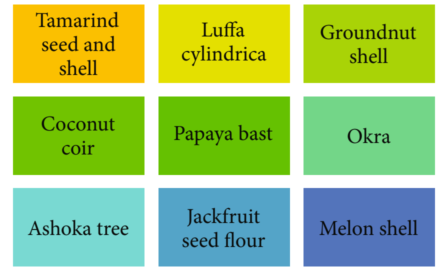



Figure 1: Schematic of some of the potential nontraditional natural filler and fiber sources reviewed in the article.

(NCC) suitable to be incorporated as a biofiller material in polymer matrices to manufacture ecofriendly composites.

Sunesh [33] has worked on the novel agrowaste-based cellulosic microfillers from *Borassus flabellifer* flower for polymer composite reinforcement. The results have showed that an agricultural residue can be converted into a valuable microsized cellulosic filler material for polymeric composite applications that can withstand processing temperature up to 200°C.

Ramesh et al. [34] have reviewed the influence of filler material on properties of fiber-reinforced polymer composites and have concluded that the use of filler material alongside the natural fibers enhances the performance attributes of the composite materials.

R Vijay et al. [35] have carried out the thermomechanical characterization studies on bio-fillers, namely *Azadirachta indica* seed powder, spent Camellia sinensis powder, and their combinations filled with jute fabrics in the epoxy matrix. The synthesized composites are evaluated both analytically and experimentally and compared with the neat epoxy. The test results revealed that the *Azadirachta indica* seed powder-filled composites exhibit better mechanical properties with lesser voids, while spent Camellia sinensis powder and its composites exhibit better thermal stability.

Ravikumar et al. [36] have explored the consequence of ecofriendly sodium bicarbonate treatment on the drilling behavior of jute fiber-reinforced polyester composites. The use of response surface methods for optimizing the composition, curing time, and process parameters has improved the machinability of natural filler-based jute fiber-reinforced composites.

Sumesh et al. [37] have focused on the impact of biowaste filler in mechanical applications. The biowastes from banana, pineapple, and coconut plants were used for preparing banana fly ash (BFA), pineapple fly ash (PFA), and coir fly ash (CFA) fillers. The results revealed that the flexural and impact properties enhanced up to 22.11% and 21.77%, with filler incorporation. The SEM results exhibited good bonding nature due to the application of filler powders. The EDX results proved the presence of silica and other inorganic content in the polymer composites adding to the improvement in properties of the composites.

Sumesh et al. [38] have studied the effect of banana, pineapple, and coir fly ash filled with hybrid fiber epoxy-based composites for mechanical and morphological study

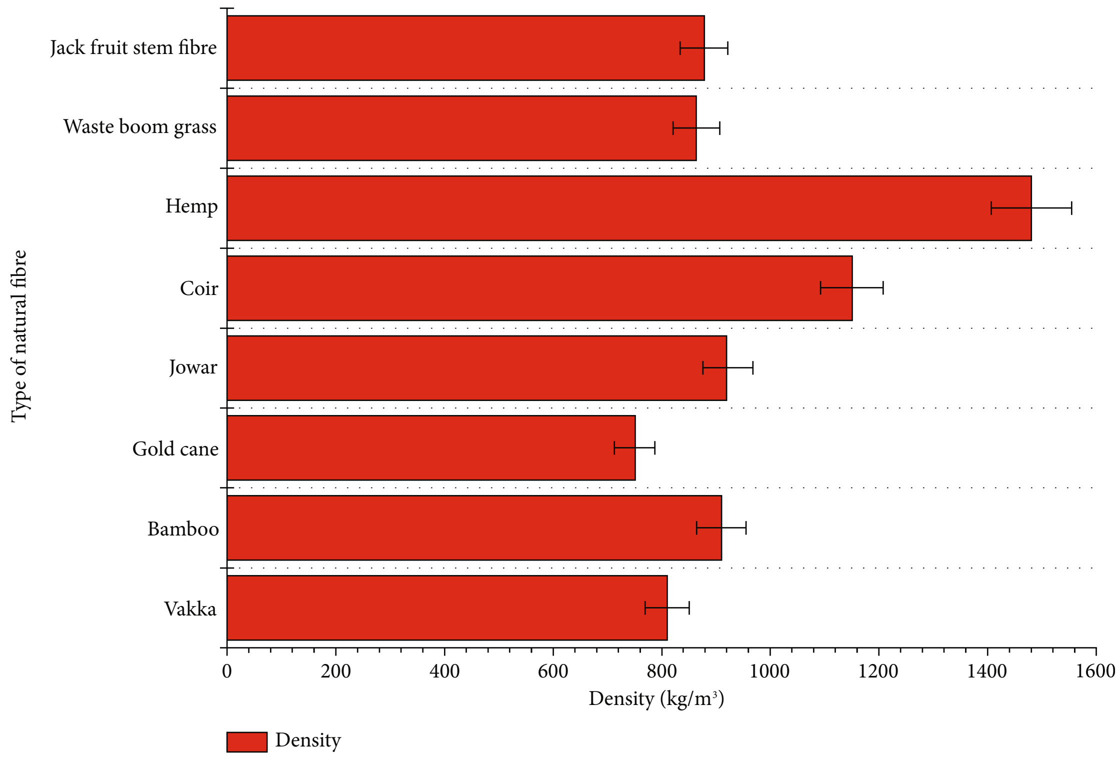



Figure 2: Type of natural fiber and their densities.

and have reported that the inclusion of the filler powders enhances the tensile strength from 23.78 to 33.79 MPa owing to the improved adhesion between the fibers and the matrix due to the addition of the filler.

Sumesh and Kanthavel [39] have carried out optimization studies by the Taguchi method and gray relational analysis to evaluate the influence of $Al_2O_3$ nanopowder, NaOH treatment, and compression pressure on free vibration and damping behavior of natural hybrid-based epoxy composites. The gray relational analysis concluded that 20 wt.% sisal and 15 wt.% coir with 2 wt.% nanoalumina and treatment with 5% NaOH at a compression pressure of 10 MPa provided optimized working condition for synthesis of the composite plates for better vibration behavior.

Sumesh et al. [40] have further studied the influence of different parameters in tribological characteristics of pineapple/sisal/$TiO_2$ filler incorporation by Taguchi optimization techniques, and the optimization results showed lower specific wear rate (SWR) by the incorporation of high $TiO_2$ filler (5 wt.%) addition in the pineapple/sisal fiber-reinforced epoxy composites.

The nontraditional filler-based natural fiber composites have been tried with ANN, GRA, and Taguchi approaches for optimization. In this regard, the works of Sumesh et al. [41] aim to optimize the mechanical performance of ramie/kenaf fibers under various parameters using GRA/TOPSIS methods.

This article encompasses a complete review of the literatures related to the use of nontraditional filler materials to improve the characteristics of the composites, which is a novel concept in comparison with the existing review particles based more on synthetic fibers and synthetic filler materials, as the need for ecocompatible and sustainable composite materials for structural applications is evolving day by day.

## 2. Different Nontraditional Filler Materials

The sourcing of different nontraditional natural fillers is an important aspect of the study. In this regard, a detailed review of the different nontraditional natural fibers is essential. Thus, the extraction methods of the naturally available materials like *Tamarindus indica* (tamarind) seed, *Arachis hypogaea* (groundnut) shell, coconut shell powder, Luffa cylindrica, coir powder, Abutilon (Indian mallow plant), and *Saraca asoca* (Ashoka tree fruit) are reviewed and discussed in detail in this section.

### *2.1. Mono Nontraditional Natural Filler-Based Biocomposites*

*2.1.1. Tamarindus indica.* The *Tamarindus indica* is also known as the tamarind plant. The plant is widely grown in India, and its seed is used to make a hybrid composite. Initially, the tamarind seed (Figure 3) is extracted manually from the fruit and washed and treated with alkali solution to remove the stickiness. Later, the brownish layer of seed is removed and ground to a fine powder using commercially available flour grinders. Tamarind shells are washed to remove any impurities and dried to render them free of moisture. They are then ball milled into powder form. Epoxy resin of grade HSC-7101 is used as a matrix and HSC-9222 as the hardener. The composite was prepared using the hand

Figure 3: Tamarind seed with brown shell.

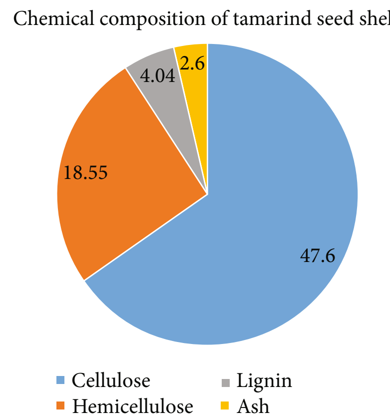



Figure 4: Chemical composition of the tamarind seed shell [43].

layup method. To obtain enhanced mechanical behavior such as impact resistance, shock resistance, vibration resistance, and water resistance, thixotropic high viscosity epoxy resin can be used with a variety of hardeners to create natural fiber-reinforced composites. Prior to the layup, tamarind powder and epoxy are mixed according to weight ratios such as 30 : 70 and 50 : 50. The samples were cured for 1 day. Cured samples were subjected to mechanical tests like the tensile test (ASTM D3039-20) and flexural test (ASTM D7264-20), followed up by water absorption test (ASTM D785). Baig and Mushtaq [42] concluded that laminates containing less tamarind shell have a lower rate of water absorption and thus a higher flexural modulus and bending strength, whereas laminates containing more tamarind shell have a higher tensile strength and hardness number.

Several findings have been reported on the variation of the mechanical characteristics of the composites with the varying wt.%. The research on the tamarind seed shell by Naik et al. [43] has revealed that the enhanced characteristics, the tensile strength, and Young's modulus of composites are 27.69 MPa and 362.39 MPa for 10% and 20% composition of tamarind seed shell in polymer matrix, respectively. The impact strength increased steadily up to 30% composition of tamarind seed shell. The hardness number varies from 72.33 to 53.33 HRN for the composite materials. This improvement is attributed to the microcoring and stronger bonding of the matrix and the reinforcements, particularly due to the inoculation brought about by the tamarind seed. The chemical composition of the tamarind shell comprises of greater weight percent of cellulose, which makes it a potential candidate for its use in composites, particularly for enhanced flexural and tensile strengths. Figure 4 gives the chemical composition of the tamarind seed shell.

*2.1.2. Groundnut Shell.* The biological name for groundnut is *Arachis hypogaea*. The powdered forms of groundnut shell (Figure 5(a)) are used as reinforcements. To extract the powder, the dirt present over the groundnut shells is removed by cleaning them with plain water. This is followed by alkali solution (of 4% concentration) treatment for around 2 hours. The groundnut shells are then washed again with plain water and sun-dried. The sun-dried shells are then ground into a fine powder (Figure 5(b)) and then sieved with different grades (0.5, 1, 2, and 3 mm) of sieve [44].

The particle board with varying ratios of groundnut shell powder and epoxy matrix (60 : 40, 65 : 35, and 70 : 30) is prepared by manual stirring and open molding and tested. The prepared composites are tested for its tensile behavior (ASTM D638), impact (ASTM D256), flexural behavior (ASTM D790), and water absorption (IS: 2380 (PART XVI)). The results showed that the composites exhibited moderate mechanical properties and low water absorption. Groundnut shell particle composites can be used as a substitute for wood in the production of particle board for use in indoor environments. This material has the potential to be used in structural, packaging, and other general applications [45].

*2.1.3. Coconut Shell Powder.* Coconut shell is an agricultural residue, usually disposed of through open burning. Toxic gases have been emitted from the open burning phase and can therefore be detrimental to human health and the environment. Thus, to reduce the risk of pollution, researchers have developed a new technology by using agrowastes to produce biocomposites. Coconut shell powder (CSP) is a solid nonfood waste, which can be potentially exploited to reduce the usage of synthetic fiber. Coconut shell is also a low-cost and low weight material that can be used to reduce the production cost of and fuel consumption for transportation [46]. This review has focused on the research carried out on the CSP loaded into different types of matrices, highlighting the fundamental, mechanical, physical, and thermal properties of CSP composites. This article also provides critical review of the development for CSP composite and the summary of the results presented in the literature, focusing in the properties of CSP with polymeric matrices and the application design for economical products.

*2.1.4. Luffa cylindrica.* The Luffa cylindrica, commonly known as sponge gourd, is mostly grown in European countries. The sponge gourd contains an inner fiber core and an outside mat core, with a multidirectional array of fibers forming a natural mat. They are extracted just by separating

(a) (b)

Figure 5: (a) Groundnut shell. (b) Groundnut shell powder after pretreatment [44].

Figure 6: Luffa cylindrica fiber [47].

and peeling the skin manually. Figure 6 gives the photographic image of the Luffa cylindrica fiber.

Saw et al. [47] and Noone et al. [48] investigated the effect of alkali treatment on Luffa cylindrica-reinforced composites and compared with glass-reinforced composites. Alkaline treatment is found to disrupt hydrogen bonds and helps to remove certain quantity of wax, lignin, and oils present on the exterior surface of cell walls of the fibers. It also depolymerizes cellulose and uncovers the small length. The spongy mat is treated with alkaline solution (5% NaOH), $ClO_2$ (oxidation), and furfuryl alcohol (FA) to remove impurities and to increase surface topology. Hand layup method and compression molding are employed to prepare the composites of treated and untreated Luffa fiber mats with epoxy resin in a mold of size $100 \times 100 \times 3\,mm^3$. The matrix-to-fiber weight ratio is kept constant at 70 : 30 in each composite. The tensile properties obtained for untreated Luffa fibers, alkali-treated Luffa fibers, and grafted Luffa fibers are represented in a tabular format for comparison. FA-grafted Luffa fiber composites show enhanced tensile properties as compared to alkaline-treated and untreated Luffa fiber composites, due to improved adhesion among the fibers and the matrix material. The properties obtained from the works of Saw et al. [47] and Noone et al. [48] are summarized in Table 1. The extracted Luffa fibers are further treated with 10 to 15 ml of sodium hydroxide (NaOH) solution and thoroughly dried for the preparation of composites. The composite material specimen is developed by the traditional hand layup process. The Luffa fiber composite ratio of 80 : 20 is prepared using LY556 (epoxy resin) and HY951 as the hardener and cured. The mechanical properties of the hybrid Luffa and glass fiber composites obtained from the works of Sreeramulu and Ramesh [49] are also summarized in Table 1. Luffa treated with alkaline solution showed better results as compared to glass-reinforced and untreated Luffa fiber, which indicates the possibility to replace glass fiber composites in structures of automobiles and other general applications.

The inclusion of the Luffa fiber reinforcements improves the mechanical characteristics of the composites owing to the stronger bonding between the matrix and reinforcements facilitated by the cellulose content in the fiber that enhances the adhesive strengthening.

Premalatha et al. [50] have studied the structural and thermal properties of chemically modified Luffa cylindrica fibers and have reported that the appropriate stearic acid treatment of the fibers improves the thermal stability and bonding capabilities.

*2.1.5. Ashoka Tree Seed.* Ashoka tree is commonly known as *Polyalthia longifolia*. The seeds collected after harvest are washed, and upper coats are removed and retreated with plain water and dried under sunlight for 12 days. The dried seeds are ball-milled to a particle size of 25 $\mu$m to 50 $\mu$m. The combination of powder and untreated vinyl ester is used to fabricate composites using compression molding. The composite is baked in an oven followed by accelerators N-dimethylaniline and styrene; the catalyst and promoters like methyl ethyl ketone peroxide (MEKP) and cobalt naphthenate are used. The prepared samples are cured for one day under a pressure of 100 kPa. The mechanical properties and thermal properties are determined by various ASTM standards like ASTM D638 for tensile test, ASTM D790-10 for flexural, and ASTM D256 for impact strength. Thermal properties like thermogravimetric and differential thermal analyses were conducted, and water absorption test was done for the sample specimen where the sample was submerged in water for 1 day. The test results revealed that

Table 1: Mechanical properties of hybrid Luffa and glass fiber composite [47–49].

| Sample | Tensile strength (MPa) | Flexural strength (MPa) | Tensile modulus (MPa) | Flexural modulus (MPa) |
|---|---|---|---|---|
| Untreated Luffa fiber [47] | 178.20 | — | 4263.84 | — |
| Alkali-treated Luffa fiber | 192.70 | — | 5184.62 | — |
| FA-grafted Luffa fiber | 226.40 | — | 5865.70 | — |
| Luffa fiber under alkali treatment/epoxy [48] | 34.1 | 57.2 | — | — |
| Untreated Luffa fiber/epoxy | 31.4 | 54.4 | — | — |
| Glass-reinforced epoxy composite | 23.3 | 50.2 | — | — |
| Luffa fiber (5 gm)+glass fiber (15 gm) [49] | 17.97 | 106.67 | 1331.24 | 4858.08 |
| Luffa fiber (10 gm)+glass fiber (10 gm) | 4.51 | 7.86 | 327.39 | 490.94 |
| Luffa fiber (15 gm)+glass fiber (5 gm) | 9.13 | 14.10 | 446.16 | 760.08 |

(a) (b)

Figure 7: (a) Okra plant. (b) Extracted okra fiber [52].

Table 2: Mechanical properties of okra fiber composites [52].

| Tensile strength (MPA) | Tensile modulus (MPa) | Flexural modulus (MPa) | Impact strength (J/mm$^2$) | Density (g/cc) |
|---|---|---|---|---|
| 263.5 | 7319.996 | 48.85 | 1.66 | 1.11 |

tensile strength increased up to 30 wt.% which leads to the use of these composites in the interior part of an automobile and aircrafts. Going beyond this limit, a weak interfacial adhesion ensued between the filler and matrix leading to a reduction in the strength. The addition of up to 35 wt.% filler to the samples had a significant impact on the flexural and impact strengths. The composite flexural and impact strengths were raised by factors of 1.60 and 2.63, respectively, over neat vinyl ester resin [51].

*2.1.6. Okra (Ladies' Finger) Plant.* Okra (gumbo or ladies' finger) plant (Figure 7(a)) is treated with water for 6 days to remove dirt from the roots of the stem. The fibers are then extracted from the stem of the plant (Figure 7(b)) and are dried under ambient conditions for 7 days. The fabrication process used is manual hand layup. The fibers are placed in layers so that they bind up with the epoxy resin. Catalyst and accelerator are added to reduce the curing time. A small load is applied over the mold and cured for 24 hrs. Postcuring is carried out for 2 hours, at 70°C. Mechanical tests like tension (ASTM D638-89), 3-point bending (ASTM D790 M-86), impact test (ASTM D256-10), and density test are performed. Table 2 depicts the mechanical properties of 60% volume fraction of fibers which exhibit optimal properties as compared to any other volume fractions from among the tested specimens. The study also showed that an increase in the volumetric fraction resulted in a significant improvement in tensile strength and tensile modulus. Figure 4 shows the images of the okra plant and the extracted okra fiber considered in the works of Potluri et al., while Table 2 gives the mechanical properties of the okra fiber in composites [52].

*2.1.7. Artocarpus heterophyllus (Jackfruit). Artocarpus heterophyllus* is commonly known as jackfruit. The rough skin of jackfruit is used as reinforcement for composite material in particulate form. Jackfruit skin is peeled manually, crushed, dried, and finally ground to 0.5 mm size. Ground skin particles are sieved to obtain 250 $\mu$m to 500 $\mu$m particulates. To remove excess lignin, the holocellulose bleaching is carried out on particles. This is further washed with plain water followed by heating to a temperature of around 70°C. For removing debris and odour, additives like sodium chlorite and acetic acid are added every 1 hr over a time period of

5 hrs. Initially, the uniform mixing of particles and polylactic acid (PLA) is carried out in Brabender Plasti-Corder, where the particles are infused at intermediate stages at a temperature of 170°C and mixed for 10 mins. The sample is then dried for 1 day at a constant temperature of 60°C. The composite granules obtained are fed in compression molding and sheets are obtained. The specimens from the prepared sheets are cut as per the ASTM standards and tested. Characterization such as infrared spectroscopy, scanning electron microscopy, and tensile testing was done under ASTM D638, and differential scanning calorimetry, thermogravimetric analysis, degradation test, and antimicrobial activity are performed. The results showed that the jackfruit skin can be used as the reinforcement up to 30% by weight and save the usage of plastics. The tensile characteristics of the composites improved when the fiber content was increased [53].

*(1) Jackfruit Seed Flour*. Marzuki et al. [54] studied the tensile properties of jackfruit flour reinforced in low-density polyethylene matrix. To start with, seeds are ground to particle sizes of 63 $\mu$m to 100 $\mu$m and are dried in an oven at 60°C. The content is mixed thoroughly, and the sample is extruded through a twin screw extruder at a temperature of 150°C and motor speed of 50 rpm. Extruded samples were then molded according to ASTM D638 for tensile test. The results showed that the use of higher percentage of filler content is not recommended due to the decline of tensile properties of the blends.

*2.1.8. Melon Shell (Abutilon).* Melon shell, also called cantaloupes, is dried under the sun for 7 days and ground. The resulting content is called uncarbonized melon shell. The dried shell is jammed in a graphite crucible and heated to 1200°C in a muffle furnace for the formation of carbonized melon shell. Hand layup technique is used to fabricate the samples using epoxy matrix LY556 and hardener of HY951. Different weight ratios (5, 10, 15, 20, and 30 wt.%) of carbonized and uncarbonized melon shells are attempted. The samples thus obtained are tested for mechanical properties like ASTM D638 for tensile behavior and ASTM D256-10 for impact behavior. The results showed weight saving potential with increase in carbonized melon content in epoxy matrix. As the amount of melon shell particles in the epoxy matrix increases, the compressive strength, elastic modulus, dielectric constant, and capacitance of the composites increased. The epoxy/carbonized melon shell composites had higher tensile strength, modulus, and impact energy than epoxy/uncarbonized melon shell particle composites. Melon shell particles added to the epoxy matrix boosted tensile strength and impact energy to a maximum of 20%. Melon shell particles can be employed to improve the mechanical and electrical properties of epoxy matrix composites for indoor and outdoor structural applications, according to the findings [55].

*2.1.9. Fish Scale.* Sekaran et al. utilized fish scales of bony fishes (*Osteichthyes*) as the reinforcing component in their composite material. Fish scales like ctenoid scales or ganoid scales are used to develop the composites using epoxy (LY556) as the matrix component and HY951 as the hardener. The prepared samples are tested for tensile behavior (ASTM D638), flexural behavior (ASTM D790), impact strength (ASTM D256), and hardness (ASTM E18-19). The sample with the volume fraction of 40% fish scale powder showed best results for tensile load; the sample with volume fraction of 30% fish scale powder showed best results for flexural load; and the sample with volume fraction of 25% fish scale powder showed best results for impact loads. The hardness of the composite material increased with an increase in the volume fraction of fish scale powder. The fish scale-reinforced polymeric composites are potential materials for pipes to carry pulverized coal in power plants, conveyor belt rollers, and low-cost housing materials [56].

Babu et al. reinforced fish scales in particulate form with epoxy grade of LY556 and hardener of HY951 and developed a new composite material using manual hand layup process and compression molding. Weight fractions of the filler were varied in percentages of 10, 15, 20, 25, 30, 35, and 40 in the epoxy matrix. Mechanical characteristics of the above samples were studied. It was found that increasing the filler material in the composite sample brought about an improvement in its mechanical properties to some degree. An optimum tensile strength is achieved at 30 percent weight fraction of fish scale filler. For 30% and 25% weight fractions of filler material, optimum flexural and impact strength are obtained, respectively. However, adding more fish scale fillers decreased the mechanical properties, which may be due to inadequate dispersion of filler in the epoxy matrix. The findings are very promising, and fish scale natural filler can be used with epoxy resin to make composite materials useful in applications such as [57].

Abhishek et al. have worked on the development of new hybrid Phoenix pusilla/carbon/fish bone filler-reinforced polymer composites and have reported that the addition of bionanofillers in hybrid polymer composites permits the enhancement of the tensile strength, flexural strength, and hardness by 22.5, 200, 100, and 15.2%, respectively [58].

*2.1.10. Papaya Bast Mass.* Coura et al. [59] have extensively worked on the papaya bast fibers that were extracted from the stem of the plant and soaked in water for 15 days. The fibers are then extracted, washed with plain water, and sun-dried for 7 days. The fabrication process used was manual hand layup and followed by compression molding. Epoxy resin of grade RenLam-M and Aradur HY96 as the hardener was used as the matrix. The fibers were cut to 5 mm to 20 mm lengths before the hand layup. Composite material specimens were prepared with 40% to 60% weight fraction of fiber content. The sample was pressured cured for 1 day at a temperature of 50°C. Mechanical testing of the samples is carried out as per ASTM standards: tensile test (ASTM D638-14), flexural test (ASTM D790-15), and density of fiber (ASTM D3800-99). They have concluded that mechanical properties of laminates prepared through longitudinal alignment of the bast fibers and without any internal voids showed superior tensile strength.

*2.2. Hybrid Nontraditional Natural Filler-Based Biocomposites*

*2.2.1. Tamarind Seed and Shell-Based Filler-Based Hybrid Biocomposites.* The hybrid composites of tamarind powder, Borassus tree trunk, and palmyra palm fiber (treated and untreated) and epoxy (LY556) matrix resin with hardener (HY906) are prepared with compression molding. The prepared composites are tested for its tensile behavior (ASTM D3039M), flexural behavior (ASTM D790-20), impact (ASTM D256-20), and active absorption (ASTM D590-20). The results showed that the particulate form of tamarind seed in the composite increases the strength significantly [60]. These types of composites have potential industrial applications such as room partitioning and false roofing.

Felix Sahayaraj et al. [61] have studied the effect of hybridization on properties of tamarind seed nanopowder incorporated Luffa cylindrica fruit waste fiber-reinforced polymer composites. They have reported that the tensile and flexural properties of the composites improved owing to the stronger bonding due to the hybridization reactions between the tamarind seed shell and the fiber in the matrix. Thus, the tamarind seed shell is a potential candidate for its use in real-time applications for improving the properties of the natural fiber-reinforced polymer composites.

*2.2.2. Coconut Coir and Groundnut Shell.* The hybrid of coconut coir and groundnut shell composites was formed and compared for its tensile and flexural strengths. The coconut coir and groundnut shell were pretreated with NaOH and washed with water and dried under the sun for 1 day. The fibers were crushed to different grain sizes of 1 mm, 1.5 mm, and 2 mm. Fabrication of the composite is done by hand layup and compression molding. Epoxy grade of LY556 was used, followed by a hardener HY951. Sample of composites was prepared by weight ratios of epoxy to fiber with different grain sizes. The samples are then cured for 15 hours. Water absorption test was carried out by keeping the samples under water for 1 day. The results revealed higher flexural strength and a lower rate of water absorption when the particulate fiber size is 1 mm. In respect of mechanical properties, coir fiber composite showed superior behavior as compared to groundnut shell-powdered composites [62]. Such properties will make these composites as the better substitutes for wood-based material in many applications. This composite is also having the potential for its use in structural, packaging, and other general applications.

*2.2.3. Coconut Shell Powder and Areca Palm Fibers.* Hybrid composites with continuous areca palm fibers and coconut shell in particulate form as the reinforcements and epoxy matrix are fabricated by hand layup technique. Initially, the areca fibers are extracted from the stem of the plant and pretreated in plain water for 15 days. The dry coconut shells are broken into smaller pieces and then ground to fine particles. The particles are then sieved using 150 $\mu$m sieve. Physical tests like density test, void content, and water absorption tests are performed as per ASTM D 792-21, ASTM D 2734-70, and ASTM D 570-98, respectively. The thermal conductivity of the prepared samples is also investigated using ASTM E1530. The results revealed that areca-coconut shell powder/epoxy composites are considerably influenced by the fiber loading. Thermal conductivity decreased with an increase in the fiber loading. Also, with an increase in fiber loading, the theoretical and experimental densities of hybrid composites see a fall, whereas void content and water absorption capacities of hybrid composites were found to increase [63].

*2.2.4. Coconut Shell and Tamarind Shell Powder.* Somashekhar et al. [64] used coconut shell in powdered form and strained in standard sieve BS 1377-1990. The particle size was around 300 $\mu$m. Polyethylene pellets were crushed in a shredder, and the tamarind shell was powdered in a grinder. Hand layup was used to create samples of three distinct composites. Epoxy resin was used as a matrix, and five separate combinations of constituents were generated by varying the matrix proportion from 5% to 25%. The samples were hot pressed at 140°C for 15 to 20 mins at a pressure of 2 MPa. The samples were cured at room temperature for 30 mins. Mechanical properties, water absorption test, and hardness tests were performed. It was discovered that combining tamarind shell powder with coconut shell powder enhanced the tensile property by around 50%. The optimum results and mechanical properties are obtained when the content is 50% coconut shell powder, 5% tamarind shell powder, and 45% epoxy resin.

## 3. Comparison of the Strength Characteristics of Different Nontraditional Natural Fibers and Their Composites

The nontraditional natural fiber-reinforced composites have significant advantages over the conventional composites due to the higher cellulose and hemicellulose contents that enhance the adhesive characteristics of the composites owing to stronger bonding and greater strength [65]. These materials are critically useful for their applications in sustainable solutions, viz., automotive dashboards, window panels, doors, and other components ranging from aerospace to marine domains [66]. Table 3 gives the comparisons of the mechanical properties of some of the nontraditional fiber-/filler-reinforced composites.

The critical review of some of the properties of these nontraditional natural fiber-reinforced composites has given a comprehensive picture of their performance capability, and it is herewith noted that the tensile, flexural, and impact properties are important for using these composites in structural applications. A comparative evaluation of the mechanism behind these enhanced properties is accomplished by morphological studies and is reported in this section. The increase in the mechanical properties is attributed to the stronger bonding between the fiber and matrix due to the homogeneous distribution of the filler in the matrix. From the works of Hariprasad et al. [69], the *Sansevieria trifasciata* fiber-reinforced polyester composites containing 15% $SiO_2$ exhibit higher mechanical strength due to the homogeneous distribution of the filler in polyester because

TABLE 3: Comparisons of the mechanical properties of some of the nontraditional fiber-/filler-reinforced composites.

| Authors, year | Natural reinforcements used | Matrix used | Major properties studied |
|---|---|---|---|
| Felix Sahayaraj et al., 2021 [67] | Tamarind (*Tamarindus indica* L.) seed nanopowder incorporated jute-hemp fibers | Epoxy resin | The authors have studied the tensile, flexural, and interlaminar shear strength and impact properties. The tensile strength and modulus of the composites are 39.52 MPa and 2.648 GPa, and the flexural strength and modulus values are 89.62 MPa and 9.24 GPa, respectively. The impact strength and ILSS values were found to be 2.35 J and 3.62 MPa. |
| Iyyadurai Jenish et al., 2022 [68] | *Cissus quadrangularis* stem fiber with red mud nanofillers | Epoxy resin | The authors have studied the hardness and wear characteristics of the composites. The hardness of the samples increased by 10.7% for 5 wt.% red mud addition and 14.2% for 10 wt.% of red mud addition. |
| Hariprasad et al., 2022 [69] | Sansevieria trifasciata fiber with $SiO_2$ and B4C filler | Polyester resin | The authors have studied the tensile, flexural, and impact properties. The polyester resin-based composite with 20 wt.% STF fiber and 15 wt.% $SiO_2$ exhibits a maximum flexural strength of 103.58 MPa and impact strength of 27.4 kJ/m$^2$ in comparison with the neat and other compositions of the composites. |
| Felix Sahayaraj et al., 2022 [61] | *Tamarindus indica* seed powder (TISP) incorporated *Luffa cylindrica* fruit (LCF) | Epoxy resin | The authors have studied the density, tensile, compression, flexural, impact, hardness, and water absorption behavior and thermogravimetric analysis. The results revealed that the inclusion of 7.5 wt.% TISP resulted in better physicomechanical and thermal properties. |
| Premkumar et al., 2022 [70] | Sansevieria trifasciata fibers | Polyester resin | The authors have studied the tensile, flexural, and impact strength of NaOH-treated and NaOH-untreated fiber-reinforced composites. It is reported that the tensile strength of the composites increases up to 48.47 MPa, the flexural strength up to 69.17 MPa, and the impact strength up to 16.34 kJ/m$^2$, with the inclusion of treated fibers. |
| Prem Kumar et al., 2022 [71] | Jute/snake grass/kenaf fiber *Annona reticulata* seed filler addition | Epoxy resin | The authors have studied the tensile, ILSS, and wear properties of the composites. The tensile, flexural, inter-lamina, and impact strengths of the composite with an equal amount of snake grass and kenaf fibers filled with *Annona reticulata* seed filler were 50.12 MPa, 132.53 MPa, 1.952 MPa, and 2.23 J, respectively. |

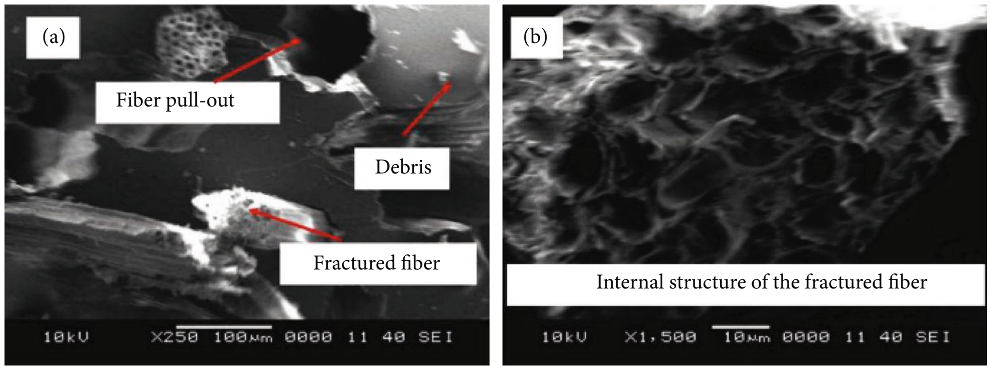
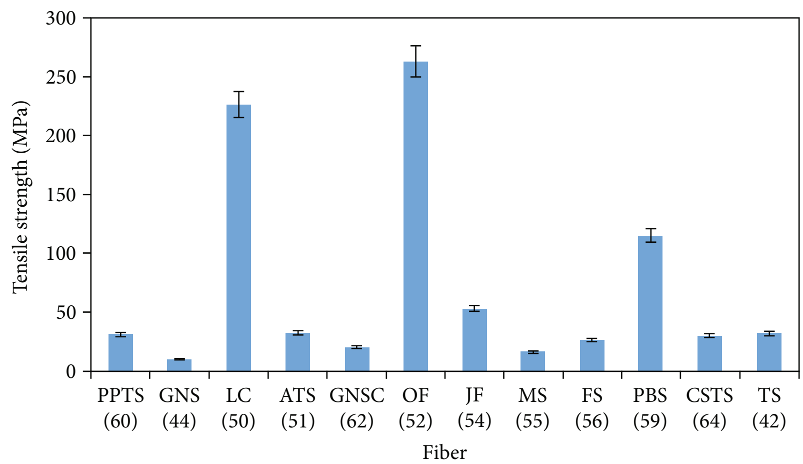
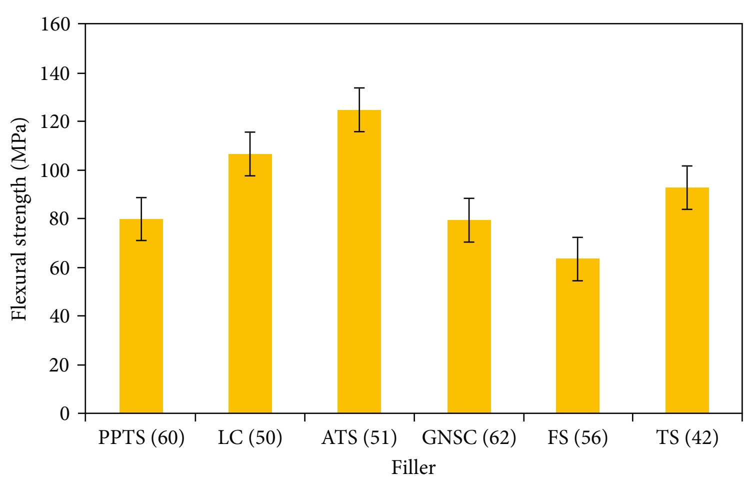



Figure 8: SEM images of *Sansevieria trifasciata* fiber-reinforced polyester composites with (a) 15 wt.% $SiO_2$ and (b) 20 wt.% $SiO_2$ nanofiller [72].




(a)




(b)

Figure 9: Comparison of (a) tensile strength and (b) flexural strength of various nontraditional fillers from various literature.

the uniform distribution of filler improves the fiber bonding significantly. However, when the $SiO_2$ concentration rises beyond 15 wt.%, the agglomerates form, resulting in a reduced bonding strength.

Further, the properties of different natural fibers are accomplished and provided in the graphical format in Figures 8(a) and 8(b), respectively. Figure 9(a) depicts the tensile properties of various nonconventional natural fiber composites. It is observed that okra fiber composites exhibited better tensile properties followed by Luffa cylindrica. The least tensile properties are represented by groundnut shell composites due to improper bonding between the

Table 4: List of abbreviations used in the graphs in Figures 8(a) and 8(b).

| | |
|---|---|
| PPTS (60) | Palmyra palm and tamarind seed powder |
| GNS (44) | Groundnut shell |
| LC (50) | Luffa cylindrica |
| ATS (51) | Ashoka tree seed |
| GNSC (62) | Groundnut shells and coir fiber |
| OF (52) | Okra fibers |
| JF (54) | Jackfruit skin powder |
| MS (55) | Melon shell |
| FS (56) | Fish scale |
| PBS (59) | Papaya bast fiber |
| CSTS (64) | Coconut shell powder and tamarind shell powder |
| TS (42) | Tamarind shell |

constituents. The maximum flexural strength was shown by Ashoka tree seed composites compared to other composites which are represented in Figure 9(b). The abbreviation used in the graphs is given in Table 4.

Jagadeesh et al. [73] have reviewed the effect of natural filler materials on fiber-reinforced hybrid polymer composites and have reported that the natural fiber-reinforced polymer composites are the new innovative class of sustainable materials having good mechanical properties for practical applications. Further, to increase the performance of composites, researchers pointed out that using filler materials essentially increases the mechanical properties and in turn minimizes the organic contents in the composite laminates. Hemath et al. [72] work on the effect of TiC nanoparticles on accelerated weathering of coir fiber filler, and basalt fabric-reinforced bio/synthetic epoxy hybrid composites have further reiterated the significance of stronger bonding between the filler content and the fiber that facilitates greater strength characteristics.

## 4. Sustainability of the Nontraditional Natural Fibers

Nontraditional fibers are considered one of the arising accoutrements of the present time, and the "sustainability" term is frequently associated with these nontraditional fiber-reinforced composites [74]. It is due to the biocompatibility and ecosustainability of the nontraditional natural fiber-reinforced composites that the utilization of these composites for real-time applications is increasing [75]. Biodegradability and recyclability can have a significant effect on both future and present-day requirements of ecocompatible developments [76].

Ecofriendly accoutrements are gaining attention throughout the world owing to continually raising environmental concerns and several research on the sustainability indices of renewability, recyclability, commercial viability, biodegradability, and ecocompatibility [77]. In this paper, the review of the sustainability aspects of the traditional natural fiber-reinforced composites is also accomplished and the key factors are identified [78]. The major attributes which make the nontraditional fibers sustainable and very much vital for their use in green composites are depicted in the schematic block diagram given in Figure 10 [79, 80].

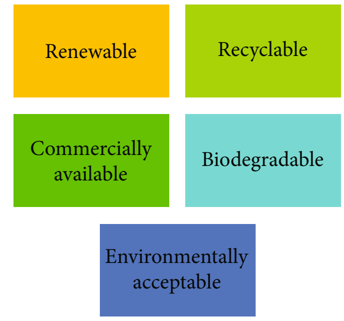



Figure 10: Factors influencing the sustainability of the nontraditional natural fillers for green composites.

The need for sustainable biocomposites for real-time applications has facilitated substantial research to be accomplished in the domain of nontraditional natural filler-based polymer-based biocomposites [81], and this review has considered all the aspects of ecocompatibility and sustainability.

## 5. Conclusion and Future Scope of Work

The review paper has made an attempt to present the summary of various works carried out by researchers on the use of nontraditional natural fibers in green composite materials while keeping different engineering applications in the perspective.

(i) An increase in the usage of these nontraditional natural fibers will reduce carbon footprints and greenhouse gas emissions

(ii) Among all the nontraditional filler-based composites studied in the present work, the okra fiber-filled composites exhibited better tensile properties, and Ashoka tree seed composites show better flexural properties, while fish scale-reinforced polymer composites showed higher impact strength

(iii) The hybrid combination of okra fibers with Ashoka tree seed powder and fish scale-reinforced composites can be used for the load-bearing applications

(iv) The manufactured composites are economical and can be processed at locations where they are available abundantly

(v) The nontraditional natural filler-based composites are environment friendly and can be recycled

(vi) These composites may eventually replace man-made synthetic fiber composites for different engineering applications such as interiors of automobiles and aircrafts, as the tensile, flexural, hardness, wear, impact, and thermomechanical stability of these natural filler-based composites are relatively higher than the composites fabricated from the conventional synthetic sources

The review provides a comprehensive text on the nontraditional fillers for the researchers to look for the alternative filler materials which are suitable for the applications in low load-bearing structures. The researchers shall further explore the possible avenues, for the usage of mango shell powder, tamarind shell powder, tamarind seed powder, tamarind fiber, and walnut shell powder in polymeric composites for various applications.

## Data Availability

The data supporting the review is already included in the manuscript, and any other additional data are readily available from the corresponding authors and will be furnished upon request.

## Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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