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Mechanical Behaviors of Natural Fiber-Reinforced Polymer Hybrid Composites

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1.1 Introduction

The use of composites in industrial applications has evolved tremendously over the years, due to the quest for better material performance and cost reduction. They have been found to have exceptional properties in terms of their physical and mechanical properties. Simply put, composites describe a heterogeneous material that comprises two or more different materials that are combined within a single system such that the new material formed now has improved properties, which are suitable for an intended application. The materials that are combined to form a composite material are known as fiber and matrix, reinforcement and binder as commonly called, respectively. The matrix material could be either a natural or synthetic polymer, while fiber material could be glass, boron, or carbon, among others (synthetic type); hemp, jute, flax, among natural type; organic; or ceramic [1]. The increasing use of composite materials in industries has been traced to the fact that they have light weight, and possess high strength as well as exceptional corrosion resistance and acoustic properties, which make them preferred to metallic and alloy materials. Their applications now span into marine, power/energy, automobile, security, aerospace, telecommunications, sport/game, military industries, among others.

Biocomposite has been defined as a composite with at least one of its components derived from biological or natural sources [1]. Their main features that drive research interest are the fact that they are biodegradable, renewable, cheap, and have natural/sustainable resources. These features underscore their environmental friendliness. Some examples of natural fibers frequently used in biocomposites are caraua, sisal, jute, abaca, and kenaf, among others [2]. Other natural fibers used in biocomposites are hemp, agave, and flax, among others [3]. Some natural fibers have been identified in the literature to be used only for craft production and these include kenaf, agave, coir, ramie, and caraua fibers [3].

Table 1.1 Commonly used natural fibers and their mechanical behaviors.

Fiber	Density (g/cm ³)	Diameter (μm)	Elongation (%)	Tensile strength (MPa)	Young's modulus (GPa)
<i>Bast</i>					
Flax	1.4–1.5	5–38	1.2–3.2	345–1500	27.6–80
Hemp	1.48	10–51	1.6	550–900	70
Jute	1.3–1.46	5–25	1.5–1.8	393–800	10–30
Kenaf	1.2	12–36	2.7–6.9	295	—
Ramie	1.5	18–80	2.0–3.8	220–938	44–128
<i>Leaf</i>					
Abaca	1.5	—	3.0–10	400	12
Banana	1.35	13.16	5.3	355	33.8
Caraua	1.4	—	3.7–4.3	500–1150	11.8
Henequen	1.4	—	3.0–4.7	430–580	—
PALF	1.5	20–80	1–3	170–1627	82
Sisal	1.33–1.5	7–47	2.0–3.0	400–700	9–38
<i>Seed</i>					
Cotton	1.5–1.6	12–35	3.0–10.0	287–597	5.5–12.6
<i>Fruit</i>					
Coir	1.2	—	15.0–30.0	175–220	4–6
Oil palm EFB	0.7–1.55	19.1–25.0	2.5	248	3.2
<i>Wood</i>					
Softwood kraft pulp	1.5	33	4.4	1000	40
<i>Cane/grass</i>					
Bagasse	1.2	10–34	1.1	20–290	19.7–27.1
Bamboo	0.6–1.1	—	—	140–230	11–17

EFB and PALF denote empty-fruit bunches and pineapple leaf fiber, respectively.

Source: Nguyen et al. [4]. © 2017, Elsevier.

Some interesting mechanical behaviors of commonly used natural fibers and many more that are not aforementioned are shown in Table 1.1.

Biocomposites have found application in many different industrial sectors, including packaging, sports articles, and ship building, but most importantly in civil and automotive sectors for nonstructural applications: soundproofing, filling material, and lightening, among others [3]. They favor applications that require low cost and lightness as compared with any other synthetic fiber-reinforced composites. They

also demonstrate good thermal and acoustic insulation capacities [3]. Generally, biocomposites are randomly oriented with short fibers that are obtained through the extrusion or molding manufacturing process [3]. Essentially, the low specific weight as well as low cost of biocomposites is a function of the low weight and low cost of most natural fibers, in combination with the low cost of the automated manufacturing processes when mass-producing them [3].

1.2 Concept of Natural Fibers and/or Biopolymers: Biocomposites

1.2.1 Natural Fiber-Reinforced Polymer Composites or Biocomposites

Natural fiber-reinforced polymer (FRP) composites or biocomposites are gaining widespread interest for many reasons. One such reason is the fact that they have shown a potential for replacement of synthetic fibers at a lower cost. They are also sustainable when compared with their synthetic counterparts [5].

Natural fibers refer to fibers whose origins are natural, that is, they are sourced from plants and animals. These origins give rise to three fundamental natural fiber types, viz:

Animal fibers: These contain proteins, such as keratin, fibroin, and collagen. Other classifications in this category are animal wool/hairs (angora wool, alpaca, camel, mohair, lamb's wool, bison, yak wool, cashmere, horse hair, goat hair, and qiviut, among others), keratin fiber (chicken and bird feathers), and silk fibers (spider silk, tussah silkmths, mulberry silk cocoons).

Plant fibers: These are often referred to as cellulosic or lignocellulosic fibers. They are classified in six categories:

- *Seed/fruit fibers:* Coir, coconut, loofah, cotton, oil palm, kapok, sponge gourd, milkweed hairs.
- *Cane, grass, and reed fibers:* Bamboo, corn, albardine, esparto, bagasse, sabai, papyrus, rape, canary.
- *Bast or stem fibers:* Blax, jute, okra, rattan, paper mulberry, hemp, kenaf, isora, urena, ramie, kudzu, roselle hemp, wisteria, mesta and nettle, among others.
- *Wood fibers:* Hardwood and softwood, among others.
- *Leaf fibers:* Caraua, pineapple, abaca, raphia, agave, caroa, banana, fique, piassava, cantala, sansevieria, phormium, Mauritius hemp, sisal, date palm, istle and henequen, to mention but a few.
- *Stalk fibers:* Derivable from barley stalk, rice stalk, maize stalk, wheat stalk, oat stalk as well as other crops. Table 1.2 shows the percentage weight (wt.%) of chemical compositions of the mostly used natural fibers.

Mineral fibers: These fibers include fibrous brucite, asbestos group (amosite, chrysotile, anthophyllite, crocidolite, actinolite and tremolite) and wollastonite.

Table 1.2 Commonly used natural fibers in hybrid composites and their chemical compositions.

Fiber	Cellulose (wt.%)	Hemicellulose (wt.%)	Lignin (wt.%)	Waxes (wt.%)
Abaca	56–63	20–25	7–9	3
Bagasse	55.2	16.8	25.3	—
Bamboo	26–43	30	21–31	—
Coir	32–43	0.15–0.25	40–45	—
Curaua	73.6	9.9	7.5	—
Flax	71	18.6–20.6	2.2	1.5
Hemp	68	15	10	0.8
Jute	61–71	14–20	12–13	0.5
Kenaf	72	20.3	9	—
Oil palm	65	—	29	—
Pineapple	81	—	12.7	—
Ramie	68.6–76.2	13–16	0.6–0.7	0.3
Rice husk	35–45	19–25	20	14–17
Rice straw	41–57	33	8–19	8–38
Sisal	65	12	9.9	2
Wheat straw	38–45	15–31	12–20	—

Source: Faruk et al. [6]. © 2012, Elsevier.

1.2.2 Polymer Matrices

Polymer matrices serve as bonding agents to fibers. They bond the fibers together and help in load transfer to the fibers. Also, the polymer matrices allow for good-quality finish of composite surfaces as well as protection of the reinforcing fibers from chemical attacks. Two common classifications of polymer matrices are thermosetting and thermoplastic resins. They are subsequently elucidated.

Thermosetting resins: Curing process (chemical reaction) occurs with this type, thus linking polymer chains and connecting the whole matrix in a three-dimensional (3D) network. It should be noted that once curing occurs, re-melting or reforming becomes impossible. These resins are highly stable in dimension, resist high temperature as well as offer good resistance to solvents, due to their cross-linked 3D structure [4]. Some thermosetting resins that are used frequently in composites are vinylesters, polyesters, phenolics, epoxies, bismaleimides (BMIs), and polyamides (PAs).

Thermoplastic resins: These resins differ from thermosetting resins, because their thermoplastic molecules are not cross-linked and can be melted when heated and made into solids and then cooled, thus allowing for reforming and reshaping repeatedly. Apart from being generally ductile, thermoplastic resins have more toughness than their thermosetting counterparts. They are broadly

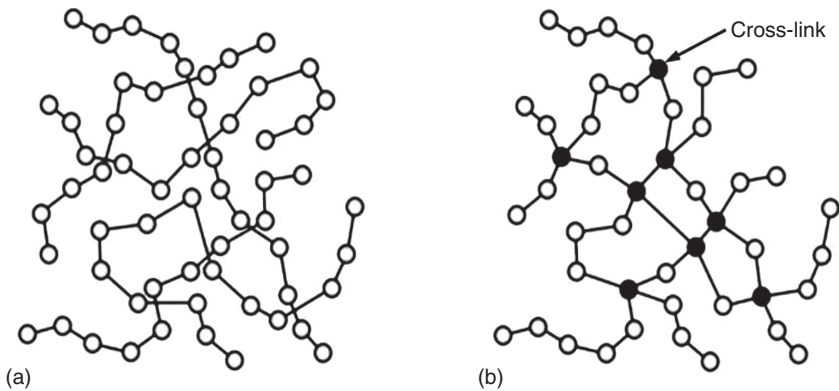


Figure 1.1 Descriptive molecular structure of both (a) thermoplastic and (b) thermoset polymers. Source: Bergstrom [7]. © 2015, Elsevier

used for nonstructural applications without fillers and reinforcements. Their mechanical properties, which are factors of attraction, include good fatigue and compression strength, excellent tensile strength, excellent stiffness, high dimensional stability, excellent damage tolerance, and excellent durability. Furthermore, their flame-retardant as well as wear-resistant features broaden their applications and make them relevant, especially in an aerospace sector [4]. Common examples of thermoplastic resins include, but are not limited to, polyvinylidene fluoride (PVDF), polypropylene (PP), polyetheretherketone (PEEK), polyphenylene sulfide (PPS), polymethyl methacrylate (PMMA, also called acrylic), polyetherketoneketone (PEKK), and polyetherimide (PEI).

Figure 1.1a,b depicts the molecular structure of thermoplastic and thermosetting resins, respectively. The cross-links in the molecular structure of the thermosetting resins (shaded molecules) are depicted in Figure 1.1b.

There are different categories that exist for the manufacturing process of polymer matrix composites (PMCs). These include squeeze flow methods, short-fiber suspension methods, and porous media methods [4]. Table 1.3 depicts some partial and complete natural and synthetic hybrid FRP composites, their resins/matrices, and manufacturing methods.

It is well known that there is no single engineering material that can be all-encompassing in terms of its applicability to operations and processes. Therefore, natural FRP composites have some limitations, despite their outstanding benefits. Table 1.4 presents some of the benefits as well as disadvantages of natural FRP composites.

The key elements that affect the mechanical response of natural FRP hybrid composites are subsequently identified [5]:

- Fiber selection, which includes the type, method of extraction, time of harvest, natural fiber aspect ratio, content, as well as its treatment
- Interfacial strength
- Matrix choice

Table 1.3 Manufacturing processes of some hybrid (mainly natural) FRP composites.

Hybrid fiber	Resin	Curing agent Catalyst	Accelerator	Manufacturing methods
Pineapple/sisal/glass	Polyester	MEKP	Cobalt naphthenate	Hydraulic press
Sisal/silk	Polyester			Hand lay-up technique
Kenaf/glass	Polyester			Hand lay-up and cold press
Woven jute/glass	Polyester			Hand lay-up
Banana/Kenaf	Polyester			Hydraulic compression molding process
Banana/sisal	Polyester			Hand lay-up method followed by compression molding
Glass/palmyra	Polyester			Hydraulic compression molding process
Jute/glass	Polyester			Hand lay-up
Roselle/sisal	Polyester			Hand lay/up technique
Silk/sisal	Polyester			Hand lay-up technique
Banana/sisal	Epoxy			Hydraulic compression molding process
Glass/glass	Epoxy	HY95 I hardener		Hand lay-up technique
Carbon/glass	Epoxy	HY225 Hardener		Hand lay-up technique
Oil palm/jute	Epoxy	Hardener		Compression molding process
Chicken feather/glass	Epoxy	<i>n-tert</i> -Butyl peroxybenzoate		Hot press
Basalt/Hemp	Polypropylene			Hot pressing
Flax, Hemp, and jute	Polypropylene			Hydraulic press
Flax/wood fiber	HDPE			Twin screw extrusion
Banana/glass	Polypropylene			Twin screw extrusion
Cork/coconut	HDPE			Screw extrusion and compression molding
Kenaf/pineapple	HDPE			Mixing and compression molding
Bamboo/glass	Polypropylene			Injection molding
Cordenka/jute	Polypropylene			Injection molding
Bamboo/cellulose	Poly lactic acid			Injection molding
OPEFB/glass	Vinyl ester			Resin transfer molding
Aramid/sisal	Phenolic			Stirring, drying, compression

HDPE, high-density polyethylene; MEKP, methyl ethyl ketone peroxide; and OPEFB, oil palm empty fruit punch.

Source: Sathishkumar et al. [8]. © 2014, SAGE Publications.

Table 1.4 Benefits and drawbacks of natural FRP hybrid composites.

Benefits	Drawbacks
<ul style="list-style-type: none"> ● Renewable source of fibers/matrices and sustainability ● Low danger/risk during manufacturing processes ● Low density, stiffness, and high specific strength ● Low process/production energy and environmental friendliness ● Lower production cost when compared with synthetic fibers, such as carbon and glass ● Low release of harmful fumes when heating and during end of life process (incineration) ● Lower abrasive attack on processing tools, when compared with synthetic FRP composites ● Possibility of predicting better balanced mechanical behaviors, such as toughness 	<ul style="list-style-type: none"> ● Lower responses, especially impact strength in comparison with the synthetic FRP composites ● Higher variability of behaviors, due to discrepancies in sources and qualities ● Lower durability in comparison with synthetic FRP composites. However, it can be enhanced significantly using treatments ● Poor fiber orientation and/or layer stacking sequence, causing weak fiber–matrix interfacial adhesion ● High water/moisture absorption, consequently causes swelling effect ● Lower processing parameters, such as degradability temperatures. Hence, it causes limiting matrix and fiber options and structural applications

Source: Modified from Pickering et al. [5]. © 2014, SAGE Publications.

- Fiber distribution
- Composite manufacturing process
- Fiber arrangement [9]
- Void presence/porosity, among others.

1.3 Hybrid Natural Fiber-Reinforced Polymeric Biocomposites

Fiber hybridization could offer a further alternative in composites. Hybrid composites are derived by a combination of two or more various fiber types in a common matrix [10]. The fibers can be arranged in different layer pattern/orientations and stacking sequences. Figure 1.2 presents some common arrangements (layering patterns) of natural FRP hybrid composites. This introduces wider spread in their properties than in the regular composite materials, which comprises only one kind of reinforcement. It also enables manufacturing engineers to channel the properties of the composite to the required structural properties. This can possibly be achieved once the hybrid composite behavior can be predicted from the constituent composites.

Operationally, producing hybrid natural FRP composites suggests an intermediate intervention to reduce the negative environmental impact of glass and carbon (synthetic) fibers on the environment, by partially replacing the glass and carbon fibers

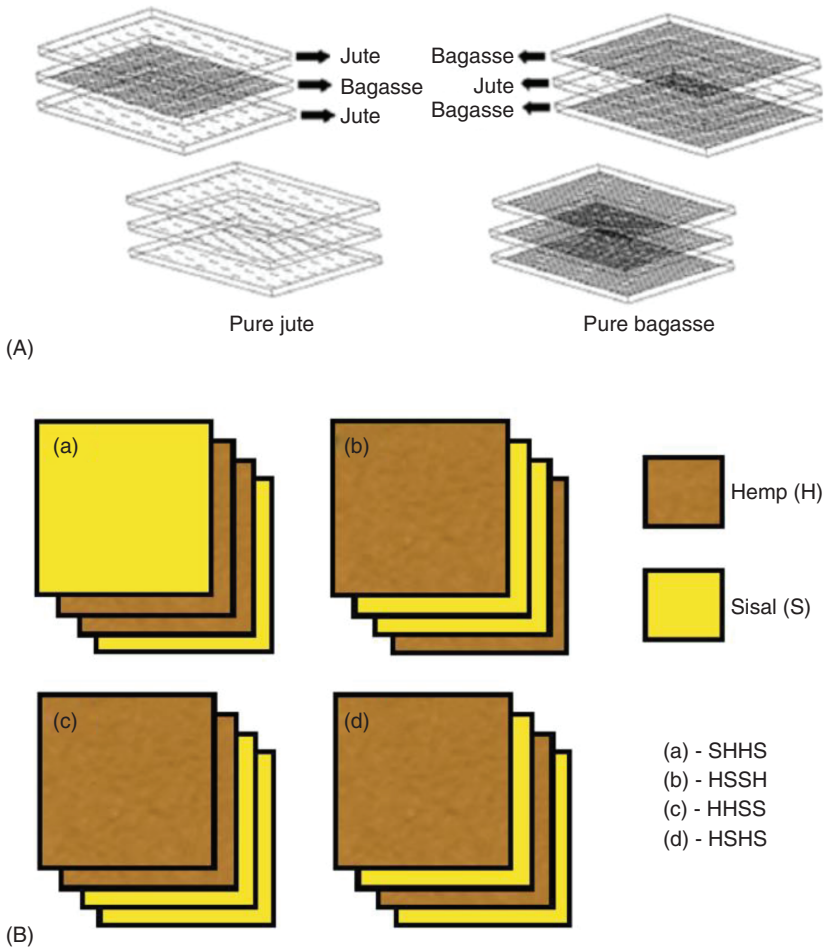


Figure 1.2 Schematic illustration of different orientations and stacking sequences of natural FRP hybrid composites. Source: Refs. [9, 11]. © 2012; John Wiley and Sons.

with such alternatives as the vegetable fibers jute, flax, hemp, kenaf, sisal, among others [12, 13]. In these substitutions, the limits are revealed by simulating the service performance in such a dynamic testing, including fatigue and impact [14, 15].

Moreover, hybrid biocomposites refer to composites in which two or more different biofibers (natural fibers) are combined in a matrix, or a mixture of natural fibers with synthetic fibers in a matrix [4]. One synthetic fiber commonly used for improving the mechanical response in natural FRP composites is glass or carbon fibers. Several types exist for hybrid composites. These types are dependent on the material constituent mixture [16, 17].

For instance, Figure 1.3 shows higher mechanical properties of unaged hybrid flax/basalt FRP composite sample A, when compared with single or non-hybrid flax FRP composite. However, the impact strength property of the aged hybrid counterpart samples B, C, D, and E changed insignificantly after 15, 30, 45, and 60 aging

days of salt-fog environment conditions (Figure 1.3c) of the hybrid types. Additional mechanical behaviors of some FRP hybrid biocomposites are presented later in Table 1.5 by considering their natural/natural fiber combined reinforcements.

In preparing hybrid FRP composites, the rule of mixture comes to play, while the volume fraction can be obtained using Eqs. (1.1)–(1.6) [8].

$$V_{c1} + V_{c2} = 1 \tag{1.1}$$

$$V_{c1} = \frac{V_{f1}}{V_f} \tag{1.2}$$

$$V_{c2} = \frac{V_{f2}}{V_f} \tag{1.3}$$

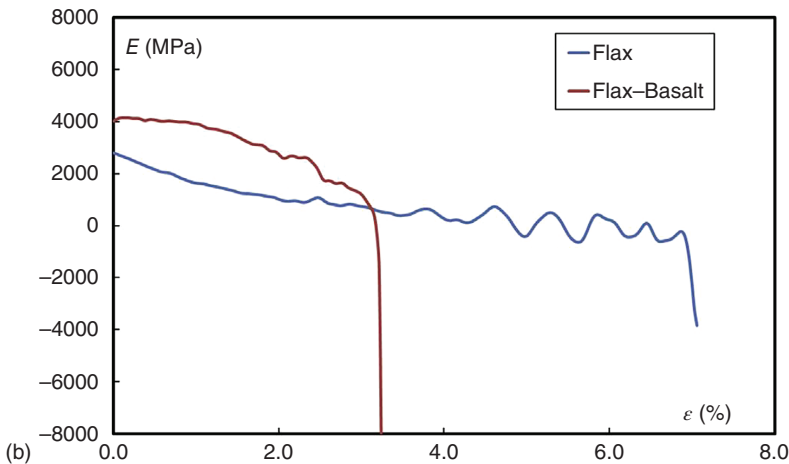
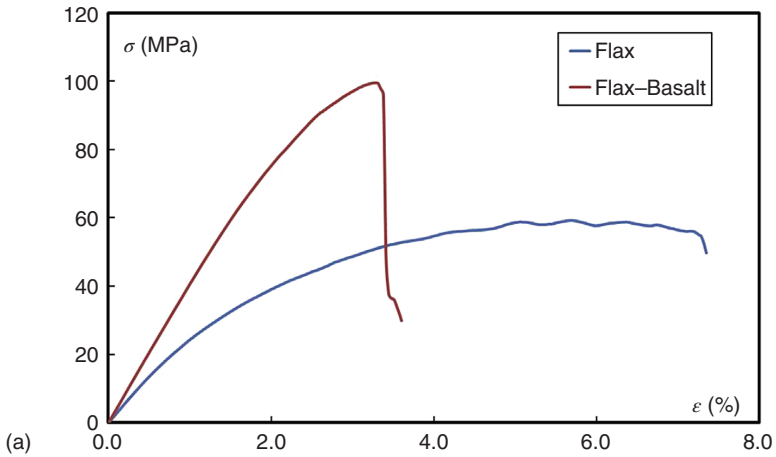


Figure 1.3 Improved mechanical properties of hybrid flax–basalt fibers FRP composites, depicting (a) stress–strain, (b) modulus–strain curves, and (c) impact strengths of aged and unaged biocomposites. Source: Fiore et al. [18]. © 2016, Elsevier.

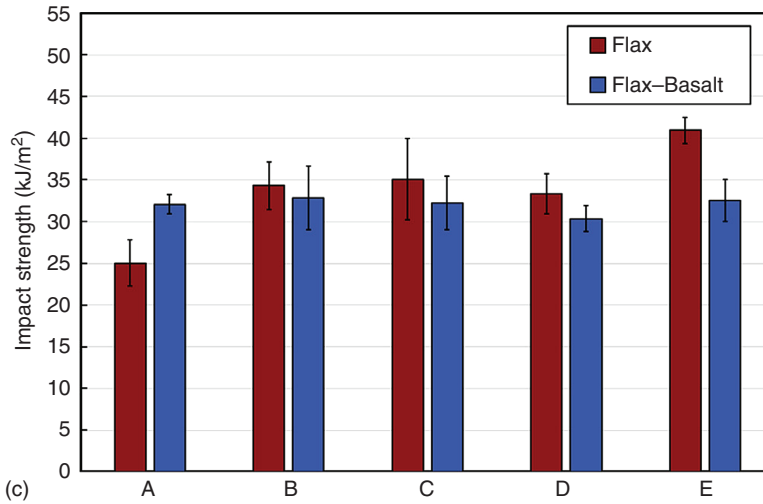


Figure 1.3 (Continued)

$$V_f = V_{f1} + V_{f2} \quad (1.4)$$

$$W_f = W_{f1} + W_{f2} \quad (1.5)$$

$$V_{f1} = \rho_c \frac{W_{f1}}{\rho_{f1}} \quad (1.6)$$

where V_f denotes total reinforcement volume fraction, V_{c1} and V_{c2} represent the first and second reinforcement relative volume fractions, V_{f1} and V_{f2} stand for the first and second fiber volume fractions, ρ_c and ρ_f designate the densities of the composites and fiber, while W_f indicates the weight of the fiber. The methodology for preparing and characterizing hybrid fiber-reinforced PMCs as well as its applications is presented in Figure 1.4.

However, the present chapter does not cover all the methodologies shown in Figure 1.4 in detail, because the scope of this chapter is not manufacturing processes and techniques of natural FRP composite materials.

1.4 Mechanical Behaviors of Natural Fiber-Reinforced Polymer-Based Hybrid Composites

There are many properties of materials that determine where they function or are used in the engineering space. The required characteristics in a proposed design will determine what combinations of materials will be relevant and which of the various mechanical properties are of interest in such instances. Notable among the mechanical properties usually considered in engineering are tensile, compressive, flexural, and impact strengths, among others. These properties are discussed in Section 1.4.1.

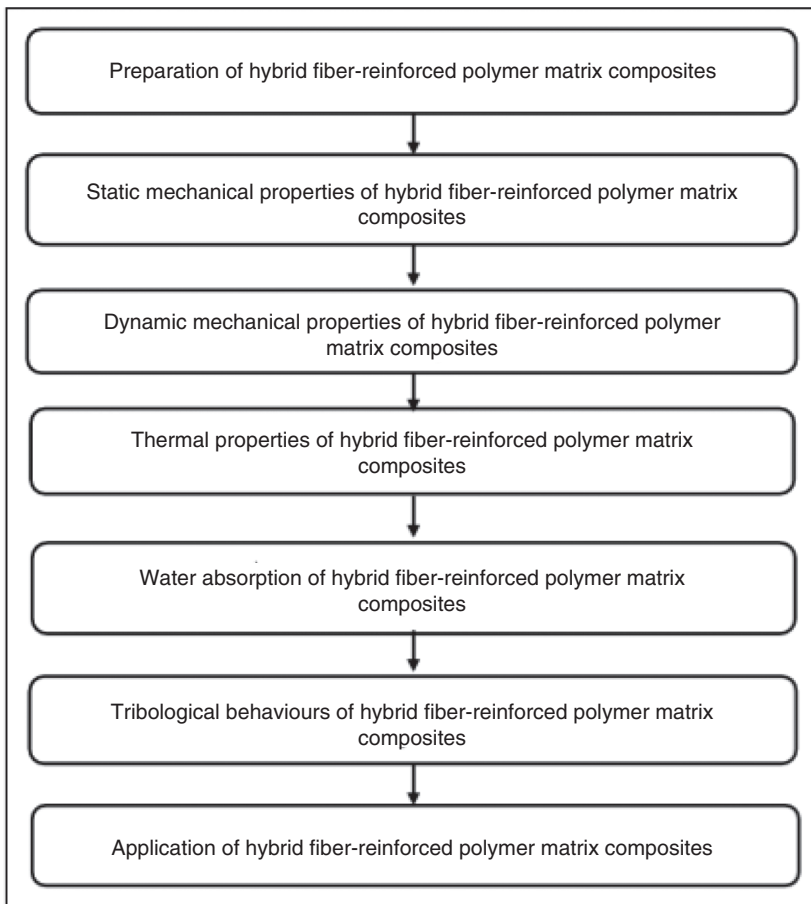


Figure 1.4 Flowchart of preparation and characterization of the hybrid FRP composites. Source: Sathishkumar et al. [8]. © 2014, SAGE Publications.

1.4.1 Hybrid Natural FRP Composites

This section discusses hybrid biocomposites in which their combined fibers are entirely natural (biofibers).

1.4.1.1 Bagasse/Jute FRP Hybrid Composites

Jute is a popular plant-based fiber (vegetable) with dominant presence in tropical countries across the Asian continent, such as China, Brazil, Nepal, Bangladesh, India, and Thailand. They account for about 95% of jute fiber (JF) production worldwide [4]. Jute is considered as a lignocellulosic bast fiber, having comparative advantages with respect to renewability, biodegradability (which makes it eco-friendly), high strength as well as high initial modulus over other fibers [11]. Bagasse, also called sugarcane bagasse, is a lignocellulosic by-product of the sugar industry, mostly utilized as a fuel in boilers and sugar factories. Compared

with other residues (by-products), including wheat straw and rice, bagasse is preferred, because its ash content is lower [19].

A study of mechanical behavior of hybrid FRP composites with short JF and short bagasse fiber (BF) bundles reinforcement was carried out by Saw and Datta [20]. They used epoxidized phenolic novolac (EPN) as resin matrix and investigated various fiber surface treatments and fiber ratios. Sodium hydroxide (NaOH) alkali solution was used to treat the JF bundles. The BF bundles were either modified using chlorine dioxide (ClO_2) and furfuryl alcohol ($\text{C}_5\text{H}_6\text{O}_2$) or left untreated. The modification of the fiber surface was necessary for quinones creation in the lignin areas of the BF bundles. The created quinones then reacted with the furfuryl alcohol, and thereby improved the BF bundles' (modified) ability for better adhesion. Their result revealed greater mechanical responses (flexural, tensile and impact properties) for hybridized BF (modified) and JF bundles (alkali-treated) in the EPN resin matrix than the BF bundles that were not modified. They obtained an optimum mechanical behavior at a BF/JF ratio of 50 : 50, as depicted in Table 1.5.

1.4.1.2 Bamboo/MFC FRP Hybrid Composites

Asian giants, India and China, are the chief producers of bamboo fiber with more than 80% of global production [21]. This biofiber is highly attractive, due to its renewable nature and low environmental impact. It grows rapidly and has comparative high strength to other biofibers, such as cotton and jute [22].

An unprecedented biocomposite (hybrid) that contained biodegradable poly-lactic acid (PLA) matrix with microfibrillated cellulose (MFC) and bamboo fiber bundles reinforcements was developed by Okubo et al. [23]. Various nomenclatures have been used for describing MFC in the literature, such as microfibril, microfibrillar cellulose, microfibril aggregates, nanofibril, nanofibrillar cellulose, nanofiber, and fibril aggregates [24]. They conducted an investigation on how MFC dispersion influenced the responses of composites reinforced with bamboo fibers by dispersing MFC in a polymer matrix of PLA by a three-roll mill calendering process. This calendering process helps to compress or smoothen a material. They used the PLA (bio-based and biodegradable) polymer matrix for interfacial bonding enhancement with the MFC. The diameter of bamboo fiber bundles was about 200 μm , while that of MFC was just a few microns, which was much smaller. Using gap settings in decreasing order of 70, 50, 35, 25, 15, 10, and 5 μm , they processed the mixture of the MFC and PLA in the three-roll mill. About 200% increase in the fracture energy was realized when they added 1 wt.% of MFC to the PLA matrix and milled the MFC/PLA composite at the smallest gap setting of 5 μm , which was quite significant. This hybrid composite combination of bamboo fiber and the PLA matrix with 1 wt.% MFC reinforcement was observed to prevent an abrupt crack channel through the bamboo fiber effectively, and thus produced a significant improvement in fracture strength. The results of other mechanical behaviors are presented in Table 1.5.

1.4.1.3 Banana/Kenaf and Banana/Sisal FRP Hybrid Composites

A good material for reinforcement in diverse polymer composites is the banana fiber. Its extraction is usually from the bark of banana trees [4]. Banana fiber has such

Table 1.5 Mechanical behaviors of bagasse/jute, bamboo/MFC, and banana/kenaf FRP hybrid composites.

Hybrid biocomposites	Fiber ratio (by weight or volume)	Flexural modulus (GPa)	Flexural strength (MPa)	Tensile modulus (GPa)	Tensile strength (MPa)	Impact strength (kJ/m ²)
<i>Natural fibers</i>						
Bagasse fiber bundles (untreated) and jute fiber bundles (treated)						
	0 : 100	0.645	31.15	0.302	11.45	6.90
	20 : 80	0.789	36.46	0.356	16.02	7.46
	35 : 65	1.101	45.32	0.420	19.45	9.53
	50 : 50	1.480	55.63	0.492	23.07	10.66
Bagasse/jute	65 : 35	1.311	51.19	0.399	21.15	8.33
	100 : 0	0.502	26.78	0.227	9.87	6.67
Bagasse fiber bundles (treated) and jute fiber bundles (treated)						
	20 : 80	1.178	42.72	0.526	18.72	10.00
	35 : 65	1.484	54.57	0.635	22.57	13.33
	50 : 50	1.748	65.22	0.753	26.77	15.93
	65 : 35	1.518	60.12	0.704	23.54	10.93
	100 : 0	0.632	30.78	0.286	11.20	8.66
MFC/PLA composites (milled to 5 μm)						
Bamboo/MFC	1 wt.% of MFC	—	—	4.61 ± 0.27	45.9 ± 4.1	—
	2 wt.% of MFC	—	—	3.95 ± 0.14	51.7 ± 2.3	—
	50 : 50, nonwoven hybrid					
Banana/kenaf	10% NaOH treatment	—	57.2	—	44	13
	10% SLS treatment		60.8		50	16
	50 : 50, woven hybrid					
	10% NaOH treatment		62.0	—	50	18
	10% SLS treatment	—	68.0		54	21

MFC and PLA represent micro-fibrillated cellulose and poly-lactic acid, respectively.

Source: Nguyen et al. [4]. © 2017, Elsevier.

advantaged mechanical properties, including good tensile strength and modulus, due to the high content of cellulose and low microfibrillar angle [25]. Kenaf fiber is a promising element of reinforcement in polymer composites, due to its interesting mechanical features such as eco-friendliness and renewability. Kenaf is usually extracted from bast fiber (kenaf plants) [4]. Sisal, on the other hand, is known to be among the toughest materials for reinforcement. It is also well known for its durability. Sisal FRP composites possess moderate flexural and tensile behaviors and high impact strength, when compared with other composites of natural fiber reinforcements. It has relevant use in some industries, such as agriculture and marine to make twines, ropes, cords, rugs, and bagging, among others [26]. Sisal and kenaf fibers, similar to other natural fibers, have poor interfacial bonding with a polymer matrix, which shows their disadvantage [27].

Moreover, Thiruchitrambalam et al. [28] conducted a study on woven as well as non-woven hybrids of banana/kenaf fiber with unsaturated polyester matrix reinforcement. They kept the fiber contents constant at 40% with equal ratio of banana and kenaf FRP composites (50 : 50 ratio) and treated the fibers with either 10% solution of NaOH or 10% of sodium lauryl sulfate (SLS) for 30 minutes. They observed that the SLS-treated specimen had better improvement with respect to mechanical behavior than the alkali-treated specimen, showing for both woven and non-woven cases of the banana/kenaf hybrid composites enhanced impact, flexural, and tensile strengths, as shown in Table 1.5.

Furthermore, Venkateshwaran et al. [29] evaluated the mechanical properties of banana/sisal FRP epoxy matrix hybrid composite and found out that hybridization increased the flexural, tensile, and impact strengths by 4%, 16%, and 35% respectively. They also reported that the 50 : 50 fiber ratio by weight enhanced the mechanical response of the banana/sisal FRP hybrid composite and decreased the uptake of moisture, as presented in Table 1.6.

1.4.1.4 Coconut/Cork FRP Hybrid Composites

A natural coconut fiber, also known as coir, is usually extracted from coconut trees. These trees are mainly grown in tropical regions of Asian countries, such as Vietnam, India, and Thailand [4]. Cork fiber is usually obtained from cork oak trees (*Quercus suber*). There is a specific species of the tree from whose bark the cork fiber is harvested. The cork oak tree is a renewable resource, as new cork bark regrows naturally [4].

Hybrid composites containing high-density polyethylene (HDPE) with reinforcements of cork powder and short coconut fibers that were randomly distributed was prepared by Fernandes et al. [30]. The interfacial bonding and compatibility between the matrix and fiber was improved by maleic anhydride, a coupling agent (CA). Their results showed 27% and 47% rise in elastic moduli and tensile strengths of the coconut/HDPE/cork hybrid composites, respectively (Table 1.6), when compared with the cork/HDPE composite. Also, they observed that using CA resulted in enhancement of the elongation at break and tensile behaviors of the hybrid composites. As a recommendation for better mechanical responses of

Table 1.6 Mechanical behaviors of banana/sisal, coconut/cork, coir/silk, corn husk/kenaf and cotton/jute FRP hybrid composites.

Hybrid biocomposites	Fiber ratio (by weight or volume)	Flexural modulus (GPa)	Flexural strength (MPa)	Tensile modulus (GPa)	Tensile strength (MPa)	Impact strength (kJ/m ²)
Banana/sisal	100 : 0	8.920	57.33	0.642	16.12	13.25
	75 : 25	9.025	58.51	0.662	17.39	15.57
	50 : 50	9.130	59.69	0.682	18.66	17.90
	25 : 75	9.235	60.87	0.703	19.93	20.22
	0 : 100	9.340	62.04	0.723	21.20	22.54
Coconut/cork	10 : 44 : 44 : 2 (wt.% of coconut/ cork/HDPE/ coupling agent) Alkali treatment	—	—	0.599 ± 0.02	20.4 ± 0.3	—
Coir/silk	10 mm fiber	—	39.53	—	15.01	—
	20 mm fiber	—	45.07	—	17.24	—
	30 mm fiber	—	42.02	—	16.14	—
Corn husk/kenaf	0 : 30 (PLA 70 wt.%)	—	—	2.117	—	—
	15 : 15 (PLA 70 wt.%)	—	—	1.547	—	—
	30 : 0 (PLA 70 wt.%)	—	—	1.221	—	—
Cotton/jute	23.7 : 76.3 (jute fabric type III)	—	—	—	—	—
	Test angle, 0°	9.9 ± 0.8	136.7 ± 4.0	7.1 ± 0.3	59.4 ± 1.7	9.3 ± 0.9
	Test angle, 45°	8.4 ± 0.7	84.6 ± 4.7	4.6 ± 0.1	21.1 ± 1.4	7.5 ± 1.0
	Test angle, 90°	7.2 ± 0.7	58.3 ± 5.4	4.1 ± 0.1	14.6 ± 0.5	5.5 ± 1.0

HDPE = high-density polyethylene.

Source: Nguyen et al. [4]. © 2017, Elsevier.

the cork-based composites, 10 wt.% of short coconut fibers and 2 wt.% of CA were proposed.

1.4.1.5 Coir/Silk FRP Hybrid Composites

Silk is a continuous protein fiber characterized by its soft, light, and thin nature and produced by different insects. The silkworm and spun synthesize silk fiber, such as silk cocoon. Large quantities of silk proteins (sericin and fibroin) are produced by the silkworm at the last stage of larval development [31]. These silk proteins are key components of the silk cocoons. Silk fiber, with its huge specific strength and stiffness, have wonderful luster and excellent drape. It prides itself as the strongest material in nature. It however has poor resistance when exposed to sunlight [4].

Using unsaturated polyester matrix, Noorunnisa Khanam et al. [32] in an investigation into the coir/silk fiber hybrid composites used various fiber lengths of 10, 20, and 30 mm. Sodium hydroxide (NaOH) solution was used to treat the coir fibers in order to eliminate their lignin and hemicellulose, thereby causing better bonding of the fiber with the matrix. Composites of 20 mm fiber length showed higher tensile and flexural strengths than the 10 and 30 mm counterparts, as shown in Table 1.6. The tensile, flexural, and compressive strengths were improved significantly in the coir/silk hybrid composites, owing to the NaOH treatment that facilitated the bonding at the coir fiber–polyester matrix interface.

1.4.1.6 Corn Husk/Kenaf FRP Hybrid Composites

Several agricultural wastes such as rice straw, corn husk, and rice husk form a huge quantity of raw natural fibers that are used in polymer composites as materials for reinforcement. Corn husks contain fibers that are rich in cellulose. They are the thin and leafy sheaths that surround corn cobs [33]. Kenaf fiber is important in paper as well as other industrial sectors, as a fiber source.

Kwon et al. [34] used PLA matrix and prepared kenaf fiber and corn husk flour hybrid biocomposites, using a constant fiber to matrix ratio of 30 : 70 by weight (Table 1.6). Different kenaf/corn husk flour ratios were examined. Before and after extrusion, the aspect ratio was measured for kenaf fibers and its influence on the mechanical behavior was investigated. The result showed that the aspect ratio post-extrusion had no influence on the values predicted from the Halpin–Tsai equation. Note that the Halpin–Tsai model for predicting elastic response of composites assumes that there is no fiber–matrix interaction and works on the basis of the orientation (geometry) as well as elastic behavior of the matrix and the fibers. They found out that the variation in the Young’s modulus of fibers affected the transfer of stress from the matrix to the fiber and reported that a factor of control to optimize mechanical behavior in hybrid biocomposites could be the reinforcements’ scale ratio in various aspect ratios.

1.4.1.7 Cotton/Jute and Cotton/Kapok FRP Hybrid Composites

Cotton fibers have been considered as fibers with the greatest importance across the globe. They usually do not have branches and the seed hairs have a single cell (unicellular). They are also rich in cellulose and can elongate up to 30 mm. The wall of cotton fiber does not contain lignin, as distinct from the secondary cell walls of most plants [35]. Cotton fibers are used widely in the textile industry. They possess some advantages, which include excellent drape, high absorbency, as well as good strength.

In the study conducted by De Medeiros et al. [36], the mechanical behavior of woven fabrics of hybrid cotton/jute with phenolic matrix (novolac type) reinforcement was investigated. The results showed strong dependence on the mechanical behaviors on fiber content, fabric characteristics, fiber–matrix adhesion, and fiber orientation. The anisotropy of the composites increased when the test angle was increased and showed dependence on fiber roving/fabric characteristics. There was an inverse proportionality between the mechanical properties and the test

Table 1.7 Mechanical behaviors of cotton/kapok, cotton/ramie, and jute/oil palm empty-fruit bunches (OPEFB) FRP hybrid composites.

Hybrid biocomposites	Fiber ratio (by weight or volume)	Flexural modulus (GPa)	Flexural strength (MPa)	Tensile modulus (GPa)	Tensile strength (MPa)	Impact strength (kJ/m ²)
Cotton/kapok	3 : 2					
	Untreated ($V_f = 60\%$)	—	—	0.884	55.70	110.53
	Alkali treatment ($V_f = 43\%$)	—	—	1.635	52.87	119.25
	Non-accelerated weather condition ($V_f = 46.6\%$)	0.709	52.40	—	—	—
	Accelerated weather condition ($V_f = 46.6\%$)	0.703	39.55	—	—	—
Cotton/ramie (ramie fibers placed longitudinally to the mold length)	10.8 : 41.1 (0° composite)	—	—	—	90.9 ± 12.7	—
	11.9 : 45.5 (0° composite)	—	—	—	117.3 ± 13.3	—
	11.9 : 45.1 (0° composite)	—	—	—	118.0 ± 6.5	—
	1 : 4					
Jute/OPEFB	OPEFB/Jute/OPEFB	—	—	2.39	25.53	—
	Jute/OPEFB/Jute	—	—	2.59	27.41	—
	Pure OPEFB	—	—	2.23	22.61	—
	Pure jute	—	—	3.89	45.55	—

Source: Nguyen et al. [4]. © 2017, Elsevier.

angle as the best performance was obtained in the specimen tested at zero degree (that is, jute roving direction). Brittle failure, though controlled, was displayed in the composites that were tested at angles of 45° and 90° as jute fiber directions (Table 1.6), while at 0° to the longitudinal direction, the tested composites exhibited an uncontrollable catastrophic failure. Jute fiber was identified as a strong material for reinforcement and when combined with cotton could prevent catastrophic failure in the fabric composites. In addition, the results obtained from mechanical behaviors of cotton/kapok FRP hybrid composites are depicted in Table 1.7, showing the effects of accelerated and non-accelerated weather conditions of alkaline-treated and untreated specimens. The results of cotton/ramie FRP hybrid composites, with ramie fibers placed longitudinally to the mold length and various ratio, are also shown in Table 1.7.

1.4.1.8 Jute/OPEFB FRP Hybrid Composites

Oil palm (*Elaeis guineensis*) is a perennial crop known for its high-value fruits from which oil is produced. It mostly grows in tropical regions, such as Southeast Asia and West/Southwest Africa. Oil is extracted by stripping the fruits (nuts) from the bunches, a process that leaves the empty-fruit bunches (EFBs) as waste material [37]. Fibers of oil palm are usually derived from the oil palm empty-fruit bunches (OPEFB) as well as mesocarp. In composite materials, the OPEFB fibers are mostly used, as they contain the highest hemicellulose content in comparison with pineapple, coir, banana, as well as soft and hardwood fibers [11].

A three-ply hybrid sample of jute/OPEFB fibers composites with epoxy resin reinforcement was prepared by Jawaid et al. [38], fixing the jute/OPEFB ratio (by weight) at 1 : 4. They investigated the void content, chemical resistance, as well as tensile behaviors of the hybrid composites. From the results obtained, the OPEFB/jute/OPEFB and jute/OPEFB/jute composites showed great resistance to chemicals: toluene (C_7H_8), benzene (C_6H_6), water (H_2O), 40% of nitric acid (HNO_3), carbon tetrachloride (CCl_4), hydrochloric acid (HCl), 5% of acetic acid (CH_3COOH), 20% sodium carbonate (Na_2CO_3), 10% of sodium hydroxide (NaOH), and 10% of ammonium hydroxide (NH_4OH). A lower void content was displayed in the jute/OPEFB/jute than pure OPEFB as well as OPEFB/jute/OPEFB composites, because the mats of the jute fiber adhered better to the epoxy resin with higher compatibility. At the outer ply, the jute fibers withstood the tensional stress due to their high strengths, and the core (OPEFB fiber) absorbed and distributed the stresses evenly within the composite sample systems. Also, it was evident from Table 1.7 that the jute/OPEFB hybrid exhibited higher tensile responses (both strength and modulus) as well as improved adhesion bond between the fiber and the matrix, when compared with pure OPEFB composite.

1.4.1.9 Kenaf/PALF FRP Hybrid Composites

A tropical plant, pineapple (*Ananas comosus*) belongs to the family of bromeliad (*Bromeliaceae*). In South America, it is next in line to banana and mango in total production across the globe [39]. Pineapple leaf fibers (PALFs) are waste products when cultivating pineapples and are extracted from pineapple leaves. It has a significant mechanical behavior, because it is high in cellulose (70–82%) as well as in crystallinity (44–60%) [40]. Combining these properties with that of Kenaf fiber, excellent tensile and flexural strengths from FRP composite are obtained, which promises a good material for different applications [4].

Aji et al. [41] studied hybridized Kenaf/PALF specimens with HDPE reinforcement, using 1 : 1 fiber ratio. They investigated into how the size of fiber and its loadings affected the mechanical responses of the hybrid biocomposites (Table 1.8). The four reinforcement lengths considered at a fiber loading range of 10–70% were 0.25, 0.50, 0.75, and 2.00 mm. The smallest of these fiber lengths (0.25 mm) yielded the best result in terms of its flexural and tensile properties, while both 0.75 and 2 mm exhibited enhancement in impact strength. As observed further, an increase in the fiber length reduced some of the mechanical behaviors, which is credited to the entanglement in fibers as against fiber attrition. An inverse proportionality was

Table 1.8 Mechanical behaviors of kenaf/PALF, roselle/sisal, and silk/sisal FRP hybrid composites.

Hybrid biocomposites	Fibre ratio (by weight or volume)	Flexural modulus (GPa)	Flexural strength (MPa)	Tensile modulus (GPa)	Tensile strength (MPa)	Impact strength (kJ/m ²)
Kenaf/PALF	1 : 1 (At 0.25 mm fiber length and 60% fiber loading)	4.114	34.01	0.874	32.24	6.167
	1 : 1					
Sisal/roselle	Dry condition, fiber length = 15 cm	—	76.5	—	58.7	1.30
	Wet condition, fiber length = 15 cm	—	62.9	—	44.9	1.28
Sisal/silk	1 : 1, fiber length = 20 mm					
	Untreated	—	46.18	—	18.95	—
	Alkali treatment	—	54.74	—	23.61	—

Source: Nguyen et al. [4]. © 2017, Elsevier.

established between the tensile and impact properties, as the rule of mixture was satisfied by flexural strength. The adhesion between the fiber and the matrix interface was good, as evaluated by scanning electron microscopy (SEM).

1.4.1.10 Sisal/Roselle and Sisal/Silk FRP Hybrid Composites

Sisal (*Agave sisalana*), from the *Agavaceae* family, is a hard-fiber plant with wide cultivation in the tropical countries of Africa, America, and Asia, though it has its origin in Mexico and Central America. Their fibers are strong and tough, and extracted from sisal plant leaves. Sisal fibers are widely utilized in composites and plastic/paper industries. The nativity of roselle (*Hibiscus sabdariffa*) can be traced to West Africa. It is a species of Hibiscus, whose plant is naturally abundant and majorly used for fruits and bast fibers. Roselle fibers have extensive applications in the textile industry and in composites, because they exhibit greater mechanical behaviors in comparison with some other naturally occurring fibers, such as jute and kenaf.

Moreover, Athijayamani et al. [42] used a fiber ratio of 1 : 1 for the sisal and roselle fibers and investigated, under wet conditions, how absorption of moisture affected the mechanical responses of short hybrid sisal/roselle FRP unsaturated polyester composites. Using different fiber contents and lengths, their results showed an improvement (increase) in the flexural and tensile strengths of the hybrid composites of sisal/roselle fibers at increased lengths and contents of fibers and under dry condition. For the wet condition, the strengths (tensile and flexural) were significantly reduced, while inverse proportionality between the impact strength and fiber length and content was observed for both conditions (wet and dry), as depicted in Table 1.8. In addition, Noorunnisa Khanam et al. [43] also

carried out a study on sisal/silk fiber ratio of 1 : 1 and prepared a polyester-based hybrid composite to evaluate various fiber lengths. Their results showed higher mechanical (tensile, flexural, and compressive) responses from the composite sample with fiber length of 20 mm than that of 10 and 30 mm counterpart hybrid composites (Table 1.8). Also, the results obtained after fiber modification depicted that the same mechanical behaviors of the alkali-treated hybrid fiber composites improved significantly.

1.5 Other Related Properties that Are Dependent on Mechanical Properties

There are other properties of biocomposites that are relevant for material analysis. Some of these are dynamic mechanical properties, thermal and water absorption behaviors, as well as tribological properties. Only tribological and thermal behaviors are subsequently discussed.

1.5.1 Tribological Behavior

Tribological behavior refers to friction-related properties of the materials. The frictional coefficient of hybrid sisal/glass fiber (GF)-reinforced epoxy composites was measured by Ashok Kumar et al. [44], using different sliding speeds of 0.2, 2.0, and 4.0 mm/s, under a constant load of 10 N. At an atmospheric temperature of 22 °C and relative humidity of 45%, both alkali-treated samples of fiber composites and untreated ones were tested. The graph of frictional coefficient against fiber length (Figure 1.5) revealed that the frictional coefficient was lower, up to 2 cm fiber length. However, the frictional coefficient increased with an increasing composite fiber length. Moreover, fiber length addition led to a decrease in the frictional

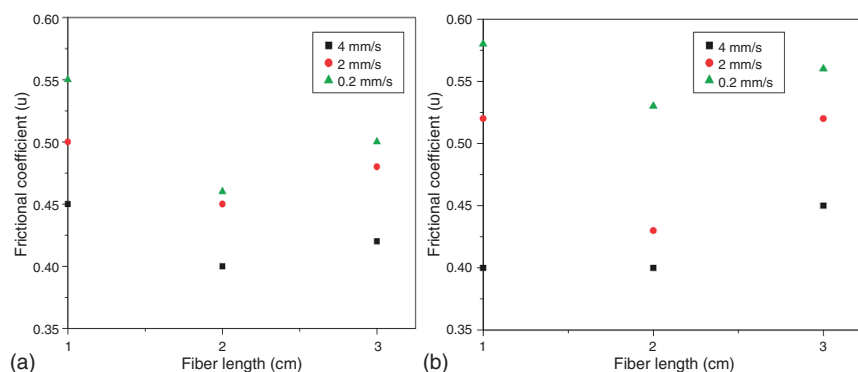


Figure 1.5 Frictional coefficients of (a) treated and (b) untreated sisal/glass FRP hybrid composites as a function of fiber length, after 50 cycles. Source: Ashok Kumar et al. [44]. © 2010, SAGE Publications.

coefficient when sliding speeds were higher. The treated fiber of the reinforced composites yielded an optimum improvement at 2 cm in comparison with the untreated samples.

Biswas and Xess [45] studied the behavior of short bamboo/E-GF-reinforced epoxy hybrid composites with respect to erosion wear, using different compositions by weight as thus: 65 wt.% of epoxy, 22.5 wt.% of bamboo fiber, 22.5 wt.% of GF; 70 wt.% of epoxy, 15 wt.% of bamboo fiber, 15 wt.% of GF; and 75 wt.% of epoxy, 7.5 wt.% of bamboo fiber, 7.5 wt.% of GF, as well as 100 wt.% epoxy. The graph of the result of erosion rate against impact velocity showed that the 15 wt.% bamboo/GF FRP composites possessed the lowest rate of erosion in comparison with the other composites.

1.5.2 Thermal Behavior

The thermal property is concerned with the response of hybrid FRP biocomposites to heat variation. Boopalan et al. [46] worked on hybrid raw jute/banana fiber-reinforced epoxy composites with regard to their thermal analysis by varying the fiber weight ratio. They used ratios of 100/0, 75/25, 50/50, 25/75, and 0/100, while varying the temperature with the use of thermogravimetric analysis (TGA) and heat deflection temperature (HDT) analysis. With the TGA, the curve depicted that the 50/50 jute/banana FRP epoxy hybrid composite demonstrated greater thermal stability. There was a shift in the temperature during degradation from a value of 200 °C to a higher value of 380 °C. For the HDT, thermal property was sustained in the 50% jute with 50% banana FRP epoxy hybrid composite at the highest temperature of 90 °C in comparison with other composite samples.

Also, thermal properties of OPEFB/woven jute FRP epoxy hybrid composites were investigated by Jawaid et al. [47], using various temperatures. It was reported in their work that there was an increase in thermal stability when woven jute fibers were added to the EFB composite in its pure state. That is, hybridizing OPEFB with the woven jute fiber caused the thermal stability to be higher as compared with OPEFB fiber. The temperature of degradation was also reported to have shifted from a value of 292 °C to a higher value of 457 °C, leaving 12.1% char residue.

1.6 Progress and Future Outlooks of Mechanical Behaviors of Natural FRP Hybrid Composites

Application of various hybrid natural FRP composites is increasing with their innovative designs and developments through optimized manufacturing techniques. The advent of automation and robots in manufacturing of hybrid natural FRP composite materials has been improving their properties. This will increase in the next century, with synergy of sophisticated processes and techniques. Mechanical properties are most important responses that require serious attention during the design and fabrication of new hybrid natural FRP composites. Other properties of hybrid natural

FRP composites, such as thermal, acoustic, electrical, water absorption, among others, are directly dependent on the mechanical behaviors.

The prospect for FRP composite materials is very high and bright. It is presumed that in the next decade, application of various hybrid natural FRP composite materials would have penetrated all facets of life. This will be made possible through enhanced mechanical behaviors of a new set of composites as fiber selection, extraction, treatment, interfacial adhesion with matrix, and processing of natural FRP composite are improved [6].

In addition, there have been significant developments in natural FRP hybrid composites in the past few decades, due to established advantages in terms of processing, low cost, biodegradability, renewability, high specific strength, sustainability, as well as low relative density. Various natural fiber types have been and are being studied, with the findings forming the basis for replacing synthetic fibers, including both carbon and glass. Primarily, the idea of biocomposites development centers around the generation of novel FRP composites that are environmentally friendly with respect to how they are produced, used, and discarded. Hence, natural FRP composites could be a valid replacement and even superior alternative to synthetic fiber composites. Their biodegradable nature offers a good solution to the problem of waste disposal often experienced with synthetic fiber, petroleum, or non-renewable polymer-based materials. The application of biocomposites is widening continually and is projected to expand more, with more effects in Europe, due to mounting legislative and public pressures.

Till now, adhesion between the natural reinforcements/fiber and matrix interface, as hybridization increases, remains a major object of concern in terms of overall performance of natural FRP hybrid composites. This is a major factor in the ultimate properties, especially mechanical responses of the biocomposites. Further cutting-edge research is therefore necessary in order to overcome this challenge. Also, there is a requirement for more research work in order to get over other challenges such as inadequate toughness, moisture absorption, and stability reduction in long-term outdoor applications. Particularly, various weather conditions, humidity, temperature, and ultraviolet radiation, have significant influence on the product service life of natural FRP hybrid composites. For example, ultraviolet exposure results in discoloring, property deterioration, and deformation.

c Lastly, identifying better extraction of raw materials, sustaining crop growth, product design, and manufacture are activities that will help to reach the goal of better natural FRP hybrid composites. Hence, further research is ongoing across the globe to overcome the aforementioned challenges of biocomposites. Their respective properties should form the basis for generating new applications and create more opportunities for these biocomposites in the present-day green environment and secured future.

1.7 Conclusions

Owing to the sustainability, environmental friendliness, low cost of production, biodegradability, as well as enhanced mechanical behaviors of natural FRP

composites through hybridization techniques, natural FRP composites are competing with and replacing some synthetic or conventional FRP composites, including both glass and carbon FRP composites. From the studies reported in this chapter, it is evident that mechanical behaviors of some natural FRP hybrid composites, especially impact, tensile, and flexural strengths and moduli, were higher than those of their non-hybrid or single FRP counterparts. Also, the degree of mechanical responses of the natural FRP hybrid composites depends on various areas of their engineering applications, as structural, semi, and/or non-structural composite systems.

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