Chapter 2
Introduction to Manufacturing of Natural Fibre-Reinforced Polymer Composites

M. Arifur Rahman, Fahmida Parvin, Mahbub Hasan, and M. Enamul Hoque

Abstract In the recent era, different environmental issues have significantly influenced the innovations in material science and technology. The burgeoning demand for clean environment has led the innovation of green materials and utilization of natural materials. Thus, the urge for the production of high-performance engineering products from natural renewable resource is growing day by day. Composites are among those versatile, high-performance materials which combine the unique mechanical and thermal properties that cannot be achieved in a single material. In the recent decade, scientists continued to explore the potential of natural fibres as the reinforcing phase for polymer composites. The important driving force for such emergence of utilizing natural resources is that they are renewable and biodegradable and impose no adverse effects on environment, whereas petroleum-based products are limited and cause environmental problems. This review gives the state-of-the-art overview on currently developed natural fibre-reinforced polymer composites focusing on structure–property relationship of fibres, different polymer matrices used to develop composites, their mechanical performances, different composite fabrication techniques, and the application of such composites in different areas. Critical issues of biocomposites have also been discussed along with their advantages and disadvantages. This article also summarized the critical issues in the manufacturing of natural fibre composites.

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2.1 Introduction

In the recent decades, scientific and technological interests have been shifting toward the development of stronger materials based on the fibre-reinforced composites. Such idea or innovation of composites is, in fact, an inspiration from biological materials such as human bone where soft tissues are reinforced with fibrous protein and exhibits unique mechanical performance. Many artificial polymer-based composites, reinforced by carbon, glass, or aramid fibres, have been developed in automotive, aerospace, construction, or sporting industries. Among many man-made fibres, glass fibre is the mostly used one for reinforcing plastics because of their comparative low-cost and high mechanical performance. In spite of being highly stable during processing, glass fibres have high density, consume high energy and cost for production, are not biodegradable, and also possess health hazards (Wambua et al. 2003). In addition, environmental concern related to the waste disposal of such nonbiodegradable composite has been growing rapidly. Since the 1990s, natural fibre-reinforced composites are evolving as convincing replacements to glass-reinforced composites in numerous uses. Natural fibres are abundant in nature; they can be grown, have cost-effective processing, and also possess unique physical and mechanical properties. Thus, they can be considered as renewable source of fibres. Plenty of different natural fibres have been used as the reinforcing phase of composites. For example, in automotive industries, hemp fibre epoxy, flax fibre polypropylene (PP), and china reed fibre PP are intensively used due to their low bulk density and lower production cost. Moreover, being thermoplastic in nature, PP- and PE-based composites can be easily recycled, and thus they have drawn a lot of attention in different industries (Mohanty et al. 2000). In addition to the cost-effective processability, NFRCs also offer a number of environmental benefits such as less pollution, less dependence on nonrenewable resources, improved energy recovery, and biodegradability.

Fabrication of natural fibre-reinforced composites has always been a challenge due to their low stability during processing. Generally, traditional manufacturing techniques, which are designed for conventional fibre-reinforced polymer composites and thermoplastics, such as compression molding, injection molding, or vacuum infusion, are still being used for the fabrication of NFRCs. These techniques are still considered to be efficient for the production of high-quality composites. However, the success in the production of NFRCs by utilizing these techniques requires proper understating of the structure—properties of natural fibres which have low thermal stability and low mechanical resistance to shear and compatibility with conventional thermoplastics (Ho et al. 2012a). Thus, pretreatment has always been an important issue for better processability of NFRCs. In many cases, chemical treatments of fibre surface are necessary to improve the interfacial interaction between fibre and matrix polymer. Thus, the correlation among the selection of materials and processing methods to the final properties of the composites is very important.

The versatile applications of NFRCs are widely known. Recent study estimates that in 2010, total global natural fibre composite materials market shipments topped 430.7 million pounds with a value of US$289.3 million, and the market is expected to grow to US$531.3 million in 2016 with an 11 % compound annual growth rate (CAGR) over the next 5 years (Faruk et al. 2014). Moreover, automotive industries are consuming most of the NFRCs and expected to be the largest market for NFRCs till 2016. Automotive industries have already adopted the technology of NFRCs due to their low weight, lower production cost, and recyclability. Thus, various products, not restricted only for interior but also nonstructural applications, have been marketed by the automotive industry. On the other hand, concerning the environmental issues related to the disposal of plastic waste, a remarkable trend of utilizing natural fibre-reinforced bioplastics can be noticed (Shen et al. 2009).

Another interesting fact is that the number of publications as patents, books, or peer-reviewed papers is increasing rapidly which indicates the growing importance of NFRCs. Figure 2.1 shows that the number of patents and publications increased rapidly after year 2000 (Figure 1, Satyanarayana et al. 2009). This chapter introduces different NFRCs and the state-of-the-art review of their manufacturing techniques as well as their potential applications.

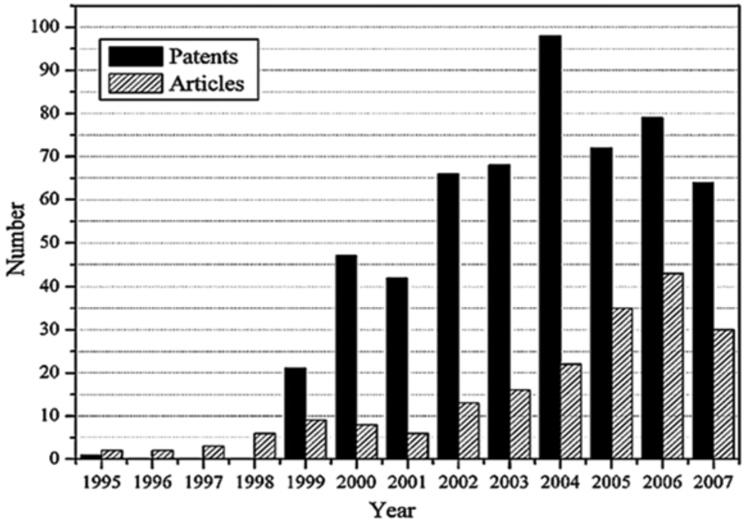

Fig. 2.1 This graph is showing the number of publications produced on natural fibre-reinforced composites each year (1995–2007)

2.2 Structures and Properties of Natural Fibres

2.2.1 Types and Sources of Natural Fibres

Natural environment comprising both flora and fauna is an enormous resource for fibres. However, in this chapter we will only focus on the plant-based fibres. Plant fibres have become a new generation of reinforcing material (Faruk et al. 2014). These fibres can be grouped into different categories based on their source and derivation of plant, animal, and mineral types (Ho et al. 2012b).

Plant-based fibres can be classified as seed hair, bast fibres, and leaf fibres, depending upon the source. Some examples are cotton (seed hairs); hemp, ramie, jute, kenaf, and flax (bast fibres); and sisal, banana, coir, and abaca (leaf fibres). Figure 2.2 shows the photographs of plant-based fibres. It is reported that Brazil and Tanzania produce the largest amount of sisal fibre, whereas Bangladesh, China, and India are the largest producers of jute fibre.

2.2.2 Structure of Natural Fibres

Plant fibres have complicated composite cell structures. Fibres are composed of cellulose microfibrils that reinforce the lignin and hemicellulose matrix. Figure 2.3 shows that the cellulose microfibrils are helically wounded along the hollow fibre axis.

Fig. 2.2 Images of plant-based fibres

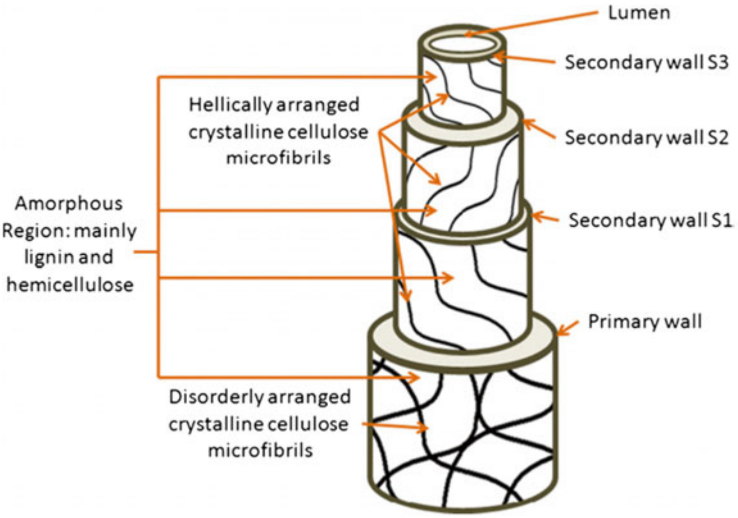

Fig. 2.3 Structural constitution and arrangement of a natural plant fibre cell. (Adopted from John and Thomas 2008)

Such helical conformation gives strength to the fibre because it requires high energy to uncoil the oriented fibrils (Dicker et al. 2014). On the other hand, hemicellulose plays key role for the biodegradation, moisture absorption, and thermal degradation of the fibre. In addition, lignin is responsible for the UV degradation. The percentage composition of each of these components varies for different fibres. Generally, the fibres contain 60–80 % cellulose, 5–20 % lignin, and up to 20 % moisture (Saheb and Jog 1999).

2.2.3 Properties of Natural Fibres

Mechanical properties of natural fibres are much lower when compared to those of the most widely used competing reinforcing glass and other man-made fibres (Table 2.1). However, because of their low density, the specific properties (property-to-density ratio), strength, and stiffness of natural fibres are comparable to the values of man-made fibres (Wambua et al. 2003). In case of natural fibre, mechanical properties are determined mainly by the cellulose content and microfibrillar angle. Young's modulus of natural fibre decreases with the increase of diameter. A high cellulose content and low microfibril angle are desirable properties of a fibre to be used as reinforcement in polymer composites (Williams and Wool 2000). The mechanical properties of natural fibre are also significantly related to the degree of polymerization of cellulose in the fibre.

Table 2.1 Mechanical properties of natural and man-made fibres (Taj et al. 2007; Saheb and Jog 1999)

Fibres	Density (g/m^3)	Elongation (%)	Tensile strength (MPa)	Young modulus
Natural fibre				
Jute	1.3	1.5–1.8	393–773	55
Sisal	1.5	2–2.5	510–635	9.4–28
Flax	1.5	2.7–3.2	344	27
Kenaf		1.5	389–930	35–53
Pineapple		2.4	170	62
Cotton	1.5–1.6	7–8	287–597	5.5–12.6
Hemp		1.6	690	
Coir	1.2	30	175	4–6
Ramie		3.6–3.8	400–938	61.4–128
Wool		25–35	120–174	2.3–3.4
Spider silk		17–18	875–972	11–13
Man-made fibre				
E-glass	2.5	2.5	2,000–3,500	70
Aramid	1.4	3.3–3.7	3,000–3,150	63–67
Carbon	1.4	1.4–1.8	4,000	230–240

2.3 Different Polymers as the Reinforcing Phase of NRFCs

2.3.1 Polyolefins

The use of natural fibres in thermoplastics such as polyethylene and polypropylene (PP) has, however, been limited, because of difficulties associated with surface interactions. This is due to the inherent hydrophilic nature of fibres and hydrophobic nature of polyolefin matrices which in consequent gives difficulty in processing. In addition, traditional chemical treatment such as the use of coupling agents is not very effective in improving the interfacial interaction between fibre–polyolefin matrixes. However, the use of functionalized polyolefins, such as MA-*g*-PP or MA-*g*-PE, as compatibilizers has resulted in substantial improvements in the properties of olefin composites (Sanadi et al. 1994). Several research works reported the use of different types of natural fibres (chemically modified or not) as the reinforcing phase for polyolefin-based composites. Sanadi et al. (1994) showed that kenaf fibre-reinforced PP can exhibit significant improvement in tensile and flexible properties over the virgin PP and $CaCO_3$-filled PP. Khan et al. (2002, 2009a, b, 2010a, b, c) explored different ways of modifying jute fibre surfaces in order to enhance PP matrix and fibre interaction that ultimately led to improved mechanical performance. They (2002; 2010a, b, c) also investigated the effect of gamma and UV radiation on the thermal stability and mechanical properties of polyolefin composites. Recently, some works have been reported on the use of oil palm, banana, and coir fibres as the reinforcing agents for PP, LLDPE, LDPE, and HDPE matrices. Ren et al. (2014) used twin screw extruder to produce bamboo pulp fibre-reinforced

HDPE composites and observed improvement in mechanical and thermal properties with 30 % fibre content. Another study (AlMaadeed et al. 2014) showed that the wood content significantly affect the mechanical properties of palm wood/LDPE composites. Increasing wood filler also contributed to the increase in water absorption capacity of the composite. Similar study was done by Darie et al. and they observed pseudoplasticity of LDPE with increasing oak wood content (Darie et al. 2011).

In a recent work, Kakou et al. (2014) developed oil palm fibre-reinforced HDPE composite with 40 % fibre. They used maleated PP as the compatibilizer and thus improved the thermal stability and mechanical properties. Apart from the production of polyolefin composites having natural fibres as the reinforcing phase, the biodegradability is always an important issue with such composites (Joseph et al. 2002; Abu-Sharkh and Hamid 2004; Arutchelvi et al. 2008).

2.3.2 Glassy Polymers

Glassy polymers are widely used for their optical clarity, high heat distortion temperature, and high-impact strength. However, such polymers require high temperature for processing which can degrade natural fibre like jute or kenaf. Thus, a limited number of works have been done on natural fibre-reinforced thermoplastic glassy polymers.

2.3.2.1 Polycarbonate

Khan et al. (2005) reported the use of jute fibre treated with HEMA (2-hydroxyethyl methacrylate) to reinforce polycarbonate (PC) matrix. They found that HEMA acted as a coupling agent between fibre and matrix and observed improved adhesion that ultimately improve the storage modulus of the composite significantly. Khan et al. (2006) also showed that similar improvement in interfacial adhesion and mechanical behavior can be achieved by treating the jute fibre with a silane monomer (γ-aminopropyltrimethoxysilane). Recent research also showed that fibre modification is necessary in order to achieve good adhesion between fibre and PC matrix. Threepopnatkul et al. (2009) showed that alkali-treated pineapple leaf fibre modified with silane coupling agents can improve the adhesion between PC matrix and fibre phase and thus improved the thermal stability as well as tensile and impact strength of the composite.

2.3.2.2 Polymethyl Methacrylate

Polymethyl methacrylate (PMMA) is another widely used thermoplastic glassy polymer which is well known for its excellent optical clarity. However, it shows very low fatigue resistance, unsatisfactory transverse strength, and low impact strength. Hence, there is a need for the improvement of its fracture resistance. Very few works have been reported on the use of natural fibres to reinforce the PMMA matrix. John et al. (2014) explored the feasibility of using oil palm fibre as the reinforcement phase for PMMA matrix. They improved the flexural strength and impact strength of PMMA composites. Another work by Sain et al. (2014a) investigated the effect of jute fibre modification on the interfacial compatibility between PMMA matrix and fibre and observed significant improvement in thermal stability and mechanical strength of the composites. Recently, Sain et al. (2014b) reported that by modifying the cellulose nanofibre by maleic anhydride and methyl methacrylate, good biodegradability can be achieved.

2.3.2.3 Polystyrene

Polystyrenes have limited use in engineering applications due to the trade-off between toughness and stiffness. For example, atactic polystyrene is widely regarded as being relatively stiff but suffers from poor toughness. A good number of research works have been done to improve the toughness and impact strength by adding inorganic particles, rubbers, impact modifier particles, and so on. However, also in this case, the use of natural fibre is very limited. Nair et al. (1996) showed that benzoylated short sisal fibre can be used to reinforce the polystyrene (PS) matrix and improve the impact strength. However, the toughness of the composite could not be improved significantly. In a recent work, Oumer and Bachtiar (2014) showed that sugar palm fibre can be used to improve the impact strength and stiffness. However, the toughness was not improved.

2.3.3 Polyamides

Polyamides are important class of engineering thermoplastics and well known for their creep resistance, stiffness, and toughness. Polyamide composites have been used in automotive industries intensively because of their relatively lightweight, durability, corrosion resistant, and attractive appearance (Johnson et al. 2004). A good number of literature can be found on the natural fibre-reinforced nylon composites. This is because nylons possess higher mechanical behavior as the matrix, and they are very compatible with lignocellulosic fibres (Graff 2005). Different natural fibres such as sisal, palm, kenaf, and flax have been used to produce nylon composites. Thitithanasarn et al. (2012) reported the development of nylon 6 composites with epoxy treated jute and observed improved interfacial compatibility that led to improved mechanical performance. Attempts have also been made to optimize the processing condition for nylon 6 composites reinforced with blend of different natural fibres (Ozen et al. 2013). Some research works have also reported the development of bio-based polyamides reinforced with fibres (Feldmann and Bledzki 2014). The advantages of such composite are their biodegradability and recyclability.

2.3.4 Bioplastics

Reinforced bioplastics are relatively new class of composites that have attracted a lot of attention in the recent era due to the increasing environmental awareness concerning the waste management of plastics. Although bioplastics are environment-friendly, they still have poor thermal and mechanical stability. However, recent studies showed that biocomposites made of bioplastics and biofibres can compete with the conventional engineering plastics. Among different bioplastics, PLLA, PCL, PVA, PBS, and PHAs and some waste proteins like gelatin are widely used. Table 2.2 shows a list of different biodegradable polymers.

Bledzki and Jaszkiewicz (2010) performed a comparative study on the mechanical properties of PLA, PHB, and PP reinforced with cellulose, abaca, and jute fibres. Figure 2.4 shows that bio-based polymer composites revealed higher tensile strength and modulus compared to petrochemical-based composites. They (Bledzki and Jaszkiewicz 2010) also observed that biofibre-reinforced PLA composites showed improved properties than reinforced PP composites.

Kim et al. (2011) compared the odor emission from PLA and PBS composites with PP- and PE-based composites. They found that PLA and PBS composites emit less odor than PE- and PP-based composites. This indicates that PLA and PBS composites are suitable for interior application and can replace PP- or PE-based composites. It is important to note that many lignocellulosic fibres and man-made fibres have been used to produce PLA composites by using traditional manufacturing technologies, and excellent thermal and mechanical biodegradability was observed as reviewed by Ganster and Fink (2010).

2.4 Critical Issues in the Processing of Natural Fibre-Reinforced Composites

There are several important issues in processing natural fibre-reinforced composites. The issues are mostly related to the structures and properties of the fibres. Thus, the properties of the composites depend on the proper selection of matrix polymer, pretreatment of fibres, and thermal stability of fibre during the processing. Such issues are discussed below.

2.4.1 Thermal Stability of Natural Fibres

The thermal degradation of natural fibres is an important concern for the processing of NFRCs. It is well known that all lignocellulosic fibres undergo chemical and physical changes when they are heated at a temperature range of 100–250 °C (Gassan and Bledzki 2001). Some of the physical changes involved the alterations

Table 2.2 Different types of biopolymers, their manufacturers, and trade names (Adopted from Mohanty et al. 2000)

Material class	Manufacturer	Product name
Cellulose acetate	Mazzucchelli	BIOCETA®
	Planet polymer	EnviroPlastic®-Z
Copolyester	BASF	Ecoflex
	Eastman	Easter Bio™
Polycaprolactone (PCL)	Birmingham polymers	Poly(caprolactone)
	Planet polymer	EnviroPlastic®-C
	Solvay	CAPA®
	Union carbide	TONE®
Poly(ester amide)	Bayer	BAK 1095
		BAK 2195
Polyethylene terephthalate (PET) modified	DuPont	Biomax®
Polyglycolide (PGA)	Alkermes	Medisorb®
	Birmingham polymers	Poly(glycolide)
	Boehringer ingelheim	Resomer®
	PURAC	PURASORB® PG
Polyhydroxyalkanoates (PHA)	Metabolix	PHA
	Biomer	Biomer™
	Monsanto	Biopol®
Poly(lactic acid) (PLA)	Alkermers	Medisorb®
	Birmingham polymers	Poly(L-lactide) and poly(D,L-lactide)
	Boehringer ingelheim	Resomer®
	PURAC	PURASORB® PL/PD/PDL
Poly(vinyl alcohol) (PVOH)	Idroplast	Hydrolene®
	Novon Planet polymer	Aqua-NOVON® Aquadro™
	Texas polymer	Vinex™
Starch and starch blends	AVEBE	Paragon™
	BioPlastic (Michigan)	Envar™
	BIOTEC	Bioplast®, Bioflex®, Biopur®
	Earth Shell	Starch-based composite
	Novamont	Mater-Bi™
	Novon	Poly-NOVON®
	Starch Tech	ST1, ST2, ST3
Other blends	Alkermers	Medisorb®
	BioPlastic (Colorado)	Biocomposite material
	Birmingham polymers	Poly(D,L-lactide-*co*-*caprolactone*)
		Poly(D,L-lactide-*co*-glycolide)

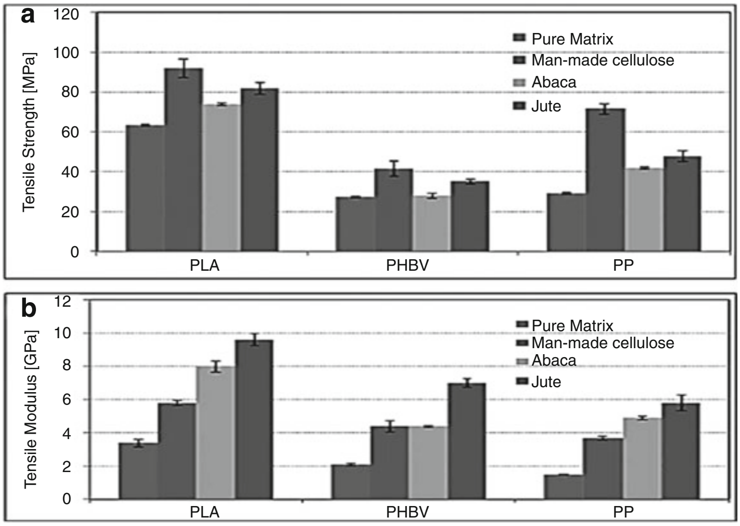

Fig. 2.4 Tensile strength (**a**) and tensile modulus (**b**) of PLA, PHBV, and PP composites with man-made cellulose, abaca, and jute fibres

in physical or chemical structures such as depolymerization, hydrolysis, oxidation, dehydration, decarboxylation, and recrystallization. For an example, Zeronian (1977) reported that the depolymerization takes place in cotton fibre when they are heated from 165 to 240 °C. The degree in the depolymerization decreased from 5,360 to 320 and also recrystallization took place. Similar observation was reported by Rusznák and Zimmer (1971). However, heating (at 160 °C) a composite reinforced with cotton fibre can cause rapid deterioration of strength (Brushwood 1988). Burger et al. (1995) observed similar loss in strength and tenacity of flax fibre. There are other reports that also showed the detrimental effect of processing temperature on the mechanical properties of natural fibre-reinforced composites. Thus, it is important to improve the thermal stability of natural fibre that will also improve the mechanical performances of the composites.

A good number of chemical and physical treatments of natural fibres, as a crucial preprocessing step of composite fabrication, have been reported in the literature. Grafting of functional monomer on the fibre surface is a popular way to improve the thermal stability of natural fibres. For example, Mohanty et al. (1989) reported the improvement in thermal stability of jute fibre by grafting acrylonitrile on the fibre surface. Khan et al. (2002, 2009a, b, 2010a, b, c) reported quite a good number of research works on the fibre surface modification by photoinduced grafting of functional monomers on the fibre surface. They also reported several work on the gamma radiation-treated jute fibres. They stated that the thermal stability of jute fibre can be improved by gamma irradiation (at a certain dose). Recent study by George et al. (2014) showed an inexpensive and environment-friendly method to improve the thermal stability of natural fibres. They improved the thermal stability of flex and hemp fibres by treating them with different enzymes such as hemicellulases, pectinases, and oxidoreductase. Treatment with hemicellulose and pectinase improved the thermal properties for both fibres. Kalia et al. (2013) wrote a nice review on the surface modification of plant fibres by environment-friendly methods. They stated that surface modification of plant fibres using chemical treatments becomes less attractive because of a number of limitations. Environment-friendly methods such as plasma treatment, treatments using fungi, enzymes, and bacteria, can be used for the surface modification of plant fibres on order to improve the thermal stability as well as the other physico-mechanical properties.

2.4.2 Dispersion of Natural Fibres in the Polymer Matrix

It is well known that the dispersion of natural fibres determines the mechanical performances of the composites. Most often bad dispersion of fibres can lead to the deterioration of the mechanical properties of the composites. Although the dispersion is a fibre size-dependent phenomenon, it is not easy to achieve good dispersion of natural fibres in the matrix. Different physical and chemical treatments of fibres can be carried out to improve the fibre dispersion in the matrix. Most often pretreatment of fibres is done in order to improve the dispersion in the matrix. Different coupling agents, such as maleic anhydride-grafted PE, mineral oil, or stearic acid, have been used to improve the dispersion. Stearic acid improves the dispersion by reducing the fibre to fibre interaction, whereas mineral oil acts as a lubricant that induce disentanglement of fibres and thus improves the dispersion.

2.4.3 Hydrophilicity of Natural Fibres

Most of the lignocellulosic fibres are hydrophilic due to the presence of hydroxyl groups, and thus they are vulnerable to microbial degradation. In addition, these fibres are incompatible to most of the hydrophobic polymer matrices. The incompatibility of fibres to the matrix contributed to poor mechanical performances of the composites. Moreover, poor wetting fibres cause difficulty in mixing. Thus, chemical and physical treatments are necessary to reduce the moisture absorption by fibres (John and Anandjiwala 2008). One of the most conventional ways of reducing moisture absorption by fibres is the hydrothermal treatment of natural fibres. Due to the increase in crystallinity of cellulose and separating portion of hemicellulose by hydrothermal treatment, water absorption is reduced (Pott et al. 1999, 2000). Photocuring of monomers on fibre surface is a popular chemical treatment to reduce the water absorption (Khan et al. 2001, 2005, 2008, 2010a, b, c).

2.4.4 Interfacial Adhesion Between Natural Fibre and Polymer Matrix

Most of the polymers are hydrophobic in nature, and thus good adhesion between fibre and polymers is the most important issue in achieving desired properties of the matrix (Tang and Kardos 1997). In addition, interface in composites plays key role in transferring the stress through the fibres to matrix. There are several ways to improve the interfacial adhesion between fibres and matrix (Herrera-Franco and Drzal 1992; Qaiss et al. 2014). For example, fibre surface can be treated with different functionalized monomers that act as coupling agents to matrix, or functionalized polymer matrix can also be used (Petinakis et al. 2014). In addition, radiation treatment of fibres such as gamma irradiation or plasma treatment of fibre surface can be useful to functionalize the surface of polymer as well as the fibres. In addition to the surface treatment of fibres, the use of a compatibilizer or coupling agents can also improve the effective stress transfer across the interface (Tang and Kardos 1997; Wang 2006). The compatibilizer can be polymers with functional groups grafted onto the chain of the polymer such as tetrafunctional organometallic compounds based on silicon, titanium, and zirconium and are commonly known as silane, zirconate, or titanate coupling agents (Tang and Kardos 1997).

2.5 Different Processing Techniques to Fabricate NFRCs

Appropriate manufacturing process is necessary in order to alter materials into the right shape without producing any imperfection into the product. There are several important criteria to find out the right processing technique. The size, shape, and desired properties of the composites are the three most important criteria for the production of composites. In addition, speed of the production and manufacturing cost are fairly more important criteria in the industries. The preliminary assessment to choose appropriate processing techniques depends on the size and shape of the composites. In addition, the properties of raw materials also determine further modification to the processing technique. Several manufacturing techniques have been discussed below.

2.5.1 Fabrication of NFRCs by Injection Molding

A great deal of research works have been reported on the fabrication of NFRCs by injection molding (Huda et al. 2005a, b, 2006a, b; Birgitha 2007). Usually polymer pellets containing chopped fibres are fed through the hopper into feed block with rotating screws. Then the viscous flow of the polymer melt carrying the fibres is forced into the mold cavities. The viscous melt then solidifies at certain pressure and temperature.

The orientation and distribution of the fibres are determined in this stage. After sufficient cooling, the composite is removed with a desired shape.

The traditional injection molding process has several limitations. For example, the fibre length is usually shorter in the final product than it is expected due to the solidification of the melt in the mold cavities. There are several other issues that need to be addressed properly in order to obtain good composites, and the issues are residual stress development during the molding that may cause cracking and long-term deformation of the composites.

Process, material, and geometric parameters should be optimized to minimize these problems to happen. Process parameters include the melt temperature, injection and screw speeds, injection pressure, and the mold temperature that can be controlled on the injection units. Increasing mold temperature results in a decreasing overall stress level, while the compressive stress region is shifted onto the surface (White 1985). According to recent research and investigations, for biocomposites, the machine temperature of biodegradable polymer, such as poly-lactic-acid (PLA) composites, was made by using injection molding process that should be restricted in the range of 150–210 °C depending on the type of PLAs and their crystallinity from diverse manufacturers.

2.5.2 Compression Molding

Fabrication of NFRCs by compression molding can be done in two different ways: hot press or autoclave process. In an autoclave process, the compounded materials are kept inside the pressure chamber under vacuum at a certain temperature, certainly above the softening point of the polymer matrix to allow sufficient flow of the fibres and matrix. After several heat–pressure cycles, a composite is formed (Mallick 1993). On the other hand, for the hot-press processing, a close mold may or may not be necessary. It is important to make sure that fibres do not break under a certain load in the hot press. Sheet molding compounds (SMCs) and bulk molding compounds (BMCs) are traditional initial charges for compression molding process.

2.5.3 Resin Transfer Molding

Liquid composite molding processes encompass resin transfer molding (RTM), vacuum-assisted resin transfer molding (VARTM), structural reaction injection molding (S-RIM), co-injection resin transfer molding (CIRTM), and other subsets where the basic approach is to separately inject the liquid resin into a bed of stationary preforms. The RTM process has become a popular composite manufacturing process due to its capability for high-volume production and cost-effectiveness. Many studies have been made on the potential of natural fibres as reinforcement with renewable polymers as matrix through RTM (Sreekumar et al. 2007; Williams and Wool 2000; Ferland et al. 1996; Kim and Daniel 2003; Ikegawa et al. 1996;

Warrior et al. 2003). In the RTM process, dry fibre preform (impregnating) or porous fibrous preform is placed into the mold cavity. Two matching mold halves are clamped tightly to avoid leakage of resin during injection process. Then, using dispensing equipment, a pressurized molten plastic is injected into the heated mold using single or multiple inlet ports in the mold depending on the complexity of the shape of a final product until the mold is filled with resin. After cooling, the part is then removed from the mold. Post-curing normally is needed to ensure the resin is fully cured (chemically reacted between the resin and its catalyst).

2.6 Structure and Properties of Various NFRCs

Natural fibre-reinforced polymer composites are gaining paramount importance day by day in the composite research field and industry due to their versatile diversified nature and renewability. Structure and properties of lignocellulosic natural fibre-reinforced polymer composites are mainly affected by the type of matrix, the content and properties of the reinforcing fibre, and the fibre–matrix interaction. Some of those properties and their related structures are described below.

2.6.1 Structural Properties

Scanning electron microscope (SEM) is one of the most important and frequently used tools for structural analysis of natural fibre-reinforced polymer composites. Figure 2.5 shows SEM micrographs of jute, coir, abaca, bagasse, bamboo, and palm fibre-reinforced polypropylene (PP) composites. The SEM image of jute fibre-reinforced PP composites (Fig. 2.5a) shows agglomeration of untreated jute in the PP matrix. This feature suggests weak interfacial bonding between the fibre and matrix. Fibre agglomeration is also seen in the SEM images of coir, abaca, and palm-reinforced PP composites (Fig. 2.5b, c, f) indicating weak interfacial adhesion between the fibre and matrix. SEM micrograph of bagasse-reinforced PP composites (Fig. 2.5d) shows fibre pullout due to the lack of interfacial adhesion between raw bagasse fibre and PP matrix. On the other hand, bamboo fibre-reinforced PP composite had broken fibre in their microstructure (Fig. 2.5e). One of the ways of improving the interfacial adhesion between the natural fibre and polymer matrix in composites is chemical treatment of the fibre prior or after composite manufacturing (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Nahar 2014).

2.6.2 Mechanical Properties

Mechanical properties of natural fibre-reinforced polymer composites are commonly characterized using tensile, flexural, impact, and hardness tests. Those properties of various natural fibre polymer composites are described below.

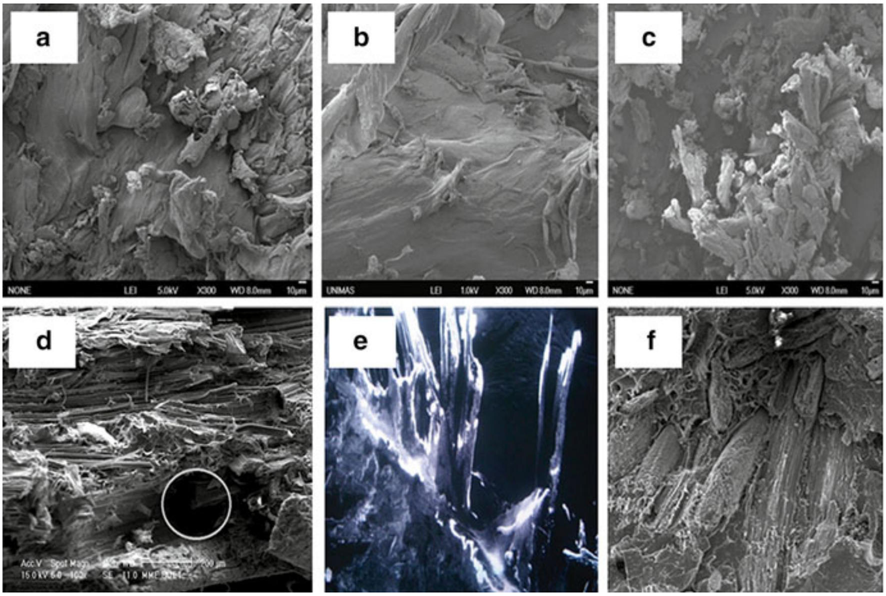

Fig. 2.5 Scanning electron micrographs of (**a**) jute, (**b**) coir, (**c**) abaca, (**d**) bagasse, (**e**) bamboo, and (**f**) palm fibre-reinforced polypropylene composites

2.6.2.1 Tensile Properties

The ultimate tensile strength and Young's modulus are the main two properties evaluated during the tensile test. Those two properties of various natural fibre-reinforced polymer composites are mentioned in Table 2.3. The tensile strength generally decreases with increase in fibre content. As the fibre load increases, the weak interfacial area between the fibre and matrix increases. This in turn decreases the tensile strength (Rahman et al. 2008; Haque et al. 2010a, b; Karim et al. 2013). Banana fibre-reinforced composites had the highest tensile strength, while palm composites had the lowest ones. On the other hand, the Young's modulus generally increases with fibre loading. During tensile loading, partially separated micro-spaces are created, which obstructs stress propagation between the fibre and matrix. As the fibre load increases, the degree of obstruction increases, which consequently increases the stiffness (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Haque et al. 2010b). The coir fibre- and banana fibre-reinforced composites had the highest and lowest Young's modulus values, respectively.

2.6.2.2 Flexural Properties

Flexural properties are some of the important parameters influencing the performance of hybrid green composites in actual applications. The variation of flexural properties of various natural fibre polymer composites is mentioned in Table 2.3.

Table 2.3 Variation of mechanical properties of various natural fibre-reinforced polymer composites (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Nahar 2014)

Type of composite	Tensile strength (MPa)	Young's modulus (GPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength	Hardness (R_L)
Jute-PP	23–29	1.6–2.4	45–54	1.7–2.8	30–51 J/m	76–91
Coir-PP	25–28	1.7–2.7	47–49	1.6–2.8	41–54 J/m	85–87
Abaca-PP	23–27	1.6–2.6	46–48	1.4–2.6	39–46 J/m	79–86
Bagasse-PP	17–22	1.2–1.4	21–34	0.8–1.6	3.3–6.2 kJ/mm^2	–
Banana-PP	36–41	0.82–0.98	–	–	10.2–12.8 kJ/m^2	–
Hemp-PP	27–29	1.6–1.8	–	–	–	–
Palm-PP	21–30	1.1–1.6	44–55	1.6–2.6	39–53 J/m	92–96

The flexural strength and flexural modulus generally increase with increase in fibre loading. Since natural fibres are high modulus materials, higher fibre concentration demands higher stress for the same deformation. Increased fibre–matrix adhesion provides increased stress transfer between them (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Haque et al. 2010b). Jute fibre-reinforced composites had the highest flexural strength, while bagasse composites had the lowest ones. On the other hand, coir fibre- and bagasse fibre-reinforced composites had the highest and lowest flexural modulus values, respectively.

2.6.2.3 Impact Strength

The impact strength is the ability of a material to withstand fracture or the amount of energy required to propagate a crack. The impact strength of different natural fibre-reinforced polymer composites is reported in Table 2.3. Generally impact strength of natural fibre-reinforced polymer composites increases with fibre addition. The impact strength mainly depends on the nature of the fibre, polymer, and fibre–matrix interfacial bonding. High fibre content increases the probability of fibre agglomeration which results in regions of stress concentration requiring less energy for crack propagation. Another factor of impact failure of composite is fibre pullout. With increase in fibre loading, bigger force is required to pull out the fibres. This consequently increases the impact strength (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Haque et al. 2010b).

2.6.2.4 Hardness

Table 2.3 shows the hardness values of some natural fibre-reinforced polymer composites. Hardness of natural fibre polymer composites generally increases with increase in fibre loading. The increase in hardness is due to the increase of stiffness of the composites with fibre loading (Rahman et al. 2008; Haque et al. 2010a; Karim et al. 2013; Haque et al. 2010b). According to Table 2.3, palm and jute fibre-reinforced composites had the highest and lowest hardness values, respectively.

2.6.3 Dynamic Mechanical Properties

Dynamic mechanical analyzer (DMA) is a frequently used characterization machine for evaluating dynamic mechanical properties of natural fibre-reinforced polymer composites. Variation of the dynamic mechanical properties of various natural fibre-reinforced polymer composites is shown in Table 2.4. The storage modulus increased with ammonium polyphosphate (APP) addition and NaOH treatment in kenaf fibre-reinforced PLA composites. This was due to better interfacial adhesion and bond strength among the matrix and fibre. NaOH treatment also reduced the loss factor values due to the same reason mentioned above (Shukor et al. 2014a). The storage modulus increased, while the loss factor decreased with increase in frequency in doum fibre-reinforced PP composites. This was due to relatively high time response of the matrix chains. Hence, at high frequencies, the composite behaved like a solid. However, at low frequencies, the polymer chains had time to relax that decreased the loss factor (Essabir et al. 2013). In kenaf fibre-reinforced polyethylene (PE) composites, the storage modulus decreased with temperature due to the softening of the matrix. On the other hand, the same property increased with fibre content due to the stiffening of the overall composite. The loss modulus and loss factor also increased with increase in kenaf fibre content in the composite.

2.6.4 Thermal Properties

Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) are the two main tools that determine the thermal properties of natural fibre-reinforced polymer composites. Variation of thermal properties of various natural fibre-reinforced polymer composites is shown in Table 2.4. The glass transition temperature increased with APP addition and NaOH treatment in kenaf fibre-reinforced

Table 2.4 Variation of dynamic mechanical and thermal properties of various natural fibre-reinforced polymer composites (Elkhaoulani et al. 2013; Shukor et al. 2014a; Essabir et al. 2013; Salleh et al. 2014; Shukor et al. 2014b; Shekeil et al. 2014)

	Dynamic mechanical properties			Thermal properties	
Type of composites	Maximum storage modulus (MPa)	Maximum loss modulus (GPa)	Maximum tan delta	Maximum glass transition temperature (°C)	Maximum thermal degradation temperature (°C)
Kenaf-PLA	5	1	1.7	67	305
Doum-PP	–	–	0.16	–	–
Kenaf-PE	8	0.7	0.24	–	–
Hemp-PP	–	–	–	–	376
Kenaf-PVC/TPU	–	–	–	–	281

PLA composites. This was due to the decrease in mobility of the matrix chains, which indicated improved interfacial adhesion between the fibre and matrix (Shukor et al. 2014a). The thermal stability of the same composites also increased with APP addition and NaOH treatment (Shukor et al. 2014b). In hemp fibre-reinforced PP composites, the thermal stability increased with increase in hemp fibre loading due to the creation of higher barrier to polymer exhaust gas formation (Elkhaoulani et al. 2013). The maximum degradation peak increased with increase in kenaf fibre content in kenaf fibre-reinforced polyvinyl chloride/thermoplastic polyurethane polymer blend composites (Shekeil et al. 2014).

2.7 Application of NFRCs

Being lightweight, economically cost-effective, highly abundant, and environment-friendly, NFRCs are useful for various applications such as products of commodity to aerospace, examples including electroactive papers, fuel cell membranes, controlled drug release mechanisms, and biosensors. Several useful applications of NFRCs in different areas are described below.

2.7.1 Automotive Applications

There is an increasing trend of utilizing NFRCs in automotive industries. Most of the major vehicle manufacturers around the world are utilizing NFRCs in various applications such as those listed in Table 2.5 (Faruk et al. 2014).

In the manufacturing of interior and exterior of vehicles, weight reduction is a key issue. Materials have to be lightweight but strong, i.e., possess good impact performance in order to avoid any occupational damage. It was reported that the reduction of vehicle weight not only reduces the fuel requirements but also reduces the greenhouse gas emission. Pandey et al. reported that a 25 % reduction in vehicle weight is equivalent to a saving of 250 million barrels of crude oil and a reduction in CO_2 emissions of 220 billion pounds per year (Pandey et al. 2010; Gejo et al. 2010; Błędzki et al. 2014).

Among various plastics, PP is the mostly used thermoplastic resin in automotive industries due to their low density, excellent mechanical properties, and processability. Several other thermoplastics such as polyethylene, polystyrene, and polyamides (nylon 6 and 6, 6) have also been used intensively in automotive industries. However, due to the raised awareness on environmental issues with the nonbiodegradable and non-recyclable polymers, most of the plastic manufacturers as well as automobile manufacturers are trying to use bioplastics as the alternative sustainable solutions. Several research works have also reported the manufacturing of bioplastic-based composites for the automotive applications. Graupner et al. (2009) reported a good number of PLA-based composites reinforced with hemp, cotton, kenaf, and

Table 2.5 Automotive models, manufacturers, and components using natural fibre-reinforced composites (Adopted from Faruk et al. 2014)

Models	Manufacturers	Components
A2, A3, A4, A4 Avant, A6, A8	Audi	Seat back, side and back door panel, boot lining, hat rack, spare tire lining
Roadstar, Coupe		
C5	Citroen	Interior door paneling
3, 5, 7 series	BMW	Door panels, headliner panel, boot lining, seat back, noise insulation panels, molded foot well linings
Eco Elise	Lotus	Body panels, spoiler, seats, interior carpets
Punto, Brava, Marea, Alfa	Fiat	Door panel
Romeo 146, 156		
Astra, Vectra, Zafira	Opel	Instrumental panel, headliner panel, door panels, pillar cover panel
406	Peugeot	Front and rear door panels
2000 and others	Rover	Insulation, rear storage shelf/panel
Raum, Brevis, Harrier, Celsior	Toyota	Door panels, seat backs, floor mats, spare tire cover
Golf A4, Passat Variant, Bora	Volkswagen	Door panel, seat back, boot-lid finish panel, boot liner
Space star, Colt	Mitsubishi	Cargo area floor, door panels, instrumental panels
Clio, Twingo	Renault	Rear parcel shelf
Mercedes A, C, E, S class, Trucks, EvoBus (exterior)	Daimler-Benz	Door panels, windshield/dashboard, business table, piller cover panel, glove box, instrumental panel support, insulation, molding rod/apertures, seat backrest panel, trunk panel, seat surface/backrest, internal engine cover, engine insulation, sun visor, bumper, wheel box, roof cover
Pilot	Honda	Cargo area
C70, V70	Volvo	Seat padding, natural foams, cargo floor tray
Cadillac Deville, Chevrolet TrailBlazer	General Motors	Seat backs, cargo area floor
L3000	Saturn	Package trays and door panel
Mondeo CD 162, Focus, Freestar	Ford	Floor trays, door panels, B-pillar, boot liner

cellulose fibres. They investigated the effect of fibre content and loading direction on their mechanical performance and found that PLA reinforced with man-made cellulose fibres showed highest tensile strength followed by hemp- and kenaf-reinforced systems.

Several other bioplastics have been commercialized by different chemical companies around the globe to replace the nonbiodegradable commodity plastics. For example, PTAT or Eastar Bio from Eastman Chemical Company, BioPET or Biomax from Dupont, Ecoflex from BASF, PLA from Dow-Cargill, and PHBV from Metabolix are other examples of environment-friendly material for automobiles.

Most of this material contains biodegradable materials, tested under composting conditions, which can degrade in the natural condition leaving behind biomass and carbon dioxide.

2.7.2 *Packaging Applications*

Polymers alone do not possess good barrier to most common gases like N_2, O_2, CO_2, or water vapor. This is why polymers are hardly used in perishable food packaging. In order to improve their barrier properties, the phase morphology can be engineered or some fillers or additives can be used. In most NFRCs, fibre phase may play significant role in improving the barrier properties of the polymers. Several research works reported that the presence of crystalline segments in lignocellulosic fibres leads to slower diffusion of gases or water molecule through the composites (Pandey et al. 2010). Lee and Wang (2006) recently reviewed the use of biofibres for packaging applications. They reported several limitations of fibre-filled plastics for packaging applications such as the hydrophilic nature of the fibres, degradability, lower mechanical properties in humid environment, and so on. However, they mentioned that such composites can be utilized in areas where plastics cannot be recycled or reused or do not have sustainable waste disposal facilities.

Natural fibre-based packaging materials have many advantages over synthetic packaging materials such as stiffness vs. weight ratio and recyclability. A good number of research works reported the production of high-performance bioplastic composites filled with fibres, and they can be applied in food and pharmaceutical packaging. In a recent study, it was found that the environmental degradation of packaging materials, which depends on the amount of organic but degradable content, is very important concerning the development of short-life disposable packaging plastics. In recent years several companies took initiatives to produce recycled packaging materials, for example, Placon Corp. has introduced EcoStar™ HS 1000, a PET-based eco-friendly recyclable material, and Toray Plastics America has developed solvent-free polyester LumiLid lidding films.

2.7.3 *Bio-Based Electronics*

Bio-based electronics is an emerging area that emphasizes the use of bio-based polymers and composites for electronic applications. The aim of such emerging research area is to create and innovate human- and environment-friendly electronics (Irimia-Valdu et al. 2014). This research area may also be useful in fulfilling the creation of low-cost, energy-efficient organic electronics. Cellulose nanocomposites with semiconductive polymer matrixes have been studied intensively for actuator and sensor applications. For example, polyaniline (PAN) reinforced with cellulose nanofibres can act as acid or base sensors or as an actuator and can be applied in heating devices (Bhat et al 2006; Nalwa 1997). Several research works reported the tuning of conductivity by modifying the surface of fibres. Cellulose fibres can be coated with conductive polymers such as PAN or polypyrrole (PPy) to improve the conductivity (Sapurina et al. 2005; Konyushenko et al. 2008). In addition, cellulosic composites have also been used as a green alternative to polyolefin-based separators in rechargeable lithium ion batteries (Kuribayashi et al. 1995; Cámer et al. 2008). Most of the cellulose-based separators are thin composite porous membranes composed of cellulose microfibres and a polymeric matrix component such as ethyl acrylate or silicon resin.

2.7.4 Other Applications of NFRCs

NFRCs have long been used in the civil infrastructures. Their applications in civil infrastructures include heat insulating boards, windows, pans, and so on. Natural fibres have also been used as the reinforcement for cements to produce low-cost building materials. Fibre cement is one such product which possesses higher toughness and crack resistance in comparison with conventional cement. In addition, fibre-reinforced composites can be utilized as the low-cost building materials in developing countries. Khan et al. developed corrugated sheets made of jute fibre-reinforced unsaturated polyester matrix (Bangladesh Economic News, April 9, 2010). They proposed that such green composite (JUTIN) also possesses good heat insulation and saltwater-resistant properties and could be best useful in coastal areas of Bangladesh. Similar initiatives have been taken in India in order to produce NFRC-based composites for building materials.

NFRCs have also been applied as fire-resistant materials. Due to the porous microstructure as well as the crystalline fractions, most natural fibres have fire-resistant properties (Chapple et al. 2010). In addition the presence of lignin in fibres also induces fire resistance to most NFRCs.

2.8 Summary and Future Prospects

Natural fibres have the potential to replace the traditional man-made fibres. They are sustainable resources for renewable natural materials. Such fibres are useful to produce lightweight and strong composites. Polymer composites with natural fibres offer a unique opportunity to maintain the sustainable development of economically and ecologically attractive technology. It is still necessary to overcome problems associated with the traditional manufacturing techniques of natural fibre-based composites. The burgeoning demand for more environment-friendly materials has already raised the interests among the scientists of different disciplines to develop a flexible and sustainable approach of composite manufacturing technology. Natural fibre-reinforced composites have already achieved wide acceptance in automotive, aerospace, packaging, construction, and other industries.

Numerous research works on bio-based polymers give the indication to the fact that the future materials will be more environment-friendly. Green chemistry has already revealed ways of producing novel biopolymers that can open new windows of thinking about polymers. Natural fibre-reinforced biocomposites can substantially replace the traditional commodity polymers, although current bioplastics still need to overcome some engineering challenges. On the other hand, recent promising works in understanding the genetic sequence of fibrous plants may offer a unique way to produce fibres with the desired properties for flexible manufacturing of composites. In fine, more bottom-up approaches are necessary to fruitfully utilize such green materials for our socio-environmental benefits.

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