Chapter 7
Natural Resources Based Green Composite Materials

M. R. Mansor, M. J. Taufiq, and A. F. Ab Ghani

7.1 Introduction

Green composites materials can be described as composite materials made from the natural resources, either as the reinforcement material, the matrix or both reinforcement and matrix. They are also known as natural fibre composites, wood plastic composites and fibre reinforced plastics. Nowadays, there have been huge efforts and accomplishment in the development of green composite materials from natural resources, especially to replace and reduce the use of synthetic composites. Green composites materials offer many advantages compared to synthetic composites for many applications. In term of material properties, the natural fibre used as the reinforcement materials for the green composites has low density property, which further contributes to a higher specific strength and specific modulus compared to synthetic composites. The low density physical property is also one of the key advantages for green composites because it helps to reduce the overall final composite weight. Furthermore, natural based fibres are also more environmentally friendly, whereby there are obtained from renewable, recyclable and biodegradable resources, as compared to synthetic based fibres which are made from non-renewable petroleum based resource. Green composite materials also offer similar product development advantages as compared to synthetic composites, such as a high degree of flexibility where the product final form with complex geometrical shape can be customized. Green composite materials are also able to be produced in varying sizes

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by using a similar manufacturing process. Besides, they are also known to have minimal impacts on users' health issues such as skin irritation, as well as encompassed fewer abrasive properties which help to reduce tool wear and prolong tool life. Another distinct advantage of green composites is lower final composite cost; due to the cheap and renewable source of constituents that making up the final composite material.

Despite the advantages, green composites also inherit several limitations that challenge~~s~~ further development of these materials, especially for higher load bearing applications. Among the most notable limitation is due to the hydrophilic nature of the reinforcement materials (which is derived from plant based natural resources). The hydrophilic nature caused dimensional instability to the final composites as well as poor fibre-matrix adhesion performance which strongly affect the structural strength of the material. Moreover, uneven fibre size also affects the structural properties due to varying load distribution along the fibre length, which causes larger stress concentration at a certain location in the final composites. These factors are currently limiting green composites application to low load bearing application.

In this chapter, an overview of green composite materials is discussed. The discussion is focused on green composites reinforced polymer matrix, as it is the most applied type of green composites made from natural resources being developed currently. Among the topics included are types of green composites, application of green composites, their mechanical properties and processing methods. This chapter also highlights the latest development of green composite materials such as hybrid green composites; green composites reinforced recycled polymer matrix and green composites with nanomaterials. The chapter is concluded with brief remarks on the opportunities which can be ventured both by researchers and practitioners, to address existing limitation such as regarding the material properties, processing and design method.

7.2 Overview of Natural Resources Based Green Composites Materials

In general, the basic structure for the formulation of green composites from natural resources comprised of the reinforcement and the matrix. The main function of the reinforcement (or the fibre) is to absorb the applied load, while the matrix acts as to hold all the fibres together to form the shape of the final composites as well as evenly distribute the load applied across the composites structure. In addition, new class of advanced green composites also encompassed nanoparticle fillers (such as nanoclays, graphene, carbon nanotubes (CNT) and cellulose crystals) to further enhance the materials existing mechanical, thermal, physical and other functional properties. Figure 7.1 shows the basic structure in green composites formulation from natural resources.

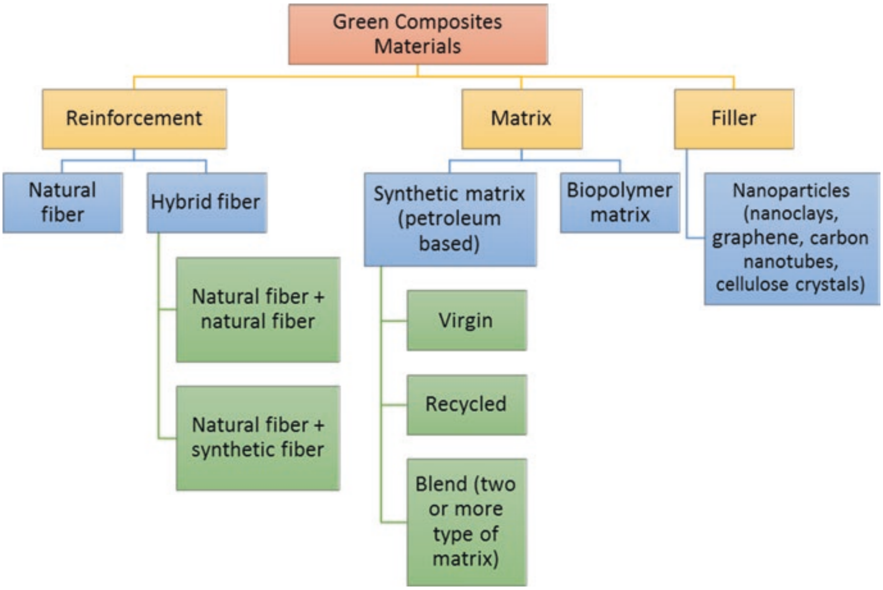


Fig. 7.1 Basic structure in green composites formulation from natural resources

Based on Fig. 7.1, there are multiple combinations on the type of reinforcement and matrices from natural resources developed for green composites. Green composites can be formulated from single fibre/single matrix up to hybrid fibre/blend of matrix. Green composites can also be formulated to become fully green composites (both reinforcement and matrix are made from natural resources) or partially green composites (either the reinforcement or matrix are made from natural resources).

7.2.1 Green Composites Reinforcement Material

Pecas et al. (2018) published a very comprehensive review on the characteristics of green composites reinforcement and matrix materials. They stated that as the main load bearing element for the green composites, the reinforcement or fibre performance depends on several factors such as fibre volume (amount of fibre used), aspect ratio, shape, orientation, arrangement and also the interfacial adhesion with the matrix. Green composites reinforcement is available in the form of particle or fibre and is characterized as continuous or discontinuous (i.e., chopped) depending on its length-to-diameter (l/d) ratio. Commonly, the fibre-reinforced phase arrangement is classed as woven or non-woven. A woven fabric is characterized by continuous interlacing of perpendicular yarns, in a regular pattern. Yarns are structures consisting of several interlocked fibres. The twist angle is responsible for the cohesion of the fibres and yarn strength up to a certain point, beyond which, the maximum fibre strength decreases due to the increase in obliquity. Moreover, the increase of the fibre twist angle is correlated with a decrease of fibre-resin bond strength, lower permeability and consequently poor mechanical properties. When continuous fibres are used, the fibre architecture can be one-dimensional and two-dimensional, or often termed as unidirectional or bidirectional. In the one-dimensional architecture, the twist angle and the level of alignment of continuous-filament yarns play a significant role in determining the maximum applied load. The reinforcement is able to hold the highest applied load in the fibre direction compared to transverse fibre directions. Pecas et al. (2018) also further explained that a non-woven arrangement consists of a flat structure without interwoven strands, with randomly oriented or unidirectional fibre orientation. Furthermore, the mat or woven can either be composed of continuous or chopped unidirectional fibres, randomly chopped fibres or suspended particles. In particular, mat composed with randomly chopped fibres does not have any preferential stress direction, but they are the preferable choice for large-scale production due to the high availability, ease and cost-effectiveness when manufacturing complex parts of isotropic nature.

Reinforcement from natural resources can be classified based on their origin into the following groups: animal, mineral and plant. Plant originated natural fibres are the most commonly accepted fibres by the industry and the most analysed by the research community due to the short growth period, renewability and wider availability (Cristaldi et al. 2010). As shown in Fig. 7.2, plant fibres are composed of cellulose, hemicellulose and lignin, which can be extracted from bast, leaf, seed, fruit, wood, stalk and grass/reed (Shah 2014). Among the types of commodity plant fibres used to manufacture green composites are kenaf, jute, flax, wood and hemp.

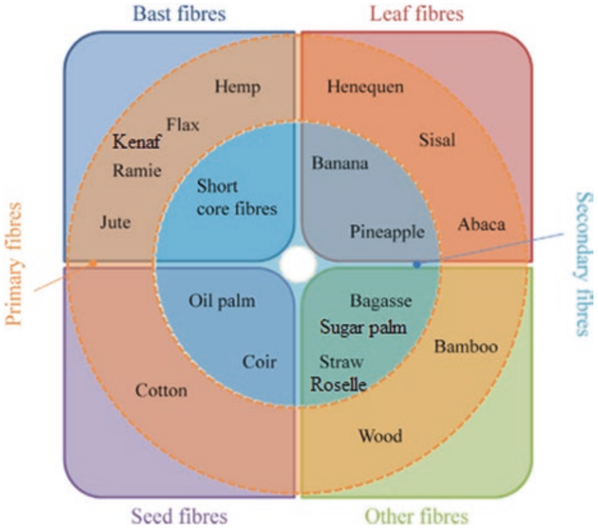


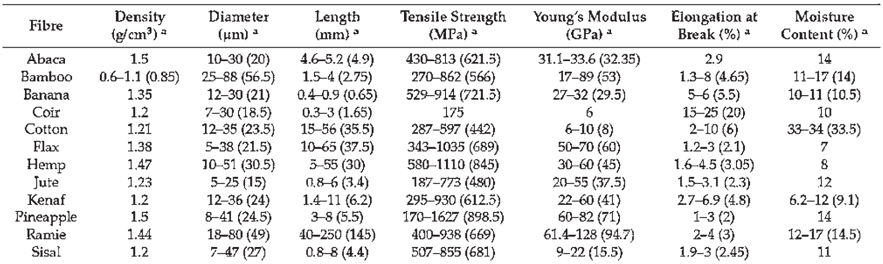
Fig. 7.2 Type of natural fibres resources (Shah 2014; Aji et al. 2009, Radzi et al. 2019a; Ishak et al. 2013)

Natural fibres offer many advantages compared to synthetic fibres such as good specific strength and modulus, low density, low manufacturing energy consumption, low cost, high acoustic damping, low carbon footprint, renewable and biodegradable (Faruk et al. 2014). However, natural fibres have negative aspects due to their low consistency of properties and quality. These fibres have higher variability of physical and mechanical properties, higher moisture absorption, lower durability, lower strength and lower processing temperature (Pickering et al. 2016). The large varieties of properties are mainly dependent upon plant species, growth conditions and methods of fibre extraction (Sanjay et al. 2016). Moreover, the related properties are also depending on the fibre cell geometry of each type of cellulose and its degree of polymerization (Ho et al. 2012; Shah 2013).

There have been many publications summarizing the important properties of plant fibres for green composites application. One of them was by Gurunathan et al. (2015) who have listed major type of plant fibres according to their physical and mechanical properties as shown in Fig. 7.3. It can be observed that among the fibres with the highest tensile strength and modulus properties available are pineapple, kenaf, flax and hemp fibres; while among the lightest fibres are bamboo, kenaf, coir and sisal fibres. Hence, consideration on the appropriate type of fibres can also be a challenge for producing green composites products, which needs to be based on the weight of importance for the selected properties to be used for the intended product. Another important information which need to be alerted for selection of suitable plant fibres are the range of values within the similar fibre itself, due to varying factors such as type of growth conditions and method of fibre extractions as stated previously.

7.2.2 Green Composites Matrices

Matrix is a vital component in the formulation of polymer based green composites. As described earlier, matrix acts as to hold all the fibres together to form the shape of the final composites as well as to distribute the load applied evenly across the composites structure. In general, there are two categories of matrix applied for



Fibre	Density (g/cm^3) [a]	Diameter (μm) [a]	Length (mm) [a]	Tensile Strength (MPa) [a]	Young's Modulus (GPa) [a]	Elongation at Break (%) [a]	Moisture Content (%) [a]
Abaca	1.5	10–30 (20)	4.6–5.2 (4.9)	430–813 (621.5)	31.1–33.6 (32.35)	2.9	14
Bamboo	0.6–1.1 (0.85)	25–88 (56.5)	1.5–4 (2.75)	270–862 (566)	17–89 (53)	1.3–8 (4.65)	11–17 (14)
Banana	1.35	12–30 (21)	0.4–0.9 (0.65)	529–914 (721.5)	27–32 (29.5)	5–6 (5.5)	10–11 (10.5)
Coir	1.2	7–30 (18.5)	0.3–3 (1.65)	175	6	15–25 (20)	10
Cotton	1.21	12–35 (23.5)	15–56 (35.5)	287–597 (442)	6–10 (8)	2–10 (6)	33–34 (33.5)
Flax	1.38	5–38 (21.5)	10–65 (37.5)	343–1035 (689)	50–70 (60)	1.2–3 (2.1)	7
Hemp	1.47	10–51 (30.5)	5–55 (30)	580–1110 (845)	30–60 (45)	1.6–4.5 (3.05)	8
Jute	1.23	5–25 (15)	0.8–6 (3.4)	187–773 (480)	20–55 (37.5)	1.5–3.1 (2.3)	12
Kenaf	1.2	12–36 (24)	1.4–11 (6.2)	295–930 (612.5)	22–60 (41)	2.7–6.9 (4.8)	6.2–12 (9.1)
Pineapple	1.5	8–41 (24.5)	3–8 (5.5)	170–1627 (898.5)	60–82 (71)	1–3 (2)	14
Ramie	1.44	18–80 (49)	40–250 (145)	400–938 (669)	61.4–128 (94.7)	2–4 (3)	12–17 (14.5)
Sisal	1.2	7–47 (27)	0.8–8 (4.4)	507–855 (681)	9–22 (15.5)	1.9–3 (2.45)	11

Fig. 7.3 Selected physical and mechanical properties of natural fibres for green composites (Gurunathan et al. 2015)

green composites, which are thermoset and thermoplastic. They are originally derived from petroleum based resources, but as the need for more environmentally friendly alternatives arise, a new class of matrix which is fully biodegradable (often term as biopolymer) are extensively developed to construct fully biodegradable green composites. This new class of biopolymer is an important solution to reduce plastics wastes issues related to petroleum based plastics nowadays.

In overall, thermoplastic materials that currently dominate as matrices for green composites are polypropylene (PP), polyethylene (PE), and polyvinyl chloride (PVC) while thermosets, such as phenolics and polyesters, are common matrices (Malkapuram et al. 2009). Elsewhere, it is also reported that PE and PP are the most widely used plastics and account for 61% of the U.S. plastics market. PE and PP have glass transition temperatures (T_g) much lower than ambient temperature, which makes them tough, and are semi-crystalline, which makes them rigid, with moderate melting temperatures (T_m), which makes them easy to process. The combination of toughness, rigidity, and processing ease along with a density of 0.93–0.96 g/cm^3 makes them useful for a wide variety of applications from bottles to automotive parts. While the semi-crystallinity gives them exceptional properties for a wide range of applications, PE and PP also exhibit the challenge in generating the largest shrinkage after molding compared to other commodity polymers, which is a direct result of the semi-crystallinity. This results in large warpage of the part while in use (Davis et al. 2019). Moreover, polystyrene, and polyamides (nylon 6 and 6, 6) are also common types of thermoplastic used especially in the automotive application. Holbery and Houston (2006) reported that the development of thermoplastic green composites is constrained by two primary physical limits: the upper temperature at which the fibre can be processed and the significant difference between the surface energy of the wood and the polymer matrix. Process temperature is a limiting factor in green composites applications. The generally perceived upper limit before fibre degradation occurs is on the order of 150 °C for long processing durations, although plant fibres may withstand short-term exposures to 220 °C. The result of prolonged high-temperature exposure may be discoloration, volatile release, poor interfacial adhesion, or embrittlement of the cellulose components. Therefore, it is important to obtain as rapid a reaction rate as possible during both surface treatment and polymer processing to limit exposure to cell wall components preventing degradation. The development of low-process-temperature surface treatments with high service capabilities is viewed as an enabling technology for the application of plant fibres in green composite materials.

Holbery and Houston (2006) also reported that among the primary thermoset resins used for green composites especially in automotive applications are polyester, vinylester, and epoxy resins. Polyester resins are widely used, particularly the “unsaturated” type capable of cure from a liquid to a solid under a variety of conditions. A range of polyesters is made from different glycols (polyethylene glycol, ethylene glycol, etc.), acids (malaeic, anhydride), and monomers, all having various properties. Orthophthalic polyester is the standard economic resin commonly used, and it yields highly rigid products with low heat resistance. Isophthalic polyester is now more common when moisture resistance is needed. Epoxy resins offer high performance and resistance to environmental degradation. On the other hand, vinylester resins have excellent chemical resistance, good thermal and mechanical properties, and the relative ease of processing and rapid cure characteristics of polyester resins. These have better moisture resistance than epoxies when cured at room temperature. Vinylester resins are similar in their molecular structure to polyesters, but differ in that the reactive sites are positioned at the ends of the molecular chains, allowing for the chain to absorb energy. This results in a tougher material when compared to polyesters.

Biopolymer is an emerging and highly potential alternative for the formulation of green composites, especially in the future. Biopolymer is made from natural resources such as plants. Sahari and Sapuan (2011) summarized several important types of biopolymer, which are further categorized into biodegradable natural based polymer and biodegradable petroleum based polymer. Among the types of biodegradable petroleum based polymer are aliphatic polyester, aliphatic-aromatic polyester, polyester amide, polyalkynene succinates and polyvinyl alchohol (PVA). On the other hand, examples of the biodegradable natural based polymer are polylactide acid (PLA), thermoplastic starch, cellulose and polyhydroxyalkanoate (PHAs). PLA is an environmentally and commercially interesting biopolymer as it has many exclusive qualities, such as good transparency, glossy appearance, high rigidity, and good processability. However, PLA also draws some limitation in term of its inherent brittleness and poor toughness (Siakeng et al. 2019a, b). Thermoplastic starch (TPS) is derived from renewable and cheap raw material feedstock such as sugar palm starch and agar (seaweed), cassava, corn, wheat, and potatoes. Starch is composed of two polymers namely amylose and amylopectin. The linear structure of amylose makes it closely resembled the behaviour of conventional synthetic thermoplastic, hence, enabled TPS to be used in various fabrication machines for its production; that is, extrusion, compression moulding, injection moulding, etc. Nevertheless, pure TPS also possesses several disadvantages such as poor mechanical strength and weak water resistance (Jumaidin et al. 2017). In overall, due to the low mechanical properties of biopolymer as compared to synthetic matrices, their application in green composite materials is limited to low load bearing conditions.

7.3 Hybrid Green Composites Materials

Single fibre/matrix green composites have been extensively studied over the years, using various types of plant fibres as the reinforcement material. However, single fibre/matrix green composites are still facing the challenges in term of low mechanical properties as compared to single fibre/matrix synthetic composites such as glass fibre reinforced composites and carbon fibre reinforced composites. To address the issue, researchers have ~~has~~ come out with a solution to use hybridization method for alleviating the properties of the green composites. The hybridization method combines two or more types of fibres in a single matrix, to form hybrid green composites. As shown in Fig. 7.1 previously, hybrid green composites may be formed using either the combination of natural based fibre-synthetic fibre or natural based fibre-natural based fibre type of reinforcements. The goal of hybridization is to improve the overall performance of the final green composites, by combining initially less strong natural fibres with stronger fibre counterparts within the same matrix. The synergetic combination enables improvement for the final green composite properties. Moreover, the hybridization method also allowed obtaining a balance between performance and cost for the final green composites, which is very beneficial for optimum use such as in application with medium load bearing requirement (Mukhtar et al. 2018).

Up to date, many efforts have been reported on the development of hybrid green composites using fully natural fibre/natural fibre type of reinforcements. This is due to the increasing drive towards formulating better eco-friendly green composite material (higher use of renewable and biodegradable contents) as well as having a lower overall raw material cost. Cavalcanti et al. (2019) have developed a novel intralaminar hybrid green composite using jute, sisal and curaua fibres. The fibres were woven together to produce hybrid jute/sisal and hybrid jute/curaua mat. The hybrid mats were later reinforced with epoxy matrix at 70 wt%. In their study, the fibres were also subjected to alkalization and mixed alkalization and silanization chemical treatments. Characterization was performed on the intralaminar hybrid green composites to determine their density, tensile, flexural impact properties. Navaneethakrishnan et al. (2019) also developed new hybrid green composites using sisal and luffa fibres. Sisal fibre was extracted from sisal leaf, whereas luffa fibres were extracted from cucumber (after its cellulose was removed). Unsaturated vinyl ester resin was used as the matrix for the green composites. They studied three hybrid sisal:luffa fibre loadings which were 10:20, 15:15, 20:10, and the hybrid green composites were fabricated using compression moulding process. In another study, Chee et al. (2019) developed hybrid green composites using bamboo and kenaf fibres. The bamboo fibre used was in the form of non-woven mat while the kenaf fibre was in the form of woven mat. Total hybrid fibre loading of 40 wt% was used to formulate the hybrid green composites, while the bamboo and kenaf fibre ratio was fixed at 30:70, 50:50 and 70:30 wt%. The hybrid composites were characterized in term of their coefficient of thermal expansion (CTE) and dynamic mechanical properties. Furthermore, Premnath (2019) also developed hybrid jute/sisal reinforced epoxy green composites. The green composites were fabricated using hand lay-up process. The jute fibre loadings were also varied. His study also involved the assessment of chemically treated (using NaOH) and untreated jute/sisal hybrid fibres in term of the mechanical properties (tensile, flexural, impact and hardness).

Other types of natural fibres were also reported as reinforcement materials for hybrid green composites. Zin et al. (2019) produced hybrid green composites by combining banana-pineapple leaf (PALF)-glass woven fibres, using epoxy matrix. Ninampure et al. (2019) developed hybrid sisal fibrils/kenaf fibres unsaturated polyester green composites. The novel hybrid fibres were targeted as an environmentally friendly alternative material for high strength electrical insulation applications. The hybrid reinforcement was varied from 10 to 40 wt%. The hybrid green composites were later characterized in term of the mechanical, thermal and electrical insulating properties. Apart from that, Asim et al. (2019) produced hybrid pineapple leaf fibre/kenaf reinforced phenolic green composites and studied their thermomechanical properties at varying hybrid fibre loadings. Elsewhere, Arulmurugan et al. (2019) also developed new aloe vera /hemp/flax natural fibre sandwich laminate hybrid green composites with the addition of barium sulfate as the filler. The hybrid green composites performance was investigated in term of their nonlinear viscoelastic behaviour, tensile and flexural strength properties.

Kumar et al. (2019) further developed hybrid Grewia optiva/Bauhinia vahlii fibre reinforced epoxy composites. This new class of natural fibres is mostly found around Himalayan region. A similar type of Bauhinia vahlii fibre was also developed as hybrid green composites, by combining the fibre with Nettle fibre (which also originated around Himalayan region). The hybrid Nettle fibre/ Bauhinia vahlii fibre was developed using epoxy matrix (Kumar et al. 2019a). In addition, Bauhinia-vahlii-weight/sisal (BVWS) fibres reinforced epoxy composites was also developed as green composites. The hybrid BVWS green composites were also added with risk husk as filler at varying filler loadings and characterized in term of the final composites physical, mechanical and sliding wear properties (Kumar et al. 2019b). The use of new and unique species of plant fibres by the aforementioned researchers showcased positive research growth towards identifying more alternative to produce high performance natural fibre for green composites application.

There are also reported studies about the effect of varying laminate stacking sequence on hybrid green composites material properties. Kumar et al. (2019) studied on the effect of different stacking sequence to the tensile, compressive, interlaminar shear strengths (ILSS) and hardness material properties for hybrid hemp/sisal reinforced epoxy green composites. The hybrid green composites were fabricated using hand lay-up and hot press method. Sanjay et al. (2019) performed a similar study using jute/kenaf reinforced epoxy and jute/kenaf/e-glass reinforced epoxy composites. They found that the addition of e-glass fibre to the jute/kenaf hybrid fibres further help to enhanced the mechanical properties. The laminates were developed using vacuum bagging method by utilizing woven fabric shape. Jothibasu et al. (2018) studied the effect of stacking sequence on the mechanical properties of areca sheath fibre/jute fibre/glass-woven fabric reinforced epoxy green composites. The natural fibres were also subjected to alkali-treatment process and the samples were fabricated using hand lay-up method. Similarly, Mohamad Hamdan et al. (2019) developed various stacking sequence for hybrid woven jute–roselle green composites to investigate its performance. They used hand lay-up method to produce the green composites. Furthermore, Khan et al. (2019) also investigated the similar effect using kenaf/jute reinforced epoxy green composites. They compared two type of stacking sequences for the hybrid fibres in the study, which are kenaf/jute/kenaf (K/J/K) and jute/kenaf/jute (J/K/J). The hybrid green composites were also fabricated using hand lay-up method.

Apart from thermoset matrix, hybrid green composites were also developed using thermoplastic matrix. For example, Radzi et al. (2019a) developed of roselle (RF)/sugar palm (SP) fibre reinforced thermoplastic polyurethane (TPU) hybrid green composites using hot compression moulding process. The fibres used were in short fibre form. They also further investigated the roselle (RF)/sugar palm (SP) fibre reinforced thermoplastic polyurethane (TPU) hybrid green composites properties when subjected to alkali treatment process (Radzi et al. 2019b). Moreover, Siakeng et al. (2019a, b) developed pineapple leaf fibres (PALF)/coir reinforced polylactic acid (PLA) green composites in short fibre form, which is targeted for food packaging application. The hybrid green composites were produced with different fibre ratios and manufactured using internal mixer plasticizer and hot press machine. In addition, Rahman et al. (2019) developed short fibre hybrid composites using the combination of kenaf and jute fibres, and bound together using polyethylene matrix. The hybrid natural fibres were also subjected to alkaline-treated and compared with untreated hybrid fibres for their physical, mechanical and thermal properties. Table 7.1 summarized the recent works available from literature on the development of hybrid green composites from natural resources.

7.4 Green Composites Materials Using Recycled Polymer Matrix

Synthetic polymer is a man-made material, containing organic or semi organic material which is malleable and can be moulded into almost any simple and complex shapes. With regards to this fact, the synthetic polymer has been used in varieties of applications in human daily life including in technical used or household appliances. The special characteristics of low density, possibly adjustable properties, easy workability, and resistance to moisture and chemicals have made the synthetic polymer can take over the conventional materials such as metal (Worsfold et al. 2019). However, the broad applications of synthetic polymer have caused another problem, faced by many countries, which is the rising of plastic waste abundance. The rising of plastic waste abundance also could reduce the landfill area. Ritchie and Roser (2018) reported that high-income country tends to produce more plastic waste. This was supported by the data collected by Dorger (2019) where it shows that China has produced the most plastic waste (59.8 million tons/year), followed by United States (37.83 million tons/year) and Germany (14.48 million tons/year). In addition, the polymer waste might be produced from the post-consumer use or even from the industrial sector. Hence, the polymer wastes need to be recycled ~~as~~ to reduce the waste abundance as well as extending the life cycle of the synthetic polymer.

The growing awareness of eco-friendly materials must be paralleled with environmental regulations and conservation. Green composite is said as an eco-friendly material, since it is made by the combination of synthetic polymer (as matrix) and natural fibre (reinforcement). For some application, the green composite can replace the conventional material, because of its characteristics of low cost, low density, nonabrasive, noncorrosive, inherent biodegradability, acceptable specific strength

Table 7.1 Summary of recent works on the development of hybrid green composites from natural resources

Year	Type of hybrid green composites	Authors
2019	Jute/sisal and jute/curaua reinforced epoxy composites	Cavalcanti et al. (2019)
2019	Sisal-luffa reinforced unsaturated vinyl ester composites	Navaneethakrishnan et al. (2019)
2019	Bamboo/kenaf composites	Chee et al. (2019)
2019	Jute/sisal reinforced epoxy composites	Premnath (2019)
2019	Grewia optiva/Bauhinia vahlii fibre reinforced epoxy composites	Kumar et al. (2019)
2019	Nettle fibre/Bauhinia vahlii reinforced epoxy composites	Kumar et al. (2019a)
2019	Bauhinia-vahlii-weight/sisal reinforced epoxy composites	Kumar et al. (2019b)
2019	Banana/pineapple leaf (PALF)/glass reinforced epoxy composites	Zin et al. (2019)
2019	Roselle (RF)/sugar palm (SP) fibre reinforced thermoplastic polyurethane (TPU) composites	Radzi et al. (2019a); Radzi et al. (2019b)
2019	Hemp/sisal reinforced epoxy composites	Kumar et al. (2019)
2019	Jute/kenaf reinforced epoxy and jute/kenaf/e-glass reinforced epoxy composites	Sanjay et al. (2019)
2019	Areca sheath fibre/jute fibre/glass-woven fabric reinforced epoxy composites	Jothibasu et al. (2018)
2019	Woven jute/roselle composites	Mohamad Hamdan et al. (2019)
2019	Kenaf/jute reinforced epoxy composites	Khan et al. (2019)
2019	Pineapple leaf fibres (PALF)/coir reinforced polylactic acid (PLA) composites	Siakeng et al. (2019)
2019	Kenaf/jute reinforced polyethylene composites	Rahman et al. (2019)
2019	Sisal fibrils/kenaf fibres unsaturated polyester composites	Ninampure et al. (2019)
2019	Pineapple leaf fibre/kenaf reinforced phenolic green composites	Asim et al. (2019)
2019	Aloevera /hemp/flax natural fibre sandwich laminate composites	Arulmurugan et al. (2019)

and stiffness, easily available, and recyclable (Naveen et al. 2019). Moreover, the current development on green composite can be seen on the using of recycled synthetic polymers as the matrix. The using of a recycled synthetic polymer may reduce the material cost and abundance plastic waste, and support sustainable product development (Taufiq et al. 2017).

In the 2018, Taufiq et al. discovered the mechanical and thermal properties of kenaf fibre reinforced recycled polymer blend composite. The recycled polymers were received from rejected unused disposable diapers, obtained from a disposable diaper manufacturer. Mainly in disposable diapers, the plastic parts were made by polyethylene, polypropylene, polystyrene, super absorbent material, and colouring materials (Espinosa-Valdemar et al. 2011). Those materials are synthetic polymer, thus suitable to be used for green composite matrix. Through electron microscopy examination on the fractured samples, it can be seen the using of recycled polymers and kenaf fibre are compatible within the range of 30–40 wt.% of fibre. The flexural strength also proved the maximum strength can be found on the point of 30 wt.% and started to decrease at 40 wt.%. On the Izod strength characteristic, the addition of kenaf fibre does not improve~~d~~ the impact strength. The deterioration of impact strength may due to fibre-to-fibre contact, causing the fibre breakage to be dominant. In addition, the increases of fibre content have affected the insufficient matrix to hold the reinforcement. The absence of coupling agent also induced the fibre pull-out, poor adhesion, and failure on matrix and reinforcement interfaces. Figure 7.4 showed the failure mode on the tested samples.

Similar work was done by Turku et al. (2018), on the using recycled synthetic polymer that received from municipal and construction wastes. Their work was comparing the durability towards manipulated weathering of the green composite made by two types of recycled polymer matrix. As reported in their earlier studies, the recycled polymer that obtained from construction contained the packaging and non-packaging plastics (Turku et al. 2017). Meanwhile, the municipal recycled polymers were made of household packaging and appliances. These construction and municipal polymer waste generally made by polyethylene, polypropylene, polystyrene, poly vinyl chloride (PVC) and other polyolefin materials, thus essential for green composite production. The freeze-thaw cycling method was used to examine the irreversible effect of water absorption on the mechanical properties of the composites made by two different matrices. The thickness swelling and water absorption more in composite made by recycled municipal plastic rather than recycled construction plastic, showing higher wettability. This caused the fibre swelling that leads to stress in matrix, inducing microcracking formation. In addition, the water absorbed also weakened the fibre and matrix interfacial interaction, which later support the reason for composites made by recycled municipal plastic have lower ~~in~~ mechanical strength and modulus. Figure 7.5 showed the SEM images of the microcracks that occurred in the matrix.

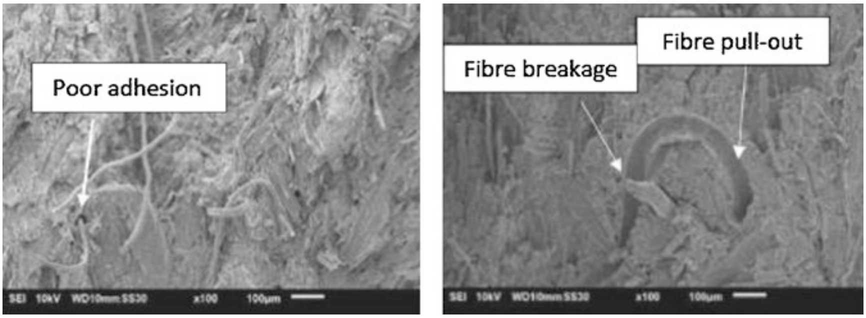


Fig. 7.4 Some of the detected failures on the Izod impact fractured samples (Taufiq et al. 2018)

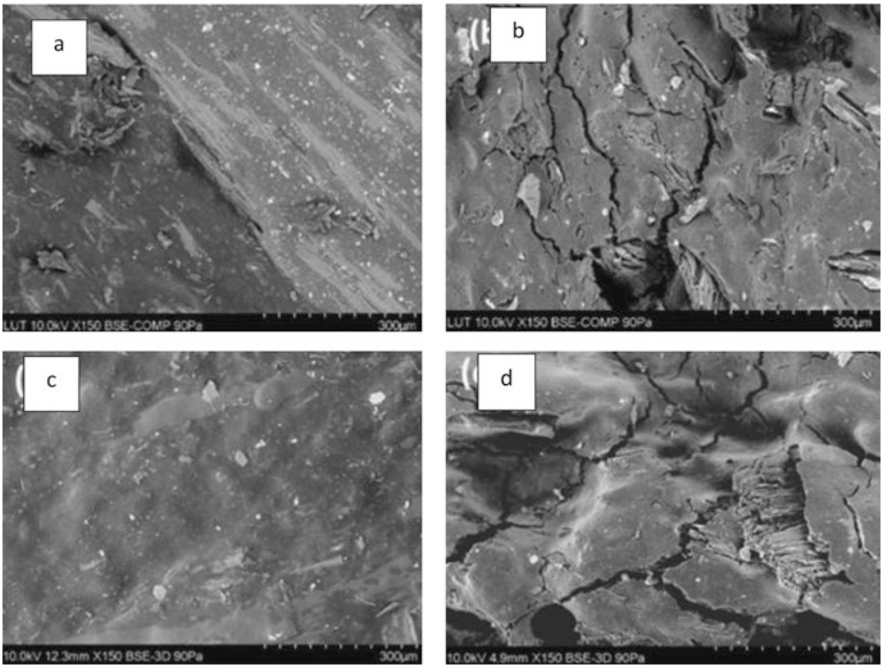


Fig. 7.5 SEM images of the microcrack on the matrix; (**a**) before, (**b**) after water absorption of recycled construction plastic, (**c**) before (**b**) after water absorption of recycled municipal plastic (Turku et al. 2018)

7.5 Green Composites Materials Using Nanomaterials

In the pursue to address the existing limitation of green composites, efforts have also been made by researchers to incorporate nanomaterials as filler material. These new classes of advanced green composites encompassed nanomaterial fillers such as nanoclays, graphene, carbon nanotubes (CNT) and cellulose crystals to enhance the materials existing mechanical, thermal, physical and other functional properties. Among the nanomaterials, nanoclays are leading the development of advanced green composites. Nanoclays play a significant role to improve composite performance by enhancing their properties such as thermal stability, mechanical strength, and barrier properties. Some of the important parameters which contribute the most in modifying the properties of various composites include the content, shape, size, and the affinity towards matrix material (Cheng et al. 2016).

Among several examples on the application of nanoclays in green composites was by Rajesh et al. (2019) who developed short madar fibre reinforced polyester composite using nanoclay to increase its mechanical properties. The composite specimens were prepared with the addition of 1 wt.% nanoclay at varying weight proportions of short madar fibre in the polyester matrix from 2.5 to 15 wt.%. Meanwhile, Mohan and Kanny (2019) developed banana reinforced epoxy green composites with the addition of Na + montmorillonite (MMT) nanoclay to enhance the unmodified composites mechanical properties (tensile, flexural and compressive strength). Shahroze et al. (2018) also used MMT nanoclay to enhance the mechanical properties of sugar palm reinforced polyester composites. They stated that, MMT nanoclay is the most widely applied constituent in composites manufacturing applications due to its good tendency for cation exchange, high aspect ratio and good swelling properties as compared to other nanoclay materials such as saponite, hectorite, stevensite, beidellite and nontronite.

Apart from nanoclays, enhancement of green composites is also performed by the addition of graphene nanomaterials. Graphene is regarded as the future materials due to its superior mechanical, thermal and electrical properties. Among the many recent works on the use of graphene in green composites is by Sarker et al. (2019). They used graphene oxide (GO) and graphene flakes in the preparation of jute reinforced epoxy composites using hot press method to improve the unmodified composites tensile strength and tensile modulus properties, tailored for various stiffness-driven applications. Naveen et al. (2019) also used varying graphene nanoplatelets loadings in improving the mechanical and moisture resistant properties of Kevlar (K)/cocos nucifera sheath (CS)/epoxy hybrid green composites. Elsewhere, Dang et al. (2019) employed GO in the formulation of ramie fibre reinforced polypropylene green composites, to enhance the materials interlaminar shear strength. The GO-ramie fibre reinforced polypropylene green composite was prepared by hot press method.

There are also many recent positive improvements regarding the green composites modified with carbon nanotubes (CNT) and cellulose nanocrystals which have been reported by the related research works. Nor et al. (2019) demonstrated the improvement of impact properties for hybrid bamboo/glass fibre polymer composites filled with CNT as nanofillers. Meanwhile, Wang et al. (2019) applied multiwalled carbon nanotubes (MWCNTs) to modify the mechanical and thermodynamic properties of flex reinforced epoxy composites. On the other hand, among the research works reported on cellulose nanocrystal for green composites was by Chang et al. (2019) who developed sisal reinforced starch composites using starch nanocrystals (SNCs). In their study, the SNCs were obtained by the hydrolysis of waxy starch. It was shown that the addition of SNC helped to improve the interfacial adhesion of the sisal reinforced starch composites. Whereas, Zhang et al. (2017) reported the development of new sisal reinforced epoxy composites with the addition of cellulose nanocrystal. In their study, sisal fibres were first treated with alkali before the cellulose nanocrystal were deposited onto the fibre surface using electrophoretic deposition (EPD) method.

7.6 Application of Green Composites Materials

Up to date, there are many reports on the application of green composites, which provide clear evidence on its positive growth for both research and development (R&D) and commercialization purpose. Grand View Research (2018), one of the leading market research and consulting firm published a forecast report on the green composites commercialization for year 2018 until 2024, stating that the global green composites market size was valued at USD 4.46 billion in 2016, and expected to steadily increase with cumulative annual growth rate (CAGR) of 11.8% from the year 2016 to 2024 (Market Research Report 2018). The company also reported that major types of natural fibres used in the current application are wood, flax, kenaf, cotton, and hemp. Wood fibre~~s~~ top the list with 59.3% of the overall natural fibre market revenue in 2015. Meanwhile in term of market application, green composites are widely used by building and construction, automotive, electronics and sporting goods industry. Product examples are decking, railing, window, building frames, mobile cases, laptop cases, tennis racket, bicycle, and snowboards. The highest market application of green composites is recorded by building and construction industry, followed by the automotive industry based on 2016 data from similar report. Among the major factors contributing to the rapid growth of green composites, especially from natural resources are spiralling demand for lightweight products from the automotive industry, rising awareness about green products, growing inclination towards eco-friendly products, and urging uptake of recyclable products mainly by consumers and industrial players.

There are two major factors that drive the rapid progress of green composites in automotive application; (1) vehicle light-weighting and (2) end-of-life performance. It is estimated that a 25% reduction in car weight would be equivalent to saving 250 million barrels of crude oil. Thus, the utilization of low-density natural fibres towards the formulation of green composite materials could lead to a weight reduction of 10–30%, which significantly contribute to achieving lightweight vehicle target. Moreover, the recycling concerns being driven by EU regulations (ELV) are forcing automotive manufacturers to consider the environmental impacts of their production and possibly shift from petroleum-based to agro-based materials. Agro based materials such as green composites derived from natural resources provide better end of life performance in term biodegradability and recyclability as compared to synthetic composites and other conventional engineering materials. Furthermore, green composites also provide advantages in term of low raw materials cost for vehicle production as they are renewable resources with high availability (Mansor et al. 2014a, b). Pecas et al. (2018) listed a very extensive list of existing green composites used in vehicle interior application such as door panels, panels and other trim components. Green composites are also reported to be used as fillers in automotive friction components (brake and clutch pads) such as areca sheath fibre (Krishnan et al. 2019), banana fibres (Zhen-Yu et al. 2019) and kenaf fibres (Abdollah et al. 2015).

However, despite the advantages, the use of green composites from natural resources are still dominated for low load bearing applications due to the relatively low structural strength of the composites as compared to synthetic composites and metals. Nevertheless, steady progress has been reported by researchers to venture into medium load bearing application using green composites. Mastura et al. (2017) have developed a conceptual design of automotive anti-roll bar using hybrid sugar palm/glass fibre composites while Ishak et al. (2018) proposed a new design of automotive car front hood using kenaf fibre-metal-laminates (FML). Furthermore, Mansor et al. (2014b) developed a new conceptual design of automotive parking brake lever using hybrid kenaf/glass reinforced polypropylene composites and while Shahruzaman et al. (2019) proposed a new concept of vehicle side door impact beam using kenaf reinforced thermoplastic composites. In addition, Mansor et al. (2015) also proposed a new design of automotive rear spoiler using kenaf reinforced thermoplastic composites, utilizing sandwich construction solution to gain higher product rigidity. The use of innovative design approach such as Theory of Inventive Problem Solving (TRIZ), biomimetics and multicriteria design making tools (such as Analytic Hierarchy Process (AHP) and Vlse Kriterijumska Optimizacija Kompromisno Resenje (VIKOR)) as demonstrated by the aforementioned researchers are spearheading new solutions for green composites to be used in higher load bearing applications.

Green composites are also reported to be applied for new application such as aerospace and military industries, particularly due to their lightweight, impact resistance and vibration damping properties. Mansor et al. (2019) summarized the recent application of green composites in the aerospace industry such as for the development of aircraft radome, aircraft interior cabin component and aircraft honeycomb structure. Furthermore, Strohrmann et al. (2019) also reported the use of hybrid flax/carbon fibre polymer composites to develop lightweight helicopter cockpit door component. On the other hand, several researchers have also reported the use of green composites in the military industry. Kumar et al. (2019) developed armour component using conch seashells/chopped banana fibres/aluminium layer reinforced epoxy composites. The hybrid green composites were later subjected to ballistic test with the short hand gun, and results obtained showed that composite successfully fit for ballistic protection application. Braga et al. (2017) constructed lightweight and efficient Multilayered Armor Systems (MAS) component using laminate shape. The MAS laminate consists of three layers, whereby the front layer was made from ceramic, followed by sisal reinforced polyester composite for the second layer, and ductile metal for the third layer. Results from ballistic tests performed using class III 7.62x51 mm ammunition showed that 30 vol.% sisal reinforced polyester composite and the conventional aramid laminates were equally efficient in terms of MAS second layer due to similar capacity of the different composites to retain the fragments generated by the interaction of the projectile with the front ceramic. Meanwhile, Yahaya et al. (2016) developed a new military vehicle spall liner component using hybrid kenaf/aramid reinforced epoxy composites. The hybrid composites were produced in laminate form using woven kenaf mat from unidirectional kenaf yarn of 800 tex at 0°/90° ply orientation.

7.7 Mechanical Properties of Green Composites Materials

Mechanical properties of the final material depend on many different parameters. The main parameters are the fibre and matrix material properties and the compatibility of matrix and the fibre bundle. Many investigations have been carried out on the potential of natural fibres as reinforcement for composites, and in several cases, the results have shown that natural fibre composites reached a good stiffness, but their final strength was not improved (Oksman et al. 2002; Mathew et al. 2005). In addition, mechanical properties were also useful in the identification and classification of materials for different applications. The considerable properties of mechanical tests are tensile strength, modulus, impact resistance, compression, hardness, and toughness. These properties also depend on the orientation of the reinforcements and atmospheric conditions.

Starch-based plastic mixtures possess poor mechanical properties such as low tensile strength, stiffness, low elongation at break, low humidity stability and release small amounts of plasticizing molecules from a starch matrix. Modification of starch, with the use of compatibilizer, reinforcement, and improvement of process conditions, are likely to make starch as a practical plastic substitution material (Zhang et al. 2007).

Elongation, thermal stability, and water resistance of PU/PDSP films increase with the addition of PU (Liu et al. 2008). Lu et al. (2005) developed PU from rapeseed oil-based polyols, and then used it to modify glycerol plasticized starch (PS) to overcome the disadvantages of starch, namely poor mechanical properties and water sensitivity. The results exhibited that plasticized glycerol starch could be combined with rapeseed oil-based PU at PU content below 20% and phase separation ascended when PU content increased. Incremental of PU into the starch matrix also intensify film resistance to water (Lu et al. 2005).

Kumar and Siddaramaiah (2005) studied the effect of coating bamboo fibres with Polyurethane (PU) and Polyurethane/Polystyrene Interpenetrating Network (PU/PSIPN) on the tensile property of the composites. Both the untreated / alkali treated bamboo fibres were coated with polyethylene glycol based PU and its semi interpenetrating network (SIPN) with PS. It was found that the tensile strength of bamboo has increased after coating with PU and PU/PS system. PU/PS coating on alkali treated bamboo fibres have shown a rise (74%) in the tensile load at break than PU (11%) coating on alkali treated fibre. Lee and Ohkita (2004) have fabricated biocomposites of poly (lactic acid) (PLA)/bamboo fibre (BF) and poly (butylene succinate) (PBS)/bamboo fibre (BF). They have investigated the effect of lysine based diisocynate (LDI) as coupling agent on properties of bio composites. They have reported that tensile properties and water resistance were improved by the addition of LDI. These improvements were due to enhanced interfacial adhesion between polymer matrix and bamboo fibre.

Chen et al. (1998) have fabricated bamboo reinforced polypropylene (PP) composites. Polypropylene was modified with maleic anhydride. Modified polypropylene was prepared by solution grafting method. They have investigated the effect of bamboo fraction, MAPP content and bamboo sizes on mechanical properties of unmodified and modified Bamboo Fibre/PP composite. Tensile modulus of modified composite increased with bamboo content up to 65 wt%, but modulus of unmodified composite was not affected by changing bamboo fraction. Tensile strength of modified composite has increased about 50 wt% bamboo fibre, but for unmodified composite, tensile strength has decreased slightly. This effect was also studied by Tran et al. (2011) using a compatibilizer of maleic anhydride polypropylene (MAPP), it enhances the flexural strength compared to pure PP, and this resultant material becomes stronger and less flexible. A similar effect was observed by Lee et al. (2009) while adding 3-glycidoxypropyltrimethoxysilane (GPS) as a coupling agent in the PLA/kenaf fibre biocomposites. The flexural strength and flexural modulus of the composites increased with increasing the content of GPS, while compared with pure PLA. This coupling agent significantly increases the interfacial strength between resin and fibres.

Thwe and Liao (2002, 2003) have fabricated the bamboo reinforced polypropylene (BFRP) and bamboo-glass reinforced Polypropylene (BGRP) composites. They have studied the effect of fibre content, fibre length, bamboo to glass ratio, coupling agent (maleated polypropylene) on tensile and flexural properties of these composites. It was reported that moisture absorption of BFRP during aging can be reduced by replacing bamboo fibre with glass fibre and by using Compatilizers. Mechanical properties of BFRP and BGRP have degraded after aging in water. Rozman et al. (2004) studied the effect of isocyanate (MDI and toluene diisocyanate-TDI) treatment on the mechanical properties of EFB composites with polyethylene (PE) as the matrix. The composite with fibre treated with MDI showed higher tensile and flexural strengths than those treated with TDI. It is well known that to achieve optimum mechanical strength and stiffness in a composite, the reinforcement fibres should be continuous and aligned in the direction of the applied load. The same is true for natural fibres.

Moreover, there are also studies which reported on the use of physical treatments such as plasma and corona discharge to improve the functional properties of natural fibres. Plasma treatment in oxygen can roughen the surface. Low-temperature plasma treatments improve the surface of natural fibres by causing chemical implantation, etching, polymerisation, free radical formation and crystallisation (Mohanty et al. 2005). For example, the wettability of wood fibres has been found to improve significantly with an increased level of corona treatment (Wallace 2005). Also, there are some methods that combine the physical and chemical methods and called physico-chemical methods such as hydrothermal and steam explosion (Jawaid and Khalil 2011; LI et al. 2007). Badri et al. (2001) prepared medium-density fibreboard (MDF) with palm-based PU as the binder. Various sizes of refined EFB ranging from 53 ∝m to 500 ∝m and fixed blending ratio of PU to EFB at 20:80 were used. The smallest size of EFB fibres gave higher impact strength and better water resistivity due to the lesser voidage between the EFB particles.

Apart from physical treatment, alkali treatment is also a common method to clean and modify the surface of natural fibres to promote enhanced fibre-polymer adhesion. Mercerization is a traditional alkali treatment based on sodium hydroxide

(NaOH, caustic soda), which improves the take up of dye in textile processing. Several studies have found that alkali treatments can improve the properties of natural fibres and the interfacial adhesion to polymers (Liu et al. 2004; Bachtiar et al. 2008). In addition, acetylation is also a treatment of particular interest in natural fibres. In this treatment, acetic anhydrides substitute the cell wall hydroxyl groups of a natural fibre with acetyl groups, rendering the surface more hydrophobic and thus less susceptible to moisture uptake and biological attack and more compatible with polymer matrices (Tserki et al. 2005). Isocyanates can be used as coupling agents to improve the bonding of natural fibres to polymers. For example, the strength and stiffness of wood-polypropylene (PP) composites can be increased by treating the fibres with polymethylene-polyphenyl-isocyanate (Wallace 2005). For example, lysine diisocyanate has been successfully used to improve the mechanical properties and water resistance of bamboo fibre-polylactide composites through enhanced fibre-matrix interfacial adhesion (Lee and Wang 2006).

There are a few epoxy resins on the market which contain a proportion of bio-based material. It is also possible to use bio-based products to improve the performance of synthetic resins. For example, the impact strength of petroleum-based epoxies can be significantly improved by blending with epoxidised vegetable oils (Mohanty et al. 2005). However, if natural fibres were to be used in combination with plastic polymers, the resulting composites would have low tensile strength values and poor interfacial adhesion because of incompatibility between the kenaf fibres and plastics (Yussuf et al. 2010; Nishino et al. 2003; Akil et al. 2011; Alayudeen et al. 2015). In order to compensate for these disadvantages, various chemical treatments have been studied. Among the chemical modifications, acetylation is one of the most commonly used methods, where the –OH groups that are responsible for hydrophilic properties of lignocellulose are modified by hydrophobic acetyl groups (Hu et al. 2011). For example, acetylation can be used to control hygroscopic properties, dimensional stability, durability, and the physical properties of plant-based materials and composites. This chemical modification strategy is regarded as an inexpensive, simple method for lowering the surface energy characteristics of natural fibres to make them more compatible with common polymers (Ismail et al. 2011; Ifuku et al. 2010). If kenaf fibres were made less hydrophilic, their compatibility with PLA would be improved, leading to an enhancement in adhesion, and thus also the strength of the resulting composites.

The flexural and tensile strengths of the kenaf–PLA composites were enhanced when the introduced acetyl constituted over 25% of the mass of fibres. With respect to the lower acetylation levels that were examined in this study, there was no improvement in the mechanical properties as compared with the untreated kenaf–PLA composites. The detrimental surface smoothening of kenaf upon brief acetylation, as evidenced by morphology studies, was a possible explanation for such mechanical performance (Chung et al. 2018).

On the other hand, modifying the composites architecture through alignment of fibre orientation could also alter the mechanical properties of the green composite produced from natural resources. Aligned natural fibres can be combined with thermoplastic polymers in a number of ways. Woven or stitched fabrics can be interleaved with thermoplastic sheets or films, often referred to as film stacking, then heated and consolidated in a press or by vacuum bagging (Arnold et al. 2007). Natural fibre-thermoplastic pultrusion is also possible (Van and Kiekens 2001), although the natural fibre materials must have sufficient strength to resist breaking during the process. Thermoplastic polymers are highly viscous so do not flow readily and care must be taken during processing, as with all thermoplastic composites, to ensure good impregnation and consolidation. By using aligned natural fibres, the mechanical properties of these biocomposites are significantly higher than non-woven mat composites (Hoydonckx et al. 2009). Compatibilizers are often used to improve fibre-matrix bonding, such as maleicanhydride for PP. In particular, the development of aligned, natural fibre fabrics suitable for composite reinforcement will provide significantly enhanced properties and will open the door to a range of semi-structural applications. Compatibilization is necessary to obtain a composite with tailored mechanical properties and good efficiency in the transferring of the stress from the matrix to the fibres.

7.8 Manufacturing Process of Green Composite Materials

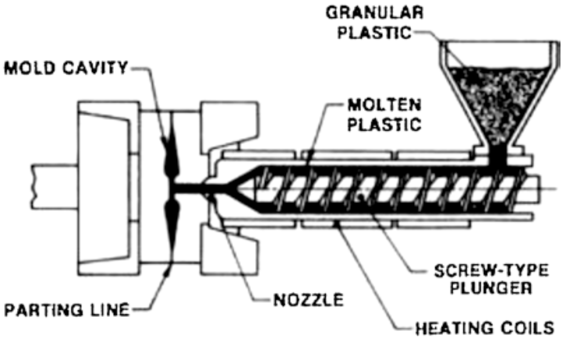
A polymer is a material made by macromolecule, built of many repeated monomer subunits, which have a significant and broad role in human daily life. Either can be synthetic or natural polymer, the synthetic polymers are petroleum-based material which falls into four major divisions, namely thermosets, thermoplastic, elastomer, and synthetic fibre. Meanwhile, the natural fibre can be found from plant, wood, or even from animal protein. The monomer can chemically react with another monomer with the same or different molecules that occurred naturally is called ~~as~~ natural polymer, where the derived or man-made polymers are known as a synthetic polymer. The broad application of polymer nowadays in human daily life including for medication, nutrition, communication, transportation, irrigation, container, clothing, recording history, buildings, highways, and so on, made the polymer industry the rapidly developed and wider than the copper, steel, aluminium and some other industries (Namazi 2017).

Although the polymer has played a significant role in our daily life, there is no perfect material that currently found. As mentioned earlier, the polymer is created by the chemical reaction of a monomer with another monomer. This can be supported by applying the heat during the chemical reaction, so that the polymer chains can break up and combined with other polymer chains. However, too much heat could cause thermal degradation which later reduces the mechanical properties of the desired material. During production, heat can be intentionally applied (by the heat of mould, or extruder) or unintentionally applied (speed or revolution of extruder or mixer). The thermal degradation could occur when the strained or entangled sections of the polymer chains to be free. The rearrangement of these freed macromolecules segments were changed from the previous arrangement polymer chains, thus affecting the crystallinity of the material. The variation of ordered molecular chains or known as the degree of crystallinity is later affecting the mechanical properties of the polymer material (Costa et al. 2007; Madi 2013). In conclusion, the temperature selected for the production process is vital so that we can minimise the thermal degradation.

Various manufacturing methods can be used for green composite production, either the injection moulding, hand layup, or rapid prototyping. The selection of manufacturing process is depending on the type of polymer used. Injection moulding is one of the manufacturing processes that involved with the using of polymer. According to Todd et al. (1994), the injection moulding commonly used for thermoplastic material, where the molten polymer is injected into the mould cavity at a certain temperature and speed. As can be observed in Fig. 7.6 below, the material in granule shape is fed into the barrel that containing the heater and forced out into the mould cavity by screw plunger. The molten polymer is left cooled inside the cavity that has been taken out. In green composite, the using injection moulding is not a new thing. Various researchers have reported the production of green composite by using this method. Montanes et al. (2019) studied the potential to use conventional injection moulding method for the green composite made by a biobased high density poly(ethylene) matrix and a high lignocellulosic filler from industrial thyme wastes. The modelling works were done with parallel with experimental validation. By using 100:0, 90:10, 80:20, 70:30, 60:40, and 50:50 HDPE:thyme wt.% ratio, the best formulation was found up to 20 wt.%, as can be found in Fig. 7.7. The process temperature and shear speed are the important parameters in this process. The higher temperature will provide lower viscosity, but it would induce thermal degradation onto the filler and the matrix. Furthermore, as the thyme fibre wt.% increases, it requires higher pressure to absolutely fill the mould cavity, which later may damage the pressure transmission. In overall, the green composites were successfully produced by using SLS method, with the optimum matrix to filler ratio not more than 20 wt.%. In addition, the simulation tools are useful in predicting and validating the rheological behaviour of high lignocellulosic content polymer systems.

In order to maintain the lower production costs at a high volume of the green composite, the hand layup method was commonly used which it requires the manual laying down the individual layers or plies of reinforcement (natural fibre). The plies can be in the form of single unidirectional or woven, which involves the

Fig. 7.6 The schematic diagram of typical injection moulding machine (Todd et al. 1994)



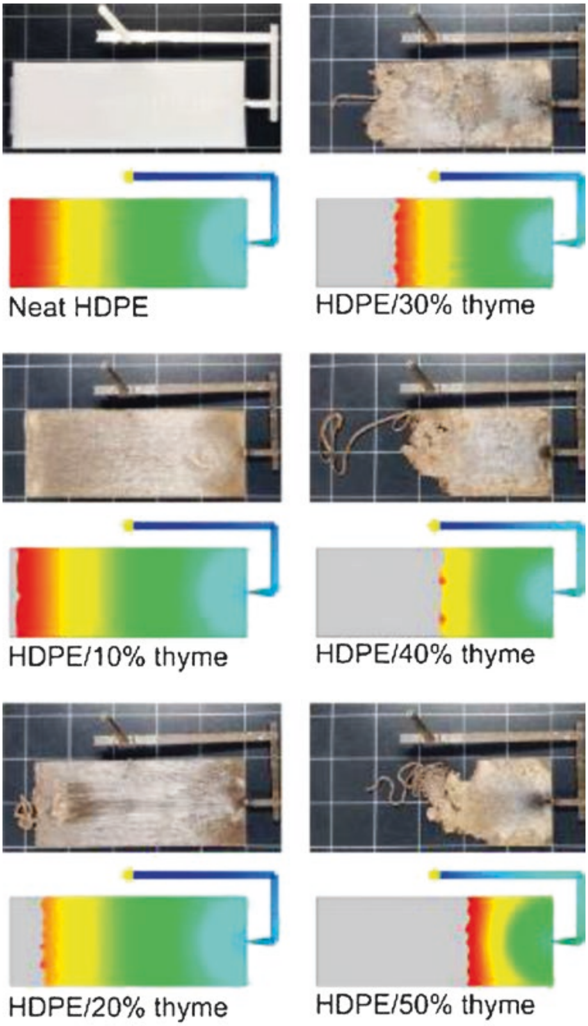


Fig. 7.7 Comparison on the injected green composite with the variation of thyme fibre ratio (wt.%) (Montanes et al. 2019)

manipulating each ply into shape by hand, and then firmly pressed with or without mould by ensuring there is no air pocket between plies (Elkington et al. 2015). Normally, thermoset polymers were chosen as the matrix as it is a liquid resin which suitable for the hand layup method. One of the modifications in hand layup method is by using two or more reinforcement plies, called as a hybrid green composite. As reported by Yahaya et al. (2015), hybridisation of the two types of reinforcement could be one of the solutions on the natural polymer limitations. Combining two types of reinforcement could obtain the full advantage of the properties of the constituents, and become an optimal, superior but economical step in improving green composites. In their research, the kenaf fibre was hybrid with Kevlar and pressed with epoxy resin. As expected, the hybridisation of kenaf fibre with Kevlar does improve~~d~~ the mechanical properties (tensile, flexural, and impact) of the green composites. In addition, the hybrid with the Kevlar as the outer part (Kevlar/kenaf/Kevlar) also produced better mechanical properties compared to (kenaf/Kevlar/kenaf). The layering sequences are portrayed in Fig. 7.8 below.

The rapid prototyping or so-called 3D printing is one of advanced manufacturing which has been available in late 1980s by using layered method and using laser light for fusing the metal powder in solid prototypes (Bagaria et al. 2011). In addition, the rapid prototyping can be used to produce a product that meeting the requirements of the shape and precision, shortening the design and evaluation cycle of a new product, and providing the actual and observable look of the model (Jiang et al. 2010). The application of rapid prototyping can be seen in automotive, aerospace, electronics, medical, and green composite. Although the usage of rapid prototyping is a fast production, there are required studies towards the effectiveness ~~on~~ of using rapid prototyping for green composite production. This is because the natural polymers are vulnerable to oxidative degradation when exposed to the ultraviolet radiation and they were which later may lead to degradation of the mechanical properties. Due to that matter, Jiang et al. (2010) reported the production of green composite containing aspen wood flour by using selective laser sintering (SLS) method. Based on their study, the green composite can be manufactured by using the rapid prototyping method if supported by the presence of additive and coupling agent. The fibre treatment with sodium hydroxide (NaOH) solution also proved in improving the binding between matrix and reinforcement which later may provide better surface roughness and mechanical properties (see Fig. 7.9). Figure 7.10 below shows the part that produced by using SLS method. In conclusion, the green composite can be produced either using conventional hand layup method, or via advanced rapid prototyping. The only matters are the production cost, the production volume, and type of materials used for the products.

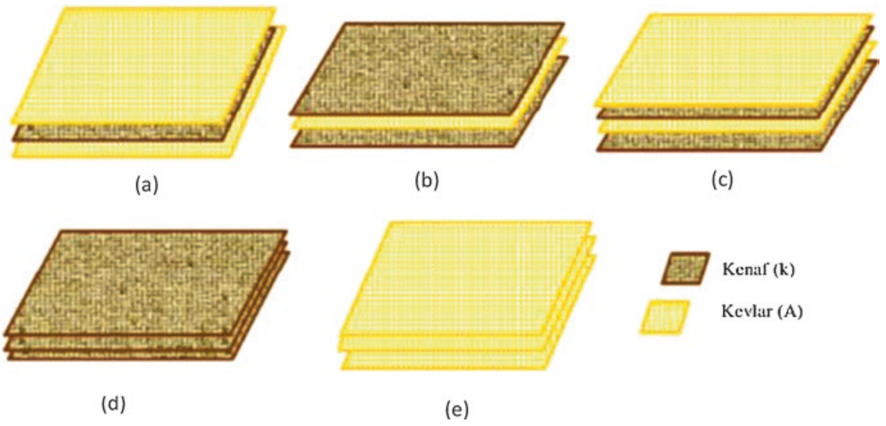


Fig. 7.8 The illustration of the layering sequences of kenaf fibre and Kevlar (Yahaya et al. 2015)

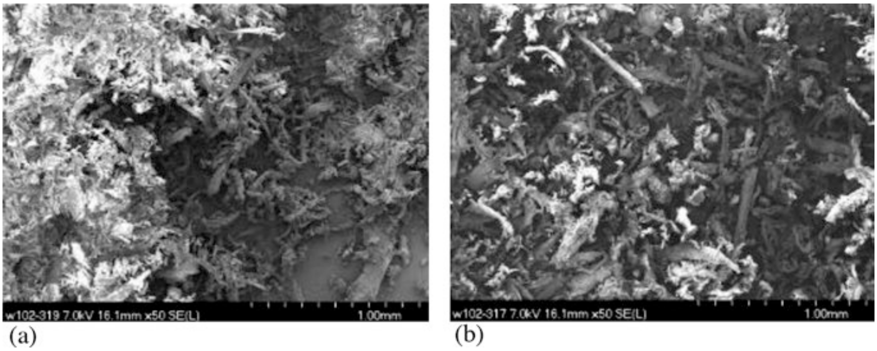


Fig. 7.9 SEM images presenting the comparison of; (**a**) untreated and (**b**) treated aspen wood flour (Jiang et al. 2010)

Fig. 7.10 The part produced by SLS method (Jiang et al. 2010)

7.9 Conclusion

Green composites in overall have shown a rapid growth both in term of research and development, as well as industrial application. At the present moment, its application is focused on low load bearing application, due to its lower mechanical properties as compared to synthetic composites and metals. However, as explained in the previous section within this chapter, many efforts have been carried out to improve the properties of the green composites.

With regards to that, many other opportunities, especially in research, are available to be pursued. Development on the new class of single fibre/matrix and hybrid composites with better material properties can be carried out by increasing the works to discover a new type of natural fibres from unique plant species and polymer matrices, especially biopolymer matrix. The development of more alternative materials could support the goal of creating many fully biodegradable green composites, which will be very beneficial in term of environmental friendly aspect. This will also help to reduce the current problem in managing plastic waste issue around the world.

Furthermore, research opportunity is also vastly opened in enhancing the material properties of natural-based green composites by utilizing the advantages of advanced nanomaterials as the filler to formulate the material. It is shown that various type of nanomaterials significantly improved the performance of the green composites. Further works can be carried out to characterize the nanomaterials effect to as many as possible green composites, as well as to fully understand how the enhancement mechanism is obtained.

It is also briefly described in the previous section how the use of innovative product development and processing method could address the material property limitation in producing an end product with higher load bearing capability. This is also an exciting area to be explored by researchers as well as composite practitioners. Clever modification of the geometrical parameters of product could reduce the maximum stress generated when a load is applied, hence enabling the use of green composites as raw material to construct it without compromising the required structural performance. In addition, the use of advanced rapid manufacturing process such as additive manufacturing should also be incorporated more aggressively to develop a product from green composites. Among the works which could be carried out is enabling the use of fibres with higher aspect ratio, improving the fibre-matrix adhesion properties, as well as creating new filament material using various type of natural fibres and matrices.

In conclusion, the future is bright for green composites, especially made from natural resources to expand its presence in more demanding applications. It is expected based on the accumulating attention and positive progress made so far, green composites from natural resources will take over the domination of synthetic composites as a commercially viable solution in many applications.

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