# Natural fiber reinforced vinyl polymer composites

2

*L.C. Hao[1], S.M. Sapuan[1], M.R. Hassan[1] and R.M. Sheltami[1,2]*
[1]Universiti Putra Malaysia, Serdang, Malaysia, [2]University of Benghazi, Benghazi, Libya

## 2.1 Introduction

The use of natural fiber reinforced polymer (FRP) composites has been the priority in many industrial sectors due to increased awareness of the environmental issues. Over the last few decades, biocomposites have gone through a remarkable evolution. The increasing research every year regarding biocomposites, starting from 32 papers from 1997 dramatically increased to 716 papers in 2015 in a journal paper website (ScienceDirect). Fig. 2.1 shows the statistics of biocomposite related topics in ScienceDirect from the 1997 to 2015. The trend is increasing every year, meaning that the biocomposite research is becoming well known due to environmental issues. Every single researcher is keen to look for the solution to substitute conventional materials that harm our world.

The good flexibility, high stiffness, and low cost of biocomposites make it in a top selection by users. Also, the limited supply of petroleum has made biocomposites even more popular. Therefore, intensive research was conducted to develop biocomposites that are compatible with conventional products (Summerscales et al., 2010; Satyanarayana et al., 2009; Venkateshwaran and Elayaperumal, 2010; John and Thomas, 2008; Shinoj et al., 2011; Mohanty et al., 2005, 2000; Hassan et al., 2010; Bledzki et al., 2002; Pickering, 2008; Thomas and Pothan, 2009). This chapter aims to discuss natural fibers and natural fiber reinforced vinyl polymer composites.

## 2.2 Natural fibers

Automotive, construction, aviation and other sectors are keen to substitute biocomposites for heavy, weak, or expensive material (Pothan et al., 2003). Natural fibers are renewable resources and will substitute all traditional materials in the future. Advantages of natural fibers can be seen in most research journals that regard it. Plenty of researchers mentioned it in their works (Elfehri Borchani et al., 2015; Anuar and Zuraida, 2011; Ku et al., 2011). Table 2.1 has stated properties of some natural and synthetic fibers. Besides the advantages of the properties, the cost of the materials is one of the main concerns. The pineapple leaf and banana leaf fibers are naturally a waste product (Jawaid and Abdul Khalil, 2011). These wastes

**Natural Fiber Reinforced Vinyl Ester and Vinyl Polymer Composites. DOI: https://doi.org/10.1016/B978-0-08-102160-6.00002-0**

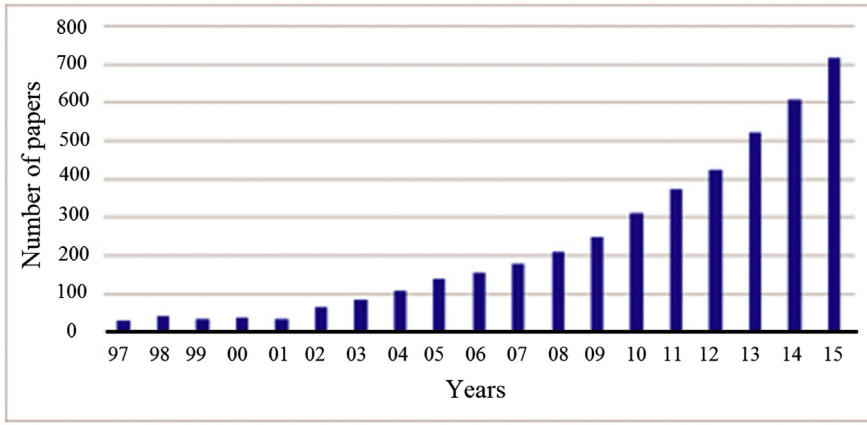


**Figure 2.1** Statistics of biocomposite related topics in the ScienceDirect from 1997 to 2015.

**Table 2.1 The properties of some natural and synthetic fibers**

| Fiber | Density ($g/cm^3$) | Elongation (%) | Tensile Strength (MPa) | Elastic Modulus (GPa) |
|---|---|---|---|---|
| Cotton | 1.5 | 7 | 400 | 12.6 |
| Jute | 1.3 | 1.8 | 773 | 26.5 |
| Flax | 1.5 | 3.2 | 1500 | 27.6 |
| Hemp | 1.47 | 4 | 690 | 70 |
| Kenaf | 1.45 | 1.6 | 930 | 53 |
| Ramie | – | 3.8 | 938 | 128 |
| Sisal | 1.5 | 2.5 | 635 | 22 |
| Coir | 1.2 | 30 | 593 | 6 |
| Softwood kraft pulp | 1.5 | 4.4 | 1000 | 40 |
| E-glass | 2.5 | 0.5 | 3500 | 70 |
| S-glass | 2.5 | 2.8 | 4570 | 86 |
| Aramid | 1.4 | 3.7 | 3150 | 67 |
| Carbon | 1.4 | 1.8 | 4000 | 240 |

*Source:* Ku, H., Wang, H., Pattarachaiyakoop, N., Trada, M., 2011. A review on the tensile properties of natural fiber reinforced polymer composites. Compos. Part B Eng. 42, 856–873. This table is reprinted with permission from Elsevier.

were disposed even before their uses are realized. Therefore, the natural fiber was available for a very low price compared to synthetic fiber (Ho et al., 2012b).

Natural fibers can be derived from plant, animal, and mineral types, which are illustrated in Fig. 2.2. As there is a higher demand for superior material now, surface modification has been applied to enhance the properties of natural fibers so that the modified natural fiber reinforced biopolymer composite is able to perform well in advanced sectors. In the recent years, cellulose fiber undergoes the most

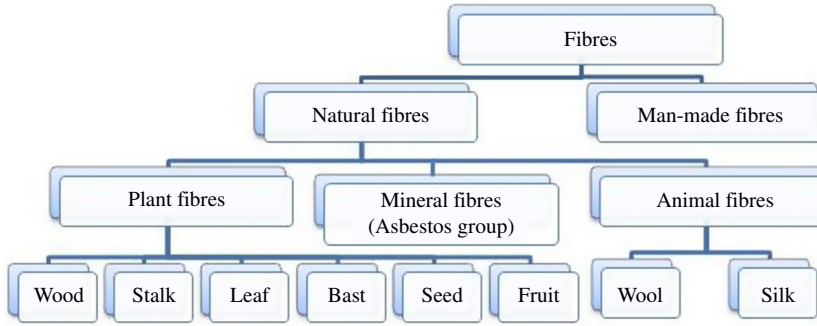


**Figure 2.2** Classification of the fibers.

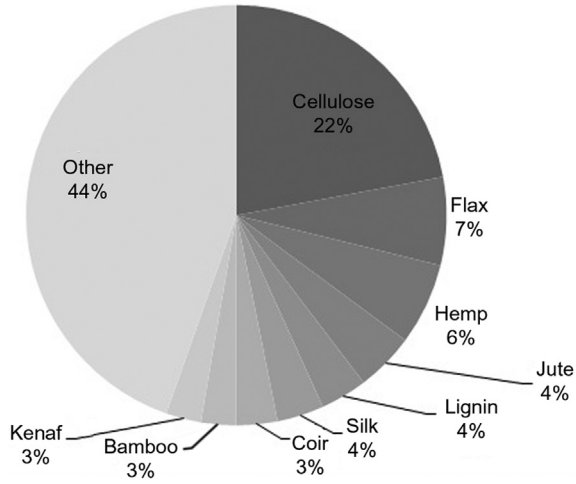


**Figure 2.3** The natural fibers used in biocomposite related topics in ScienceDirect from 2011 to 2015.

intense research in the last 5 years Fig. 2.3 shows the natural fiber that has been used in biocomposite related topics in ScienceDirect from 2011 until 2015. The cellulose fiber consists of about a quarter of total natural fiber used in biocomposite research. The cellulose fiber has outstanding strength properties as it is constructed by pure cellulose.

Regarding environmental issues, biodegradable natural fiber can reduce solid waste yield and handling matter (Ho et al., 2012b). Other than landfill issue, the energy consumption is another point of environmental concern. A lower energy is required to produce the same amount of natural fiber compared to synthetic fiber. It only takes 15 MJ of producing energy for 1 kg of kenaf fiber, while glass fiber consumed 54 MJ (Akil et al., 2011). The natural fiber has lower densities of 1.2−1.6 g/cm$^3$ than 2.4 g/cm$^3$ of glass fiber (Huda et al., 2006). The lower density

of natural fiber has more volume or quantity of fiber for the same weight. This scenario ended up with the natural fiber being fabricated with much lower energy but a higher quantity of fiber. Additionally, natural fiber is nonabrasive to equipment; this helped to prolong the life period of machine tools, and reducing t maintenance cost (Akil et al., 2011). The environmentally friendly production process of t natural fibers offers a good working condition to workers and reduces the risk of respiratory problems compared to intrapleutral fibrous glass that causes chest pain, breathing trouble, sore throat, and cough (Jawaid and Abdul Khalil, 2011; Newball and Brahim, 1976). The natural fiber shows a good damage tolerance and better elongation when the load applied (Jawaid and Abdul Khalil, 2011). Spider silk is an animal based natural fiber that gathered during its web making process (Ho et al., 2012b). The spider silk allows more than 200% of elongation of its original length, and needs about triple kevlar fiber breaking energy to break the silk (Bonino, 2003). In this chapter, the discussion focuses on the natural fibers from the plant.

However, a low interfacial bonding between the natural fiber and polymer has reduced the properties and performance of the composites. The void in the composite has turned itself into a stress concentration point and crack propagation starting point (Milanese et al., 2011). Several methods have been introduced to modify the natural fiber surface to improve the fiber-matrix interfacial such as coupling agent, preimpregnation, and graft copolymerization (Herrera-Franco and Valadez-González, 2004).

Hydrophilic in nature, natural fiber is incompatible with hydrophobic polymers (Akil et al., 2011; Alvarez et al., 2004; Baiardo et al., 2004). Water absorption behavior of the natural fiber has led to fiber swelling at the fiber-matrix interphase (Mehta et al., 2004). This may cause a drop in mechanical properties of the composite (Ku et al., 2011). High water absorption is the main disaster for the natural fiber reinforcement composite.

Expelling the dye compounds in the wastewater into clean water will dissolve oxygen in the water and prohibit the sunray passing through the water (Sajab et al., 2011). Variety of methods can be used in removing dye compounds, e.g., adsorption (Rafatullah et al., 2010), membrane filtration (Amini et al., 2011) or electron-catalytic degradation (Ma et al., 2009). The conventional method shows an effective result yet another form of solid waste has been produced. Therefore researchers have found a cheaper and potential absorbent of natural fiber. High water absorption of natural fiber is a useful feature of dye compounds removal in wastewater (Hassan, 2015).

### 2.2.1 Chemical component of natural fibers

Lignocellulosic fiber is a scientific name that refers to natural fiber, because all plant fibers are constructed by few constituents (cellulose, hemicelluloses, and lignin). Most of plant fibers contain 50%−70% of cellulose as shown in Table 2.2.

The structure of a plant cell wall is shown in Fig. 2.4 and the structure is known as macrofibril. The natural fiber is constructed by millions of macrofibril, while a

**Table 2.2 Chemical content of natural fiber**

| Fiber | Cellulose | Hemicellulose | Lignin | Extract | Ash content | Water soluble |
|---|---|---|---|---|---|---|
| Cotton | 82.7 | 5.7 | – | 6.3 | – | 1.0 |
| Jute | 64.4 | 12.0 | 11.8 | 0.7 | – | 1.1 |
| Flax | 64.1 | 16.7 | 2.0 | 1.5–3.3 | – | 3.9 |
| Ramie | 68.6 | 13.1 | 0.6 | 1.9–2.2 | – | 5.5 |
| Sisal | 65.8 | 120 | 9.9 | 0.8–0.11 | – | 1.2 |
| Oil palm EFB | 65.0 | – | 19.0 | – | 2.0 | – |
| Oil palm Frond | 56.0 | 27.5 | 20.48 | 4.4 | 2.4 | – |
| Abaca | 56–63 | 20–25 | 7–9 | 3.0 | – | 1.4 |
| Hemp | 74.4 | 17.9 | 3.7 | 0.9–1.7 | – | – |
| Kenaf | 53.4 | 33.9 | 21.2 | – | 4.0 | – |
| Coir | 32–43 | 0.15–0.25 | 40–45 | – | – | – |
| Banana | 60–65 | 19 | 5–10 | 4.6 | – | – |
| Sun Hemp | 41–48 | 8.3–13 | 22.7 | – | – | – |
| Bamboo | 73.83 | 12.49 | 10.15 | 3.16 | – | – |
| Hardwood | 31–64 | 25–40 | 14–34 | 0.1–7.7 | $<1$ | – |
| Softwood | 30–60 | 20–30 | 21–37 | 0.2–8.5 | $<1$ | – |

*Source:* Jawaid, M., H.P.S. Abdul Khalil, 2011. Cellulosic/synthetic fibre reinforced polymer hybrid composites: a review. Carbohyd. Polym. 86, 1–18. This table is reprinted with permission from Elsevier.

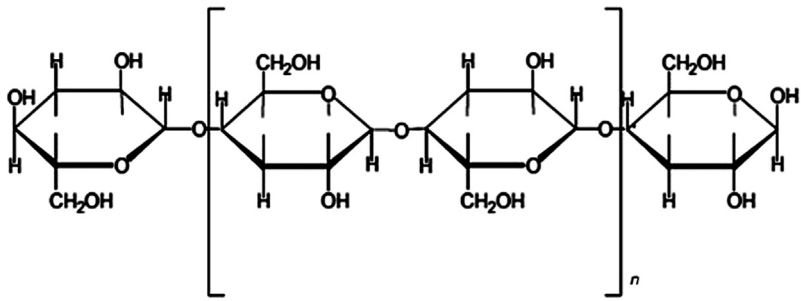


**Figure 2.4** Chemical structure of cellulose.
*Source:* Akil, H.M., Omar, M.F., Mazuki, A.A.M., Safiee, S., Ishak, Z.A.M., Abu bakar, A., 2011. Kenaf fiber reinforced composites: a review. Mater. Design 32, 4107–4121. This figure is reprinted with permission from Elsevier.

macrofibril is framed by microfibril that consists of cellulose, hemicelluloses, and lignin (Ho et al., 2012b). Each of the macrofibrils consists of an outer layer of a primary cell wall and three inner secondary cell walls. Lumen is the open empty spaces located at the center of macrofibril, and it reduces the bulk density of the natural fiber. The lumen was widely deployed as an acoustic and thermal insulator

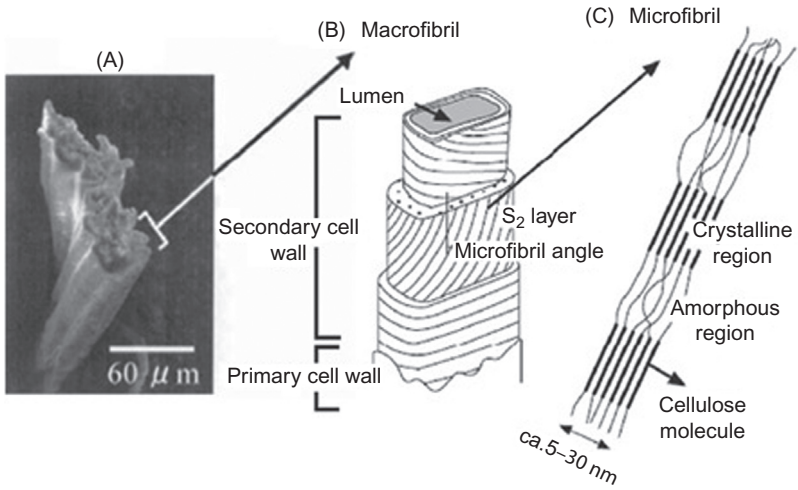


**Figure 2.5** Evolution of natural fiber (A) scanning electron micrograph of kenaf bark fiber, (B) macrofibril (C) microfibril of natural plant.
*Source:* Baillie, C., 2004. Green Composites: Polymer Composites and the Environment, Boca Raton, Woodhead Publishing Ltd. This figure is reprinted with permission from Woodhead Publishing Ltd.

in nature. On the other hand, the microfibril in the secondary cell wall is composed of crystalline cellulose or crystalline hemicellulose and amorphous lignin, arranging alternately with a width of 5−30 nm (Baillie, 2004). Each layer of microfibril has implicated in a designated angle to hold the fiber from every directions.

Cellulose is the main constituent for all the plant fibers (Chawla, 1998). It is composed by C, H, and O elements with a formula of $C_6H_{10}O_5$ (Fig. 2.5). The cellulose influences the major characteristic of the plant fibers. Highly hydrophilic in nature, plant fiber was due to the hydroxyl groups (−OH) that was found in the cellulose chain (Baillie, 2004).

Hemicellulose is one of the most abundant materials after cellulose. Hemicellulose contains a highly branched chain and built by several sugars such as glucose, glucuronic acid, mannose, arabinose, and xylose (Summerscales et al., 2010). Lignin is a highly complex amorphous structure; it acts as a cementing material and fills the spaces between the cellulose and the hemicelluloses (Mohanty et al., 2002; Salit, 2009). Cellulose, hemicellulose, and lignin are bonded together by covalent bonds. The functioning of natural fiber components are shown in Table 2.3.

### 2.2.2 Cellulose fiber

Cellulose is the most abundant component in the world. Every biomass consists of a major part of cellulose. Cellulose is the main source of high performance for plant fibers, the Young's modulus of crystalline cellulose was found higher than Kelvar

**Table 2.3 Chemical content and function of natural plant fibers**

| Chemical content | Polymeric state | Molecular derivatives | Function |
|---|---|---|---|
| Cellulose | Crystalline, highly oriented large molecule | Glucose | Fiber |
| Hemicellulose | Amorphous, smaller molecule | Polysaccharides, galactose, mannose, xylose | Matrix |
| Lignin | Amorphous, large 3-D molecule | Phenyl propane, aromatic | Matrix |
| Extractives | Some polymeric and nonpolymeric | Fat, fatty acid, phenols, terpentes, waxes | Extraneous |

*Source:* Salit, M.S., 2009. Research on Natural Fibre Reinforced Polymer Composites. Universiti Putra Malaysia Press. This table is reprinted with permission from.

and potentially stronger than steel (Lin and Dufresne, 2014). Regardless of its source, cellulose consists of linear homopolysaccharide with β-D-glucopyranose linked with β-1-4-linkages. There are several ways to produce nanocrystalline cellulose (NC), acid hydrolysis (Neto et al., 2013), and microbial hydrolysis (Satyamurthy and Vigneshwaran, 2013). On the other hand, cellulose nanofibers (CNF) can be done by mechanical grinding, high shear homogenization, a combination of mechanical actions with chemical or enzyme hydrolysis. Enzymatic hydrolysis of CNF runs in short time, to avoid further cellulose degradation (Zhu et al., 2011). The source and form of cellulose reinforcement and production method is plotted in Table 2.4.

NC is the most famous reinforcement additive for many applications. Its dispersion state, weight ratio, and phase behavior are important factors on performances. NCs are needle-shaped cellulose particles and less than 100 nm in size (Neto et al., 2013). NC has several terms in literature: NC (Zhang et al., 2012; Maddahy et al., 2012; Cha et al., 2014), cellulose nanowhiskers, cellulose whiskers, crystalline cellulose (Rahman et al., 2014), cellulose crystals, and cellulose nanocrystals (Mtibe et al., 2015; Kumar et al., 2014).

There are two path ways to extract cellulose from lignocellulosic biomass, a chemical pulp treatment solubilize lignin and hemicelluloses, followed by bleaching with oxidizing agents shown in Fig. 2.6. Alkaline treatment was the most famous pathway (de Carvalho Mendes et al., 2015; Rathod et al., 2015). Another extracting treatment was the steam explosion process; better enzymatic hydrolysis resulted. Milled biomass is subjected to a high pressure for a short period of time. The fiber is then exposed to normal pressure after the steam opens. The fibers experience an explosion and the break down of lignin and hemicellulose due to a sudden drop of pressure. Water soluble hemicelluloses can be easily removed by water extraction while other chemical treatments needed to eliminate lignin components. After pulp or steam explosion extraction, controlled sulfuric acid hydrolysis was used to isolate the NC. Acid hydrolysis in the amorphous region is faster than the hydrolysis of crystalline regions. After that, repeated washing with water for dilution to stop

**Table 2.4 Source and form of cellulose reinforcement and production method**

| Sources | Fiber form | Method | References |
|---|---|---|---|
| Kenaf fiber | CNF | Mechanical grinding | Babaee et al. (2015) |
| Canola straw | CNF | Mechanical grinding | Yousefi et al. (2013) |
| Softwood | CNF | High shear homogenization | Zhao et al. (2013) |
| Commercially bleached eucalyptus kraft pulp | CNF | Mechanical actions with chemical or enzyme hydrolysis | Qing et al. (2013) |
| Cotton | NC | Microbial hydrolysis | Nadanathangam and Satyamurthy (2011) |
| Bamboo fiber | NC | Microbial hydrolysis (*Trichoderma reesei*) | Zhang et al. (2012) |
| Sugarcane bagasse | NC | Acid hydrolysis | Kumar et al. (2014) |
| Jute fiber | NC | Acid hydrolysis | Rahman et al. (2014) |
| Sugarcane bagasse | NC | Acid hydrolysis | Zhu et al. (2011) |
| MCC | NC | Acid hydrolysis | Cordero et al. (2015) |
| Bleached softwood pulp | NC | Acid hydrolysis | Wang et al. (2015a) |
| MCC | NC | Acid hydrolysis | Voronova et al. (2015) |
| Corn husk | NC | Acid hydrolysis | de Carvalho Mendes et al. (2015) |
| MCC | NC | Acid hydrolysis | Atef et al. (2014) |
| Green seaweed, *Ulva lactuca* | NC | Acid hydrolysis | Rathod et al. (2015) |
| Grinded cellulose, KimWipes® | NC | Acid hydrolysis | Lalia et al. (2014) |
| MCC | NC | Acid hydrolysis | Voronova et al. (2013) |
| Flax and hemp fiber | NC | Microbial hydrolysis (*Aspergillus oryzae*) | Xu et al. (2013c) |
| Bleached aspen kraft pulp | NC | Acid hydrolysis | Xu et al. (2013a) |
| Cotton cellulose | NC | Acid hydrolysis | Pirani and Hashaikeh (2013) |
| Banana peel | CNF | Chemical and enzyme hydrolysis | Tibolla et al. (2014) |
| Cotton fiber | NC | Microbial hydrolysis (*Trichoderma reesei*) | Satyamurthy et al. (2011) |
| Banana pseudo-stem | CNF | Chemical hydrolysis | Cordeiro et al. (2012) |

(*Continued*)

**Table 2.4 (Continued)**

| Sources | Fiber form | Method | References |
|---|---|---|---|
| Bleached eucalyptus kraft pulp | CNF | Chemical hydrolysis | Tonoli et al. (2012) |
| Bleached eucalyptus kraft pulp | CNF | High shear homogenization | Syverud et al. (2011) |
| Pineapple leaf fiber | CNF | Chemical hydrolysis | Cherian et al. (2011) |

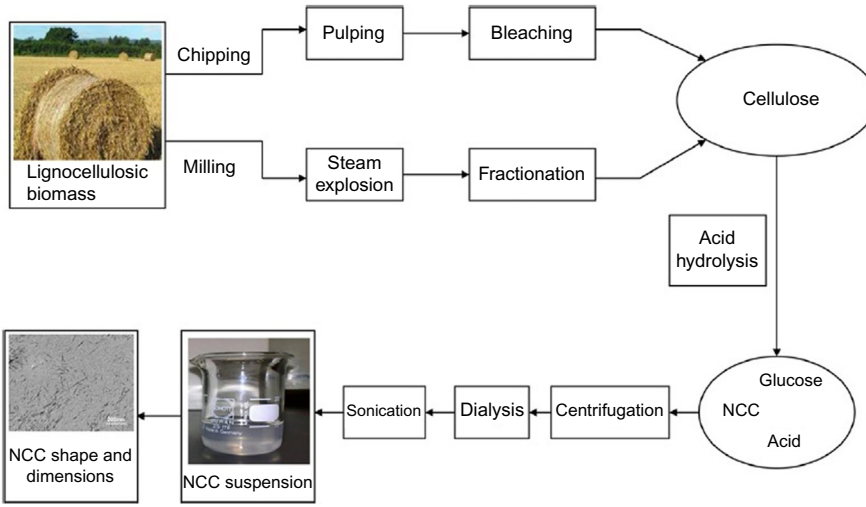


**Figure 2.6** Scheme of NC production steps.
*Source:* Brinchi, L., Cotana, F., Fortunati, E., Kenny, J.M., 2013. Production of nanocrystalline cellulose from lignocellulosic biomass: technology and applications. Carbohyd. Polym. 94, 154–169 (Brinchi et al., 2013). This figure is reprinted with permission from Elsevier.

hydrolysis reaction and remove free acid molecules; a huge amount of water was needed to dilute the product. Mechanical dispersion and sonication are used to disperse agglomerated nanoparticles. Lastly, drying of the products to gain sold NC. Unfortunately, a long production time of NC limited the commercial availability of NC and it once was being accused of environmental issues due to polluted washing water in production. Remaining sulfate groups on the surface of fiber may hydrolyze continues, unexpected drop of properties may happen. These made sulfuric acid hydrolyzed NC unsafe for health care applications. One work has studied the phosphotungstic acid hydrolyzed NC, higher thermal stability, and reusable acid through extraction with diethyl ether were found. This has dramatically reduced environmental pollution done the acid waste (Liu et al., 2014).

Therefore, microbial hydrolysis of NC was proposed under a controlled anaerobic medium. Microcrystalline cellulose (MCC) sole carbon sources, from biomass, are placed in a salt medium with anaerobic gas (10% hydrogen, 10% $CO_2$ and 80% Nitrogen) and shaking condition. The anaerobic microbial medium accelerates the enzyme to produce extra NC as storage for future usage. However, a long period of NC production was short of microbial hydrolysis. The process of hydrolysis took a few days in complete dark, to avoid photosynthesis. It was found that microbial hydrolyzed NC has larger dimension due to lower regions of amorphous needed to be cleaved (Peng et al., 2011). *Trichoderma reesei* is one of the most productive enzymes used in microbial hydrolysis.

Superior properties of cellulose reinforcement have gone through intensified study. Almost all cellulose reinforcement attributed to biomedical materials, since no damage on DNA was found by bacteria CNF, making it suitable for use in the cell system (Hannukainen et al., 2012; Norppa, 2012; Väänänen et al., 2012). A good strength support is a requirement by medical implant and biodegradable progression accompanied by formation of new tissues. Pineapple leaf fiber nanocellulose-polyurethane blood valve was reported biostable in 6 months, resistant to fatigue and hemodynamics. Pineapple leaf hydrolyzed nanocellulose composite exhibited good elasticity, with reasonable strength performance (Cherian et al., 2011).

CNF aqueous suspensions can form hydrogels to provide a suitable environment to support cell growth by controlling CNF contents without human borne components. CNF hydrogels helps to differentiate the human hepatic cell (HepG2 and HepaRG) as well as enhance the spheroid formation of the cells. At a high shear stress situation, the aqueous CNF has small viscosity to support the injectability, while the material is converted into an elastic gel at a low shear stress situation. These findings have allowed the drug and chemical testing on this artificial CNF hydrogel (Bhattacharya et al., 2012).

Bone regeneration therapy by using nanocellulose biocomposites had been proven to be effective and practically workable. However, only a few publications reported the animal experiments. Microbial hydrolyzed nanocellulose biocomposites were the most famous material on bone regeneration study (Tazi et al., 2012; Saska et al., 2011; Fan et al., 2013). A wound infection caused by high bacterial levels, is a highly dangerous scenario in the medical field. Nanocellulose can provide a porous network structure in the biocomposites, to accelerate the antibiotics or medicines into the wound (Andresen et al., 2007). This significantly reduces the chances of a wound infection. Silver (Ag) has been studied most extensively as antimicrobial agents cooperated with nanocellulose biocomposites (Rai et al., 2009). The antimicrobial effect of the Ag depends on the particle size and shape; dendritic Ag showed better performance than the sphere Ag fillers (Xiong et al., 2013).

### 2.2.3 *Flax (*Linum usitatissimum *L.) fiber*

Flax (*Linum usitatissimum*) is one of the bast fiber and one of the first textile material. Flax textile applications were traced back to 5000 BC in Egypt (Dewilde, 1983). Canada is the main producer of flax from 1994 and it exports more than

90% of flax to Europe, United States, and Japan. Fast growth rate of flax enables it to be harvested in about 3 months from March to July (Baiardo et al., 2004). The fine and long flax fiber are suitable for high quality textiles while short flax fiber is suitable for canvas, towel. Lower fiber grades were utilized as fiber reinforcement in composites to enhance the properties of the composites.

An experiment investigates the strength of the flax composites by the influences of different fiber direction and stacking sequences (0, 90, ±45, 0/90) (Liang et al., 2015). The compression strength of the glass fiber composites was 76% greater, mainly because the flax fiber pull-out led to the damage on the composite.

Maleic anhydride graft copolymer is a very effective additive for natural fiber composites (Karmaker and Youngquist, 1996; Sanadi et al., 1995; Felix and Gatenholm, 1991; Chuai et al., 2001; Olsen, 1991). The maleic anhydride polymer helps to reduce the hydrogen bonding on fiber that tends to attract fiber together, avoiding fiber agglomeration (Kazayawoko et al., 1997). Besides, the covalent bonding between the hydroxyl group of the cellulose and anhydride group is one reason for strength improvement (Felix and Gatenholm, 1991; Matias et al., 2000; Cantero et al., 2003), resulting in better interfacial properties and load transfer. One research studying the influential of several types of maleic anhydride-polypropylene copolymer (MAPP) for short flax fiber reinforced polypropylene (PP) composites (Arbelaiz et al., 2005) revealed that the 5% of E43 or 10% of G3003 MAPP has the best mechanical properties. The molecular weight difference between two copolymers leads to a different degree of entanglements. Furthermore, the recycled MAPP treated flax fiber reinforced PP composites have showed a slight change of properties prompt that flax fiber is environmentally friendly to recycle.

A flax fiber composite application has been proposed in orthopedics; a hybrid epoxy polymer composite reinforced with ordinary carbon fiber and flax fiber as a potential orthopedic fracture plate (Bagheri et al., 2015). High water absorption behavior of flax/epoxy composite may cause additional cell death. This hybrid composite shows no negative effect as medical grade stainless steel. Besides, the same hybrid carbon/flax/epoxy composite material has been applied on bone plate application (Bagheri et al., 2013). Yet this bone plate is made of sandwich structure that thin carbon fiber reinforced epoxy sheets were bound to the outer surface of flax fiber reinforced epoxy core rather than homogenous mixing. Compared to the orthopedic metal plate, this new hybrid composite is more close to human cortical bone.

Another study has investigated the rheological properties treated flax/cement mixture for the construction field. The four type of fiber treatments are Rheomac Deco Oleo, hydrothermal treatment, matrix precoated (Sawsen et al., 2015), and alkaline treatment (Sawsen et al., 2014). The first treatment is commercial water repellent to lower the water absorption of flax fiber. The second method is to clean the fiber with boiling water in order to release extractives and the precoated method treated with matrix to decrease the moisture content inside fiber as well as to enhance the interface with matrix. The alkaline treatment of the flax fiber was to aim to remove the noncellulosic polysaccharide compounds, and increase active react sites for better interfacial with matrix. As a result, a commercial water

repellent product and precoated matrix are helped to reduce water saturation rate on fiber. Besides, the flexural and compressive strength have been enhanced. On the other hand, the water boiling method does not influence the water saturation rate nor the strength of flax fiber. 10 wt% of the flax fiber reinforced cement has greatly reduced the density of cement due to the air bubbles and light weight flax fiber (Aamr-Daya et al., 2008). The air bubbles and flax fiber serve as sound insulation. This may deteriorate the strength of the composite, but still above the basic requirement of construction materials.

Flax fiber reinforced epoxy composite is also being applied in the automotive sector for sound absorption and vibration damping (Prabhakaran et al., 2014). It is possible to create a superior acoustic and vibration damping performance without losing its light weight. The flax fiber reinforced composite has enhanced 20% more sound absorption ability and an extra 51% vibration damping. The lumen in natural fiber is the reason for a better sound proof characteristic.

### 2.2.4 *Hemp (*Cannabis sativa *L.) fiber*

Hemp (*Cannabis sativa L*) is one of the most famous annual crops that is grown for its long and strong bast fiber and seed for more than 12,000 years. Hemp grows in a very wide range of climates, ranging from Western to Asia. In western countries, the cultivation has been interrupted for decades due to the synthetic fiber competition and high labor cost. In 2011, hemp was cultivated on 61, 318 ha in the world (Salentijn et al., 2015). Hemp fiber can be divided into a bast and core section. The primary bast fiber is 20−50 mm long while secondary bast fiber only has 2 mm in length. The bast hemp fiber is made up of thick and lignified cell walls of cellulose (55%−77%), hemicellulose (2%−22%), pectin (0.8%−18%), and lignin (2.9%−13%) (Bismarck et al., 2006; Baltazar-y-Jimenez and Bismarck, 2007; Garcia-Jaldon et al., 1998; Jarman, 1998; Wang et al., 2007; Kostic et al., 2008; Gassan and Bledzki, 1996; Bolton, 1995; Mougin, 2006; Kozlowski and Wladyka-Przybylak, 2004). Hemp fiber is a cheap, abundant and renewable resource, having outstanding tensile strength; and it is suitable to reinforce in composites (Lu and Oza, 2013; Rouison et al., 2006; Shubhra et al., 2010; Etaati et al., 2014, 2013; Niu et al., 2011, Song et al., 2012; Aabdul Khalil et al., 2012a; Shahzad, 2012).

Maleic anhydride graft copolymer is a very effective additive for natural fiber composites. The covalent bonding between the hydroxyl group of cellulose and the anhydride group is one reason for strength improvement, resulting in better interfacial properties and load transfer. High density polyethylene (HDPE) was blended with the hemp short fiber with coupling agent maleic anhydride-grafted polyethylene (MAPE) (Wang et al., 2014c). The results have proved that the presence of 2 wt% has MAPE enhanced the strength as well as higher flame resistivity. MAPE improves the fiber-matrix adhesion which decreases the cracks observation on the composites. Another finding for short hemp fiber reinforced PP composite has shown good enhancing static and dynamic mechanical properties of composite, doubling its strength and stiffness brought by coupling agent (agent maleic anhydride-grafted PP and silane coupling agent) (Panaitescu et al., 2015). The

enhanced properties of the hemp-based composite have prompted to substitute the use of synthetic glass fibers.

The biodegradable natural fiber has no relation with the issue of recycling. However, the natural fiber reinforced conventional synthetic polymer composites have a great interest on this topic as it is not fully decomposed. A recycled PP/hemp fiber composite has maintained its properties due to the stable fiber aspect ratio (Bourmaud et al., 2011). The increased impact energy at failure for recycled composite was due to the lower viscosity; this has been made a favorite by the automotive industry for ease fabrication.

A concrete block was made from hemp-lime composites to study its performance (Arnaud and Gourlay, 2012). The high porosity of the sample (80 wt% hemp fiber) presented good thermal and acoustic properties. However, the hemp hurd in hydrophilic nature absorbs large quantities of water, making it fail in conventional mixing since the hemp fiber absorbed a large amount of water, resulting in long setting and drying times. Unfortunately, the properties of concrete composite were depending on its density. Higher density increases all kinds of properties (hardness, bending strength, compression strength, Yong's modulus, and thermal conductivity) (Elfordy et al., 2008). Therefore, the user has to locate a balance point between mechanical properties and thermal insulation from the type of application. If prebuilt structure is needed, low thermal conductivity could be used. On the other hand, denser blocks should be used for structural integrity of the construction.

A study aimed to substitute steel helicopter Eurocopter AS 350 Écureuil rotorcraft interiors with hemp fiber reinforced epoxy composites (Scarponi and Messano, 2015). The lower fuel consumption was expected for lighter weight and lower pollution for green material. The outcomes have showed more than 55% of weight reduction on the substitution parts without significantly increasing the cost. However, the durability and fire behavior of this material require more studies before it is apply on the helicopter.

### 2.2.5 Jute

Jute has been long found in India on family farms based. It was twisted into twine and rope. Jute fiber is known as the golden fiber because of its long and golden brown color. It is widely used to make sack and garden twine. The jute plant can grow easily with a little fertilizer. It can grow about 3.5 meters tall with a stem diameter of 20 mm. It is an annual crop with a very high photosynthesis rate; this helps to convert carbon dioxide into oxygen. A hectare of the jute plants could absorb about 15 tons of carbon dioxide and produce 11 tons of oxygen. Additionally, the plant enriches the soil as used in crop rotation, the leftover on soil maintaining soil fertility. Some research on the jute fiber has been done for reinforced polymer composites (Gopinath et al., 2014; Pantamanatsopa et al., 2014; Yallew et al., 2014; Dong et al., 2014; Kikuchi et al., 2014; Arao et al., 2015).

The jute fiber reinforced composite has been widely chosen to compound as a sound barrier material or fire barrier (Fatima and Mohanty, 2011). 5 wt% of the jute fiber content reinforced latex composite has found more than half smoke

emission reduction and shows the best limiting oxygen index (30.2). The highly flame propagation behavior of the jute/latex composite has been lowered down by 1 wt% of sodium phosphate fire retardant. In short, the jute fiber reinforced latex composite with fire retardant filler is potentially used to substitute conventional materials in a high temperature working surrounding.

Alves et al. (2010) has discussed the environmental factors to fabricate a frontal bonnet for an off-road vehicle. The topic has been surveyed including availability of materials (jute fibers and glass fibers), human safety, and ecosystem equality in several stages (production, use, and disposal phase). This case study has demonstrated that jute fiber was the best choice to enhance the environmental image of the whole vehicle, and lower fuel consumption due to the light weight of jute fiber. Human safety on the production was mentioned in discussion as well as increasing the tool life by using jute fiber composite. Jute fiber composites are also able to prevent the potential environmental pollution for vehicle disposal due to its biodegradable characteristic.

### 2.2.6 Lignin based carbon fiber

In recent times, carbon fiber was produced by petroleum based Poly-Acrylic-Nitrile (PAN) feedstock by solution spinning (Baker et al., 2012). The global shortage of petroleum based precursors, environmental pollution, and most importantly the high production cost of the carbon fibers, have restricted carbon fiber in further

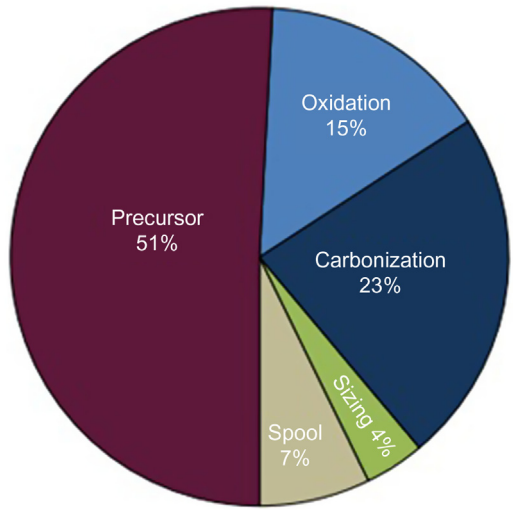


**Figure 2.7** The cost of carbon fibers production in each step.
*Source:* From Mainka, H., Taeger, O., Stoll, O., Körner, E., Herrmann, A., 2013. Alternative precursors for sustainable and cost-effective carbon fibers usable within the automotive industry. Society of Plastics Engineers (Automobile Division)–Automotive Composites Conference & Exhibition, 2013. Previously Published in the Proceedings of the 13th-Annual SPE ACCE, Novi, MI, September 11–13, 2013. Reprinted with permission.

development. Fig. 2.7 has stated the cost of carbon fiber production in each step, more than 50% of the cost accounted for the precursor PAN production. Therefore, the alternative precursor, lignin, with the significant low production cost has been introduced. Lignin is the most suitable option as it has a high carbon content and it is found in most of the plant at 15%−40% of dry weight (Ragauskas et al., 2014; Zhou et al., 2014). Lignin is a waste product of the paper mill, hence it is cheap and available in mass (Mainka et al., 2015).

Lignin can be categorized into three types: hardwood lignin, softwood lignin, and grass lignin. Hardwood lignin is preferred as carbon fibers' precursor. Two extra fabrication steps have added into lignin based carbon fiber productions. These steps are washing and drying the lignin powder followed by pelletizing. Hydrophilic nature of lignin was not suitable for fiber spinning, the moisture making it bad in flow ability. Pelletizing of lignin allowed it process in melt spinning and lower moisture content (Mainka et al., 2015).

Phenolic resin was one of the famous carbon precursors other than PAN. High microporosity of carbonized fibers and high carbon yield are a feature phenolic resin precursor. A bamboo lignin-phenol-formaldehyde (LPF) resin has been used as a carbon fiber precursor (Guo et al., 2015). The LPF-derived carbon fiber shows a homogenous diameter distribution and long fibrous morphology. The fiber exhibits good dimensional and thermal stability performance.

A study has add the PAN and cellulose nanofibrils into lignin based carbon fiber to increase its electrospinability and produce a porous core, respectively (Xu et al., 2013b). Fig. 2.8 shows the SEM micrograph of lignin-cellulose nanofibrils-PAN

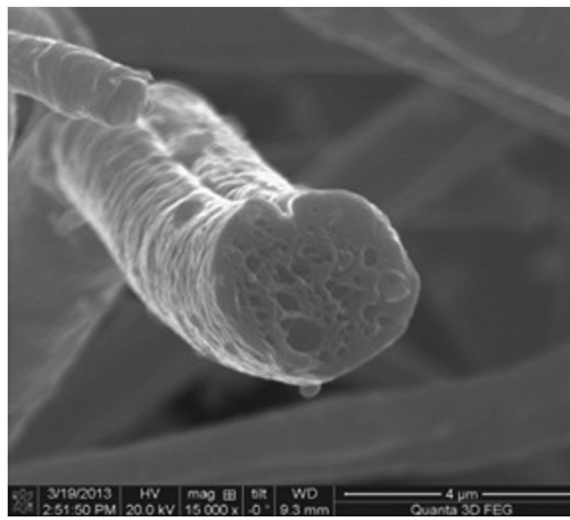


**Figure 2.8** The SEM micrograph of lignin-cellulose nanofibrils-PAN core−shell fiber. *Source:* Xu, X., Zhou, J., Jiang, L., Lubineau, G., Chen, Y., Wu, X.-F., et al. 2013b. Porous core-shell carbon fibers derived from lignin and cellulose nanofibrils. Mater. Letters, 109, 175-178. This figure is reprinted with permission from Elsevier.

core−shell fiber. The porous core fibers provide a much larger surface area and porosity than solid fibers for the similar diameter. A larger surface area provides better interactions between fibers and surroundings, therefore better performances.

Carbon physical properties are varied widely with the allotropic form. A lignin derived carbon has been studied in the electrochemical energy storage field, such as lithium ion batteries. The lignin derived carbon obtained shows excellent lithium storage capacity and perfect rate capability due to its unique hierarchical porous structure (Zhang et al., 2015d). The galvanostatic charge-discharge testing has been conducted at 200 and 500 mA/g current density. Both densities show lignin derived carbon having a stable cycling performance. After 5000 charge/discharge cycle of the lignin derived carbon, over 96% of capacitance retentions remained, demonstrating potential of lignin derived carbon on electrochemical energy storage sector (Hu et al., 2014).

Porosity of the lignin derived carbon fibers has attributed to better interactions, higher energy storage capability, better thermal stability, and lower cost. The lignin derived carbon fiber blended in polylactic acid (PLA) has shown deteriorated strength when increasing the PLA content (Wang et al., 2015d). The previous work suggested that microvoids found on the fiber surface due to the volatilization of PLA. The voids act as a stress concentration point and break the fiber before applying the maximum loading.

### 2.2.7 Silk fiber

Silk fiber is a natural protein fiber. It could come from a moth or spider. Most of the commercial silk fiber was made by *Bombyx mori* moth silk. *B. mori* moth silk fiber is the most famous silk for high strength properties. Nangnoi Srisaket (Somvipart et al., 2013), *Hysperophyiax occidentalis* (Wang et al., 2014a), *Antheraea pernyi* (Du et al., 2015; Wang et al., 2015c), *Antheraea yamamai* (Numata et al., 2015), *P. ricini* (Panda et al., 2015) and *Antheraea mylitta* (Bhattacharjee et al., 2015) silk fiber were studied in previous work. *B. mori* silk fiber is extracted from a cocoon which is mainly composed by fibroin and sericin binder. The silk fibroin is a natural fibroin protein with semicrystalline structure, providing fiber stiffness and strength. The sericin binder acts as an adhesive binder to hold the structure of the fiber. The silk fiber has been chosen as a reinforcement in biopolymer recently, especially in tissue engineering and medical industries (Eshkoor et al., 2013a,b; Ataollahi et al., 2012; Ude et al., 2013a,b; Chen et al., 2012a,b).

Another type of the silk fiber was produced by a spider. Six types of different silk fiber can be produced by a single spider: major ampullate silk, minor ampullate silk, flagelliform silk (capture core threads), aciniform silk, tubuliform silk (outer eggsac), and piriform silk (Lewis, 2006). Fig. 2.9 has showed the spider silk glands, silk types, and its uses.

Wool and silk fibers differ in many perspectives. Wool fiber is formed by keratin protein and growth from an outer skin layer like from a sheep or goat. Silk fiber is a protein fiber from silk glands of an insect.

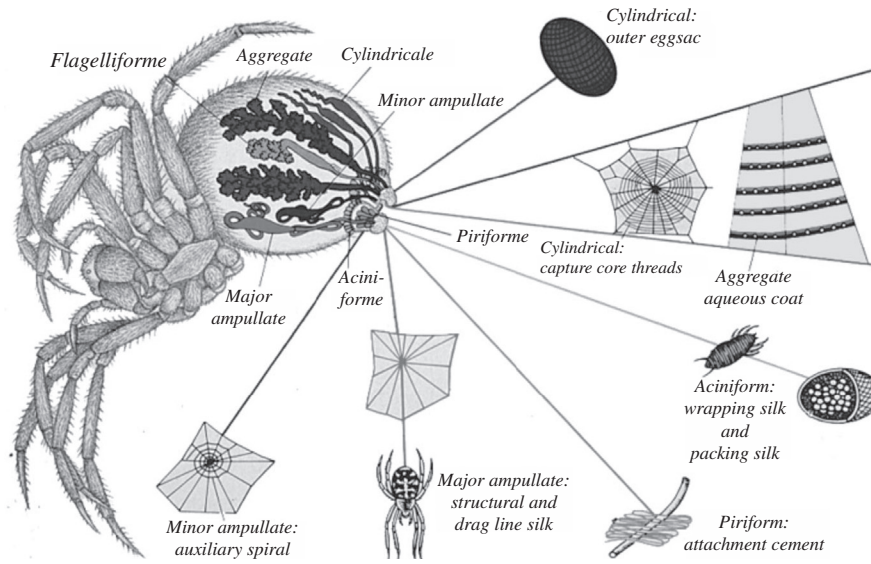


**Figure 2.9** The spider silk glands, silk types, and uses.
*Source:* Tokareva, O., Jacobsen, M., Buehler, M., Wong, J., Kaplan, D.L., 2014. Structure−function−property−design interplay in biopolymers: spider silk. Acta Biomater. 10, 1612−1626 Tokareva et al., 2014. This figure is reprinted with permission from Acta Materialia Inc.

Cultivation of silkworm is known as sericulture. A female silk moth produces about 300−400 eggs at once. The female died immediately after producing the eggs while the male moth died a short period after that. The eggs took about 10 days to become larvae. The larvae then consume a lot of mulberry leaves until it grows mature under optimum temperature and food quality. This feeding period takes about 6 weeks. The silkworm starts to contract its body and extrude the silk composed by the fibroin protein and coated by gum-like protein sericin from its mouth. A hammock of silk spins out first to support the structure of cocoon. The silk cocoon then spins out and covers itself completely. Several grams of cocoon is made by a single continuous silk in length of 700−1500 m. The larva now transforming itself into pupa by developing a hard skin, finally grows up into an adult moth. The ideal growth spot of silk can be a corner, a box, or a bunch of twigs. Fig. 2.10 shows the life cycle of *B.* mori silkworm.

Degumming treatment is a treatment to remove the sericin coating in the cocoon structure before it can reel several silks together into a single thread (Ho et al., 2012a; Wang et al., 2015b) and is followed by dry-spinning (Yue et al., 2014) or wet-spinning (Kim and Um, 2014; Zhang et al., 2015c) or electrospun (Yoon et al., 2013; Solanas et al., 2014; Ko et al., 2013) to extract the fiber.

Due to its outstanding strength, intensified studies were done on it. The harvesting method has influenced the final performance of the silk fiber. An exciting study

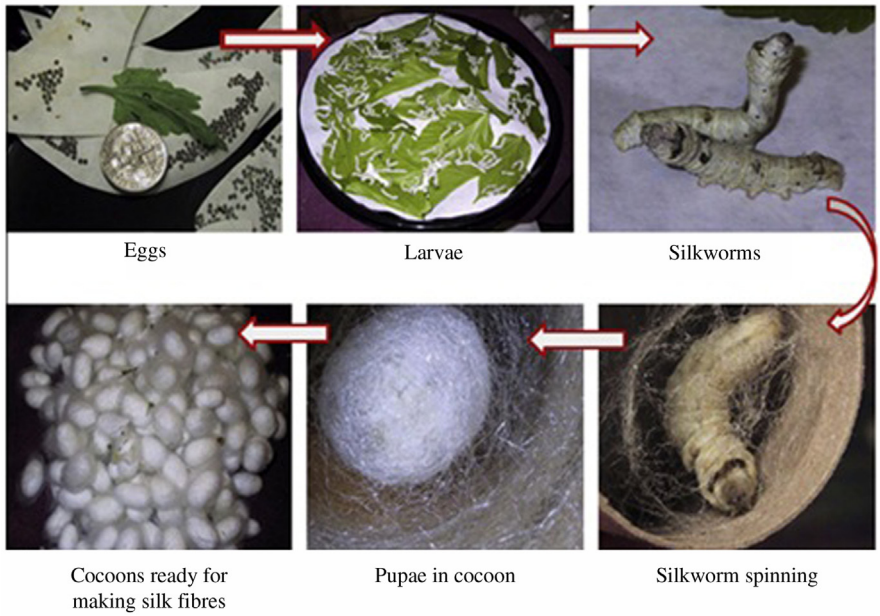


**Figure 2.10** Life cycle of *Bombyx mori* silkworm.
*Source:* Ude, A., Eshkoor, R., Zulkifili, R., Ariffin, A., Dzuraidah, A., Azhari, C., 2014. *Bombyx mori* silk fibre and its composite: a review of contemporary developments. Mater. Design, 57, 298−305 (Ude et al., 2014). This figure is reprinted with permission from Elsevier.

has examined the properties of cultivated *B. mori* silk fiber by feeding the silkworm mulberry leaves with nanomagnetic powder. The magnetic properties in natural, superior strength and thermal properties were the result of this feeding. The successful results allowed us to modify the silk fiber by feeding a specific component to obtain a desired performance of the fibers (Wang et al., 2014b). The strength of fine silk fiber was reported surprisingly high but elongation at break has no effect with the fiber diameter (Tsukada et al., 1996). Meth-acrylamide treatment on the fiber has increased the Young's modulus value with lower elongation at break (Kawahara et al., 1996). One study even showed the *B. mori* silk fiber has better strength than glass fiber (Pérez-Rigueiro et al., 1998).

Almost all silk fiber researches in recent were attributed to biomedical materials. Biocompatible on human tissues with good strength of silk fiber has been promoted in a drug delivery system (Wenk et al., 2011; Pritchard and Kaplan, 2011; Mwangi et al., 2015; Mottaghitalab et al., 2015), scaffolds for tissues (Melke et al., 2016; Teimouri et al., 2015; Zhang et al., 2015a) and wound healing (Patil et al., 2015).

To obtain metalize silk fabric for biomedical material, tris (2-carboxyethyl) phosphine (TCEP) was applied on silk fiber. A smooth iron metal layer has deposited on the fiber surface showing a good electrical conductivity and antibacterial properties (Yu et al., 2015). The antibacterial ability has claimed to be responsible

by the amount of silver metal layer on the silk fiber surface (Meng et al., 2016; Calamak et al., 2015). The silver metal particles were employed to increase antibacterial activities of silk fibers against Gram-negative and Gram-positive microorganisms (Amato et al., 2011).

When developing products for drug delivery, nature size of the raw product needed to be considered for different drug releasing profile. Milled silk particles has a high surface area, able to reached equilibrium loading in a short time (10 minutes) while silk fiber need 3 days to reach equilibrium when delivering the drug. This shows the silk fiber was releasing the drug faster than silk particles, while the smaller the particle size, the lesser the amount of drug released (Kazemimostaghim et al., 2015). Bhardwaj et al. (2015) has milled several types of nonmulberry silk fiber into particle size for a long term drug releasing system.

To apply the silk fiber on the application, integrity of the product cannot be ignored especially in biomedical. Biopolymers are the best option in terms of biocompatibility with silk fiber. By controlling the portion of silk fiber and poly(3-hydroxybutyric-acid-*co*-3-hydroxyvaleric-acid) (PHBV),the rate of composite degradation was tuneable. Higher silk fiber content induced a rapid degradation whereas sustain period for larger matrix content (Miroiu et al., 2015).

Polypyrrole (PPy) has been extensively applied on tissue scaffold due to its good conductivity. However, its brittleness and nonbiodegradable behavior made it away from nerve regeneration application. A hybrid polymer (PPy and poly($_{L}$-lactic) acid) reinforced with spider silk fiber composite was fabricated for good biocompatibility and cell adhesion with stable conductivity (Zhang et al., 2015b). The degradation rate and the spinnability properties were controlled in the silk fiber reinforced hybrid polymer composite.

Other than medical sector, silk fiber was recruited in automotive applications due to its superior impact resistance (Oshkovr et al., 2012, 2013; Ataollahi et al., 2012). The silk fiber reinforced in epoxy polymer composites in 12, 24, and 30 layers of lamination were tested under quasistatic compression testing; crash load and energy absorption behavior were studied. The energy absorption capability varies with its fiber length and the layer of laminations in composite. The higher energy absorbed the safer of the passenger in the event of a crash.

Seawater contains thousands of species of organisms. *B. mori* silk fiber was used in a study to replace the glass fiber for erodible antifouling marine coating. The surface treated silk fiber has better compatibility, resulting in a promising potential material in marine applications as an antifouling agent (Buga et al., 2015).

### 2.2.8 Coir fiber

Coir fiber is the natural fiber extracted from the husk of the coconut. The coir fiber is the thickest and most resistant of all commercial natural fibers. Low decomposition rate is the key advantage for making durable products. Early century ropes made from coir fiber have been discovered. The coir fiber's high strength is the main reason for the rope production for centuries. There are generally two types of coir fiber the brown fiber from mature coconuts and the finer white fiber from

immature green coconuts after soaking up to 10 months. Coir fiber is one of the most lignin-rich natural fibers (Gu, 2009).

Tons of coir residues are produced when producing the coir fibers. Especially the shells were fully utilized as a combustion source fuel, a potential alternative fuel for diesel engine or fertilizer (Wever et al., 2012; Tiryaki et al., 2014). Recent investigation on the coconut shell as reinforcement in polymer matrix has found excellent results (Essabir et al., 2014). Therefore, the fruit shell can be used as another form of reinforcement in composites sector.

Coconut coir fiber cement mortar has been made in Thailand and used as a roof sheet to reduce the heat transfer and energy conservation. In that study, a researcher has mentioned that natural fiber based composite building materials are more suitable for hot and humid weather regions like Thailand. It's believed that the hydrophilic nature of natural fiber has a lower duration in a high humidity surrounding.

Coir fiber treated with 2% of alkali was used to reinforce polyester composites; the results have shown better tensile strength and reduction of the strength beyond 2% of sodium hydroxide (NaOH) concentration (Rout et al., 2001). The enhancement in mechanical properties is attributed to the improved wetting of alkali treated coir with polyester by removing the hemicelluloses and lignin component (Arrakhiz et al., 2012). However, one alkaline pretreatment on brown coir fibers has reported poor results compared to normal coir fibers (Gu, 2009). The lower tensile strength of the composite was found for an increment of concentration of NaOH for alkaline treatment. The intension of improving adhesive ability of the fiber with the matrix by alkaline treatment had been overcome by the strength loss of the fiber. Other than a strength properties investigation, a fiber shrinkage phenomenon has been studied by Rahmanand and Khan (2007). The results show 20% of alkaline treated coir fibers having the maximum shrinkage and weight losses. This is because high NaOH concentrations take a large amount of water in the crystal structure and hence fiber become swollen. When the water has eliminated, structural shrinkage and weight losses were found.

The effect of lignin content on composite properties was studied. The half of lignin content in coir fiber has been eliminated by sodium chlorite (Muensri et al., 2011). The lignin removal has no significant effect on mechanical properties, but slightly reduces the water absorption of the samples. This was suggested by a researcher that the remaining lignin content is still sufficient to cover the fiber surface, showing that the excess lignin content has no effect on composite properties.

Mechanical properties of coir FRP matrix composite have intensively been studied. The strength of the coir fiber tends to decrease with the amount of fiber in the polyester matrix. This showed that the random arrangement of coir fiber in the matrix are not improving the composite's strength (Monteiro et al., 2008). Glass fiber reinforced plastic has been widely used as a comparison for natural FRP composites. Poor interfacial strength between the coir fiber and matrix was the main issue of lower strength properties (Harish et al., 2009). Poor bonding structure of composite encourages the crack propagation and void formation.

Natural fiber is a good low cost adsorbent substitute; coir fiber is one of them. The results have shown a good adsorption capacity toward methylene blue (Etim et al., 2016). The fiber was also chemically modified to remove the heavy metal ions like Ni(II), Zn(II), and Fe(II) (Shukla et al., 2006). The oxidized coir fibers possess better efficiency in adsorbing metal ions and could be regenerated using alkali and reused with maximum efficiency for at least up to three times.

### 2.2.9 Bamboo fiber

Bamboo belongs to a grass family known as Bambusoideae. The high photosynthesis ability, high growth rate, low density, and low cost made it stand out from other plant fibers (Ray et al., 2004; Osorio et al., 2011; Thwe and Liao, 2003; Riaño et al., 2002). There are more than a thousand species of bamboo in its family. Asia and South America are major bamboo growing locations (Aabdul Khalil et al., 2012b; Gratani et al., 2008). Bamboo culm is constructed by a hollow cylinder, while internodes are the place where branches grow. Every section of the bamboo fiber has different properties. Different types of bamboo fiber give different status of properties due to difference of length, diameter, constituent composition, and lumen size. Fig. 2.11 shows different types of bamboo fiber with its fiber length, fiber diameter, and lumen diameter. Some of the fibers have large fiber diameter but small lumen and vice versa. The chemical constituents of bamboo fiber have been reported as 73.83% of cellulose, 12.49% of hemicellulose, 10.15% of lignin, 0.37% of pectin, and 3.16% of aqueous extract (Table 2.2). Every constituent in the bamboo fiber plays the same role as general natural fiber. Fiber at the plant base has lower strength with smaller density but better force resistance (Lo et al., 2004). Nigeria has introduced the bamboo reinforcement in the construction sector (Atanda, 2015). High properties of the bamboo composite are comparable to mild steel for use in construction (Alade et al., 2004). Scaffolding, mud house based, and roofing were reported that constructed by bamboo, having a high strength/weight ratio (Oyejobi and Jimoh, 2009).

Bamboo fiber consists of many vascular bundles that provide strength to the bamboo plant. Vascular bundles are shown in Fig. 2.12. Xylem and phloem found in vascular bundles were meant to transfer water, nutrients, and sugars to the whole bamboo plant (Ray et al., 2004). High water absorption is a common disadvantage of natural fiber, and also a disadvantage of bamboo fiber. The hydrophilic nature of the bamboo fiber causes a poor interfacial bonding between the hydrophobic polymer and fiber. This has decreased the tensile strength and modulus by 37% and 48% respectively after the bamboo fiber has immersed in water for 6 days (Godbole and Lakkad, 1986). Alkali treatment and steam explosion treatment on bamboo fiber had reported a better interfacial bonding and lower water absorption (Phong et al., 2011). Another biochemical treatment has been studied by Saikia et al. (2015), to enhance the tensile strength and prevent the sample degradation. This treatment helped improve the bamboo fiber properties, while maintaining the ecology and environment.

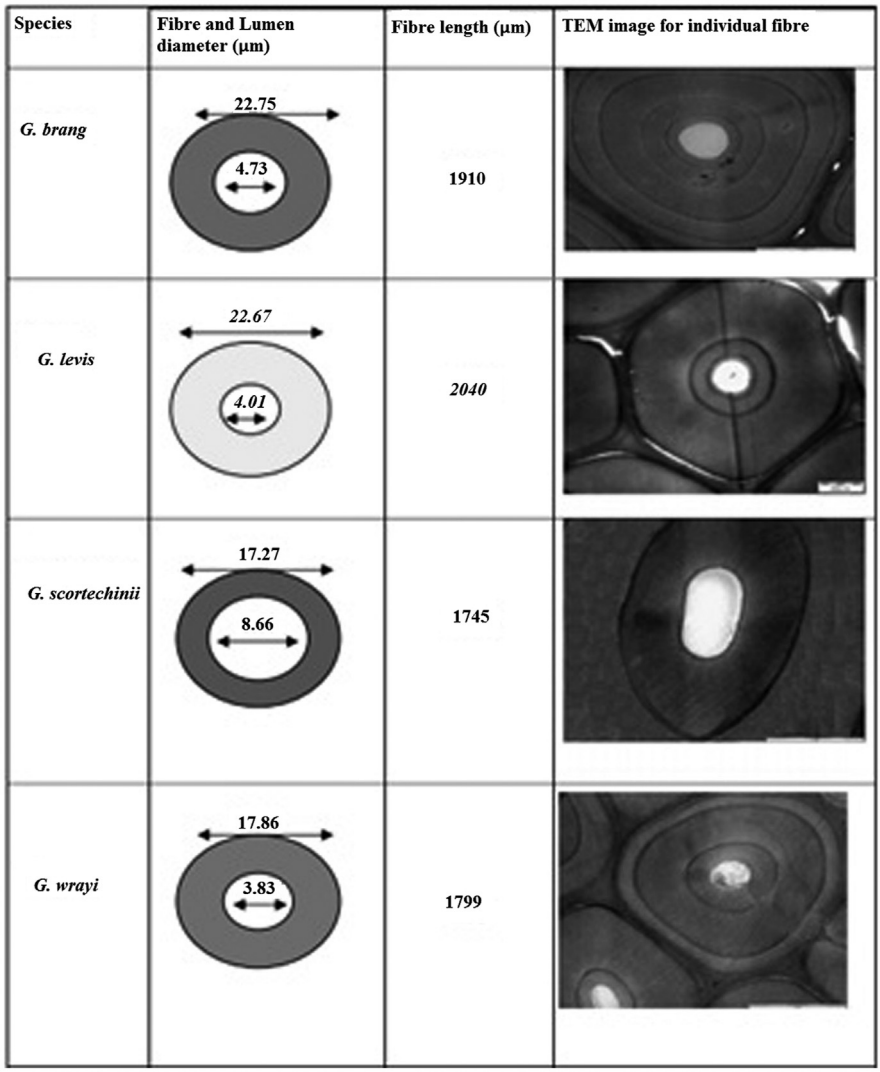


**Figure 2.11** Bamboo fiber length, fiber diameter, lumen diameter, and TEM micrograph. *Source:* From Tamizi, M., 2010. Fundamental and characteristic study of cultivated Malaysia bamboo-Selective genus Gigantochloa [Ph.D. thesis], Universiti Sains Malaysia, 211 p. Used with permission from Dr. Mohd Tamizi Mustafa.

The bamboo fiber reinforcement has been widely used in polymer composites construction (Wahyuni et al., 2014). Reinforcement of bamboo fiber exhibits better strength properties. A negative charge was found in bamboo fiber surfaces and has been used for cationic coating to eliminate electrostatic repulsion to have better properties (Ott et al., 2002). One research has studied the uniform distributed layer

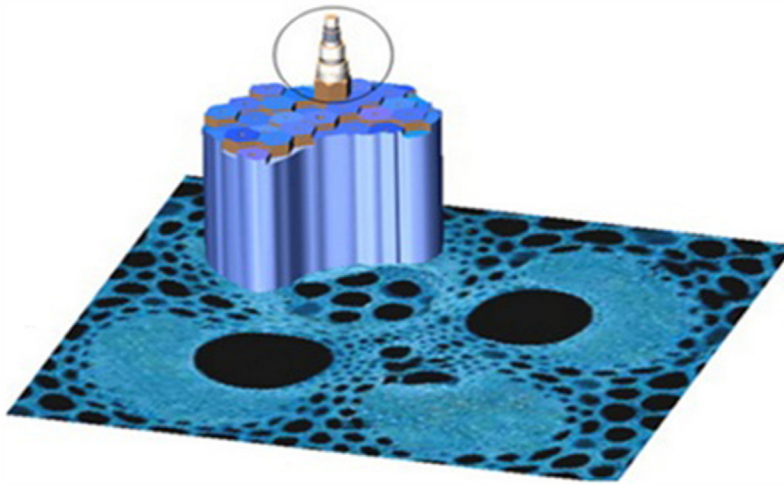

**Figure 2.12** Vascular bundles of bamboo plant.
*Source:* Fuentes, C., Tran, L.Q.N., Dupont-Gillain, C., Vanderlinden, W., De feyter, S., Van vuure, A., et al., 2011. Wetting behaviour and surface properties of technical bamboo fibres. Colloids Surfaces A: Physicochem. Eng. Aspects 380, 89−99 (Fuentes et al., 2011). This figure is reprinted with permission from Elsevier.

of nanoparticles on bamboo surfaces (Liu et al., 2015). A high absorption rate happened in the first 50 minutes due to the strong electrostatic attractive force by the bamboo fiber. This coating has shown that the bamboo fiber is able to enhance its properties by cationic coating to be used in an advanced sector. However, limited thermal stability of natural fiber has limited the reinforce bamboo fiber application (Mohanty and Nayak, 2010).

### 2.2.10 *Kenaf (*Hibiscus cannabinus *L.) fiber*

Kenaf fiber is one of the famous natural fibers used as a reinforcement in polymer matrix composites (PMC). Kenaf, known as *Hibiscus cannabimus L.*, is an herbaceous annual plant that grows in a wide range of weather conditions, growing more than 3 m within 3 months (Nishino et al., 2003). The highest growth rate may up to 10 cm/day. However, the difference of growth parameter influencing the properties of kenaf fiber such as length of growth season, plant population, cultivar, planting date, photosensitivity, and plant maturity. The stem of the kenaf plant is straight and is not branched along the stem. It is built up by bark and a core. Therefore it is easy to separate the stem by either chemicals or enzymatic retting. The bark has contributed 30%−40% of the dry weight for the stem while the wood-like core makes up the remaining weight. Long bast fiber type was used to make composite boards, textiles, pulp, and paper industry.

Rouison et al. (2004) has revealed the two main attracting reasons of kenaf fiber. Kenaf plant absorbs the nitrogen and phosphorus in the soil. These minerals were helped to increase the cumulative weed weight, crop height, stem diameter, and fiber yield. Kuchinda et al. (2001) suggested that the nitrogen application at 90 kgN/ha has a significant effect for kenaf plant growing. Another attracting reason is the high photosynthesis ability of kenaf (Nishino et al., 2003). The triple of photosynthesis rate of kenaf (23.4 mg $CO_2/dm^2/h$) compared to conventional tress (of 8.7 mg $CO_2/dm^2/h$) under 1000 μm $mol/cm^2/s$ helped to reduce the carbon dioxide while producing oxygen (Lam and Liyama, 2000).

The lighter and porous kenaf core fiber is rich in hemicellulose and lignin content (Alireza and Mohd, 2003). It has reported a better bonding ability than bast fiber since lignin acts as a cementing agent in fiber (Paridah et al., 2009). Kamal et al. (2009), has made a new particleboard by the kenaf core fiber with PP. The performance of the particleboard is satisfied, expect its high flammability, caused by the nature of kenaf fiber and petrochemical polymer product. Therefore, some modifications have been done on the board in order to solve this issue. A few types of fire retardant filler (DAP, MAP and BP (Boron)) were added into the sample and tend to get a better result. Untreated particleboard only spent 50 seconds to ignite while BP® was able to extend its ignition period to 2 minutes. It only has 8.52% of the burnt area with 0.69% of the weight loss. The boron is capable to provide protection to postpone the heat transfer (Horrocks and Price, 2001). Furthermore, other fire retardant filler candidates were chosen to conduct the same investigation and hoping to get the better result (Aisyah et al., 2013).

The bast kenaf fiber has better strength properties than core fiber; hence it is more suitable for high strength applications. A study has used the kenaf bast fiber to reinforce in the concrete composite to compare its properties with the plain concrete (Elsaid et al., 2011). The results had indicated the mechanical properties of the concrete composite were comparable to plain concrete specimen. Furthermore, the concrete composite has showed even distributed cracking and higher toughness. Therefore the concrete composite is claimed to be potential material for construction application.

The automotive sector has implanted the natural fiber reinforcement composite into the design for decades to achieve lower fuel consumption, lower cost, and more environmental friendly. However, the poor mechanical properties of the renewable materials have limited the idea of using natural fiber. Davoodi et al. (2010) have focused on combining the kenaf and glass fiber to improve the properties for a car bumper beam. The promising mechanical properties of this hybrid composite material have showed the potential of the natural fiber in the automotive sector. On the other hand, there are five concept designs of the kenaf fiber polymer composite automotive parking brake lever introduced by Mansor et al. (2014). One of the concept designs was selected for further development. Several selection processes and computerized analysis were done to replace the existing heavier steel-based parking brake lever, while maintaining the strength and performances.

## 2.3 Natural fiber reinforced vinyl polymer composites

### 2.3.1 Vinyl polymers

Vinyl polymers are an important part of the plastics industry, owing to the physical and chemical properties. The applications of such polymers are in many fields, such as packaging, adhesives (for textiles, paper, and woods), toys and sporting goods, medical field, electrical applications, construction and automotive industries. The vinyl polymers are the polymers that are made from vinyl (ethenyl) monomers (the molecules contain carbon−carbon double bonds and one hydrogen atom is substituted with other groups, Fig. 2.13). The vinyl monomers can be polymerized owing to the double bonds and formed polymers named vinyl polymers. The polymerization of this category of monomers is the additional polymerization in which the double bonds are changed into single bonds. Consequently, no vinyl groups are contained in the generated polymer. The vinyl polymers are classified into different categories based on the substituent groups of the vinyl monomers. If the substituent is an H, alkene, alkyl, aryl, or halogen, the polymer category is a polyolefin. If the substituent is cyanide or carboxylic group or its ester or amide, the category is an acrylic polymer (Brydson, 1999; Chanda and Roy, 2006). Fig. 2.13 illustrates the chemical structures of some vinyl polymers.

There are three classifications for vinyl polymers: vinyl plastics, thermosets, and rubbers. The thermoplastic, such as polystyrene and poly (vinyl chloride), is defined as the polymer that can be softened on an application of heat without a change in its properties. There are two classes of molecular arrangement for thermoplastics,

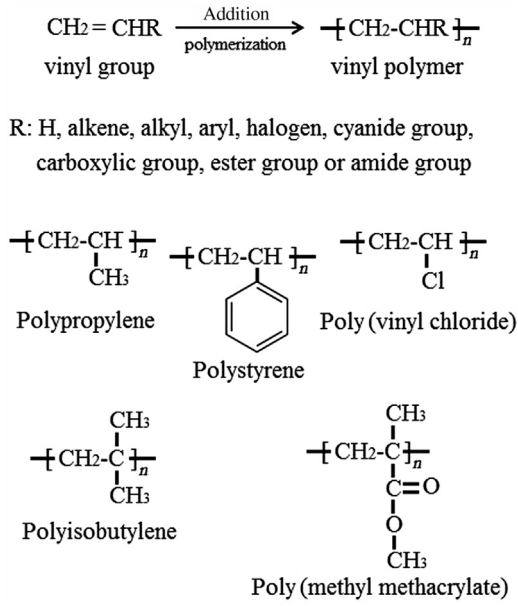


**Figure 2.13** Chemical structure of vinyl polymer with some examples.

amorphous and semicrystalline. Thermoplastics offer low processing costs, low energy for manufacture and low density. Thermosetting polymer is a resin; vinyl esters are the main types of the vinyl thermoset polymer. Thermoset polymer is the polymer that strengthened during heating and cannot be reheated after the initial heating. The thermoset advantages are high thermal stability, creep resistance, resistance to deformation, high hardness and rigidity, and low density. Rubber polymer, such as polyisobutylene, is the elastomer polymer that has poor molecular symmetry and very low $T_g$ (in the range of $-40^{\circ}C$ to $-80^{\circ}C$) for reasonable advantages and uses in industry (Mishra and Yagci, 2009).

### 2.3.2 Fiber reinforcement on vinyl polymers

Vinyl polymers have several advantages such as low cost, easy processability, and good chemical resistance. However, they also have low strength and modulus. The fiber form of materials is stronger and stiffer than the bulk material, thus fibers are effective reinforcement materials. Natural fiber reinforced vinyl polymer composites are widely used in different fields due to the biodegradability, environmentally friendly, high strength, stiffness, high impact resistance, low electrical conductivity, excellent corrosion resistance, low density and low cost. One of the drawbacks of natural fibers is the high degree surface hydrophilicity, consequently the moisture absorption. Several chemical treatments including alkaline, silane, acetylation, benzoylation, acrylation and acrylonitrile grafting, and maleated coupling agents can be applied to enhance the adhesion between the fiber and polymer. The indication of chemically-treated natural fibers efficiency is seen through the improvement of mechanical strength of resultant composites compared with the pristine polymer. The overall mechanical properties of natural FRP composites are highly dependent on the morphology, aspect ratio, hydrophilic tendency, and dimensional stability of the fibers used (Kabir et al., 2012; Bledzki and Gassan, 1999). In Section 2.2, *t* natural fibers and examples of the use of natural fibers in different applications were discussed. In this section, selected natural fiber reinforced vinyl polymer composites will be discussed.

#### 2.3.2.1 Short fiber reinforcement

Nair et al. (1996) studied tensile properties of short sisal fiber and benzoylated sisal fiber reinforced polystyrene composites. The influence of fiber length, fiber content, fiber orientation, and surface treatment of the fiber on the tensile properties of the composite were considered. The researchers found that the benzoylation treatment of the sisal fiber leads to improve the adhesion between the fiber and the polystyrene matrix. The tensile properties of the resulting composite were enhanced with the benzoylation treatment of the fiber (Fig. 2.14). This indicated a better compatibility between benzoylated fiber and polystyrene. The tensile properties of the composites show a gradual increase with fiber content whereas almost independent of fiber length, although the ultimate tensile strength shows marginal improvement at 10 mm fiber length (Table 2.5).

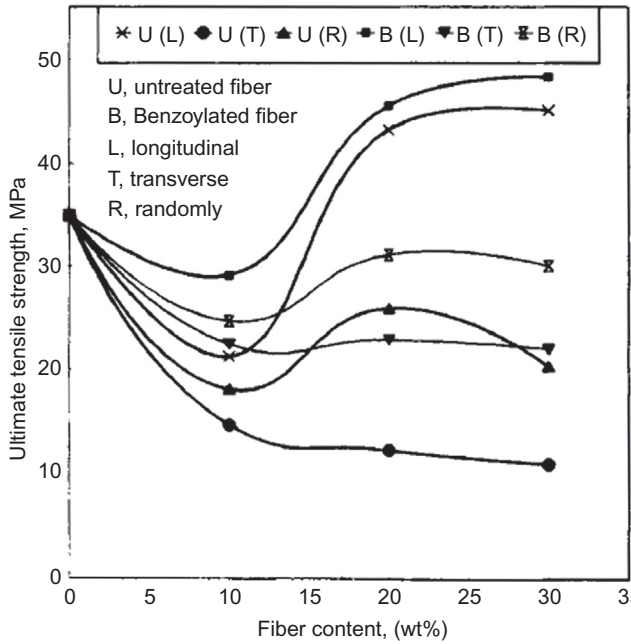


**Figure 2.14** Variation of the tensile strength of untreated and benzoylated sisal fiber composite as a function of fiber content.
*Source:* Nair, K., Diwan, S., Thomas, S., 1996. Tensile properties of short sisal fiber reinforced polystyrene composites. J. Appl. Polym. Sci., 60, 1483−1497. This figure is reprinted with permission from John Wiley & Sons, Inc.

**Table 2.5 Tensile properties of polystyrene untreated sisal fiber composite as a function of fiber length**

| Fiber Length (mm) | Elongation at break (%) | Ultimate tensile strength (MPa) | Young's modulus (MPa) |
|---|---|---|---|
| 2 | 6 | 21.12 | 666 |
| 6 | 9 | 21.3 | 629.6 |
| 10 | 9 | 25.06 | 657.1 |

*Source:* Nair, K., Diwan, S., Thomas, S., 1996. Tensile properties of short sisal fiber reinforced polystyrene composites. J. Appl. Polym. Sci., 60, 1483−1497. This table is reprinted with permission from John Wiley & Sons, Inc.

### *2.3.2.2 Long fiber reinforcement*

Unidirectional composites of PP with flax and hemp yarn fibers were studied by Bledzki et al. (2004); the fibers were modified by mercerization (NaOH) and maleic anhydride-polypropylene copolymer (MAH-PP) coupling agent to improve the properties of the composites. In general, the tests demonstrated that by optimizing

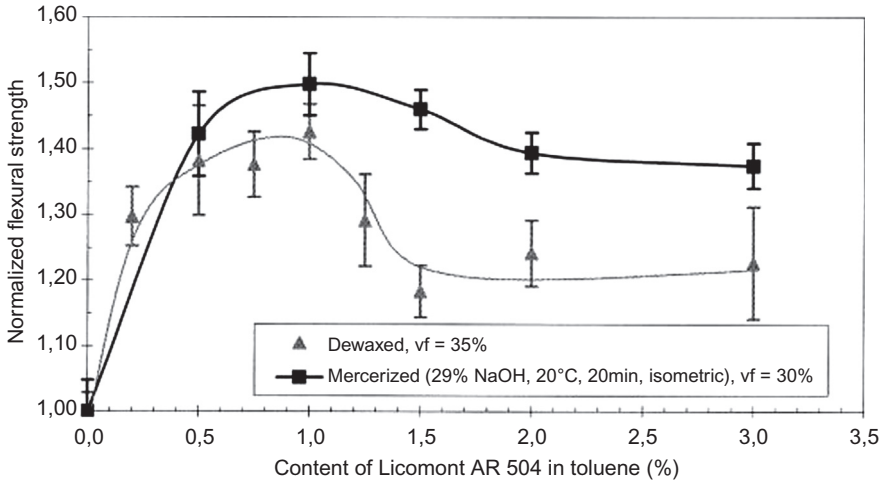


**Figure 2.15** The MAH-PP content versus a normalized flexural strength of unidirectional flax-PP-composites with and without fiber mercerization; vf is fiber volume content. *Source:* Bledzki, A., Fink, H.P., Specht, K., 2004. Unidirectional hemp and flax EP-and PP-composites: influence of defined fiber treatments. J. Appl. Polym. Sci. 93, 2150−2156. This figure is reprinted with permission from Wiley Periodicals, Inc.

the fibers structure and its surface by using the technique of chemical treatments, the fiber properties can be homogenized and controlled in a broad range (Bledzki et al., 2004). Fig. 2.15 shows the MAH-PP content versus a normalized flexural strength of unidirectional flax-PP-composites with and without fiber mercerization. Moreover, processing parameters of manufacturing the composites influence the final properties of the composites (Angelov et al., 2007).

### 2.3.2.3 *Hybrid composites*

Hybridization is the incorporation of two kinds or more of fibers into a single matrix material or vice versa, and the resulted composite is referred to as hybrid composites. The properties of the hybrid composites are dependent on the constituent materials and they are directly proportional to the additive properties (Bunsell and Harris, 1974; Summerscales and Short, 1978; Mishra and Yagci, 2009). Herzog et al. (2005) investigated the durability of the FRP composite-wood hybrid products. The researchers used composites pressure resin infusion system of fabrication to produce E-glass/vinyl ester FRP material directly on a wood surface; a hybrid composite resulted. They studied the shear stress, the percentage of wood failure experienced in shear, and the delamination of the glass FRP composite-wood interface when subjected to an accelerating aging test. The researchers reported that composite-pressure resin infusion system-fabricated hybrid exhibited a shear strength equal to, or greater than, the control methods of reinforced FRP-glulam manufacture. In their research, they recommended that the FRP used should be

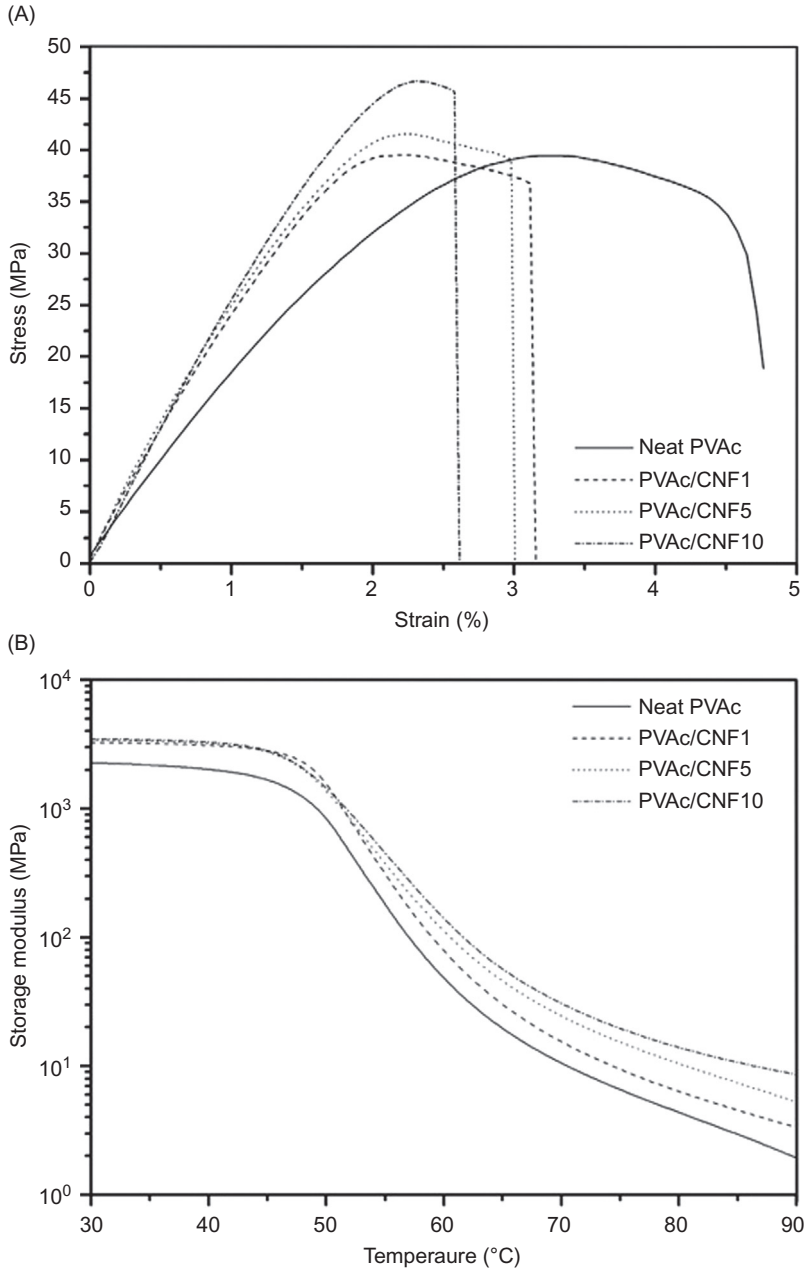


**Figure 2.16** Mechanical properties of neat PVAc and PVAc/CNF nanocomposites (A) the tensile stress−strain curves at a loading rate of 5 mm/min, and (B) storage modulus. *Source:* Gong, G., Pyo, J., Mathew, A.P., Oksman, K., 2011. Tensile behavior, morphology and viscoelastic analysis of cellulose nanofiber-reinforced (CNF) polyvinyl acetate (PVAc). Compos. Part A: Appl. Sci. Manufact. 42, 1275−1282. This figure is reprinted with permission from Elsevier.

thick enough to eliminate failure during testing; a postcuring procedure was recommended (Herzog et al., 2005; Mishra and Yagci, 2009).

The mechanical and physical properties of the vinyl ester reinforced with oil palm of empty fruit bunch fibers (EFB) laminated at different layer arrangements with glass fiber hybrid composites were investigated by Abdul Khalil et al. (2009). The researchers found that the mechanical properties, water absorption, and density of hybrid composites exhibited higher properties than control composites.

### 2.3.2.4 Nanocomposites

Gong et al. (2011) prepared and studied polyvinyl acetate (PVAc) nanocomposites. The CNF was used as reinforcement. The results demonstrated that the storage modulus, tensile modulus, and tensile strength were increased with increasing CNF content (Fig. 2.16). The creep strain of PVAc was reduced, whereas the creep elasticity and viscosity calculated from Burger's model were increased by the addition of CNF. Hybrid nanocomposites were prepared and investigated by Ching et al. (2015). The researchers used nanocellulose and nanosilica as reinforcements for PVA. The incorporation of the small percentage of the nanomaterials successfully improved the mechanical properties.

## 2.4 Conclusion

The natural FRPs have developed due to the advantages over other FRPs. The natural fiber composites are low density, low cost, high mechanical properties, and biodegradability. A review and discussion were presented in this chapter on the natural fibers and natural fiber reinforced vinyl polymers. There are some common defects that occur in processing fiber reinforced vinyl polymer composites. Consequently the effect on the final products needs to be considered. The common defects are including the resin cure, the fiber size and volume fraction, void volume content, the fiber distribution, misaligned or broken fibers, and fiber−polymer matrix adhesion.

## Acknowledgments

The authors would like to acknowledge all the people involved. Without their support, this discussion would not have become a reality. The authors would like to offer a special thanks to University Putra Malaysia and the library of Sultan Abdul Samad for the support in the research works and references.

## References

Aabdul khalil, H., Kang, C., Khairul, A., Ridzuan, R., Adawi, T., 2009. The effect of different laminations on mechanical and physical properties of hybrid composites. J. Reinfor. Plast. Compos. 28, 1123−1137.

Aabdul khalil, H., Bhat, A., Yusra, A.I., 2012a. Green composites from sustainable cellulose nanofibrils: a review. Carbohyd. Polym. 87, 963–979.

Aabdul khalil, H., Bhat, I., Jawaid, M., Zaidon, A., Hermawan, D., Hadi, Y., 2012b. Bamboo fibre reinforced biocomposites: a review. Mater. Design 42, 353–368.

Aamr-Daya, E., Langlet, T., Benazzouk, A., Quéneudec, M., 2008. Feasibility study of lightweight cement composite containing flax by-product particles: physico-mechanical properties. Cement Concr. Compos. 30, 957–963.

Aisyah, H., Paridah, M., Sahri, M., Anwar, U., Astimar, A., 2013. Properties of medium density fibreboard (MDF) from kenaf (*Hibiscus cannabinus* L.) core as function of refining conditions. Compos. Part B: Eng. 44, 592–596.

Akil, H.M., Omar, M.F., Mazuki, A.A.M., Safiee, S., Ishak, Z.A.M., Abu bakar, A., 2011. Kenaf fiber reinforced composites: a review. Mater. Design 32, 4107–4121.

Alade, G., Olutoge, F., Alade, A., 2004. The durability and mechanical strenght properties of bamboo in reinforced concrete. J. Appl. Sci. Eng. Technol. 4, 35–40.

Alireza, J.H., Mohd N.M.Y., 2003. Pulping and papermaking properties of Malaysian cultivated kenaf (Hibiscus cannabinus). Proceeding of the second technical review meeting on the national project kenaf research project, 2003 Serdang, pp. 108–115.

Alvarez, V.A., Fraga, A.N., Vázquez, A., 2004. Effects of the moisture and fiber content on the mechanical properties of biodegradable polymer–sisal fiber biocomposites. J. Appl. Polym. Sci. 91, 4007–4016.

Alves, C., Silva, A., Reis, L., Freitas, M., Rodrigues, L., Alves, D., 2010. Ecodesign of automotive components making use of natural jute fiber composites. J. Cleaner Production 18, 313–327.

Amato, E., Diaz-Fernandez, Y.A., Taglietti, A., Pallavicini, P., Pasotti, L., Cucca, L., et al., 2011. Synthesis, characterization and antibacterial activity against gram positive and gram negative bacteria of biomimetically coated silver nanoparticles. Langmuir 27, 9165–9173.

Amini, M., Arami, M., Mahmoodi, N.M., Akbari, A., 2011. Dye removal from colored textile wastewater using acrylic grafted nanomembrane. Desalination 267, 107–113.

Andresen, M., Stenstad, P., MØRETRØ, T., Langsrud, S., Syverud, K., Johansson, L.-S., et al., 2007. Nonleaching antimicrobial films prepared from surface-modified microfibrillated cellulose. Biomacromolecules 8, 2149–2155.

Angelov, I., Wiedmer, S., Evstatiev, M., Friedrich, K., Mennig, G., 2007. Pultrusion of a flax/polypropylene yarn. Compos. Part A: Appl. Sci. Manufact. 38, 1431–1438.

Anuar, H., Zuraida, A., 2011. Improvement in mechanical properties of reinforced thermoplastic elastomer composite with kenaf bast fibre. Composit. Part B: Eng. 42, 462–465.

Arao, Y., Fujiura, T., Itani, S., Tanaka, T., 2015. Strength improvement in injection-molded jute-fiber-reinforced polylactide green-composites. Composit. Part B: Eng. 68, 200–206.

Arbelaiz, A., Fernandez, B., Ramos, J., Retegi, A., Llano-Ponte, R., Mondragon, I., 2005. Mechanical properties of short flax fibre bundle/polypropylene composites: influence of matrix/fibre modification, fibre content, water uptake and recycling. Composit. Sci. Technol. 65, 1582–1592.

Arnaud, L., Gourlay, E., 2012. Experimental study of parameters influencing mechanical properties of hemp concretes. Construct. Build. Mater. 28, 50–56.

Arrakhiz, F., El achaby, M., Kakou, A., Vaudreuil, S., Benmoussa, K., Bouhfid, R., et al., 2012. Mechanical properties of high density polyethylene reinforced with chemically modified coir fibers: impact of chemical treatments. Mater. Design 37, 379–383.

Atanda, J., 2015. Environmental impacts of bamboo as a substitute constructional material in Nigeria. Case Stud. Construct. Mater. 3, 33–39.
Ataollahi, S., Taher, S.T., Eshkoor, R.A., Ariffin, A.K., Azhari, C.H., 2012. Energy absorption and failure response of silk/epoxy composite square tubes: experimental. Composit. Part B: Eng. 43, 542–548.
Atef, M., Rezaei, M., Behrooz, R., 2014. Preparation and characterization agar-based nanocomposite film reinforced by nanocrystalline cellulose. Int. J. Biol. Macromol. 70, 537–544.
Babaee, M., Jonoobi, M., Hamzeh, Y., Ashori, A., 2015. Biodegradability and mechanical properties of reinforced starch nanocomposites using cellulose nanofibers. Carbohyd. Polym. 132, 1–8.
Bagheri, Z.S., El sawi, I., Schemitsch, E.H., Zdero, R., Bougherara, H., 2013. Biomechanical properties of an advanced new carbon/flax/epoxy composite material for bone plate applications. J. Mech. Behav. Biomed. Mater. 20, 398–406.
Bagheri, Z.S., Giles, E., El sawi, I., Amleh, A., Schemitsch, E.H., Zdero, R., et al., 2015. Osteogenesis and cytotoxicity of a new Carbon Fiber/Flax/Epoxy composite material for bone fracture plate applications. Mater. Sci. Eng. C 46, 435–442.
Baiardo, M., Zini, E., Scandola, M., 2004. Flax fibre–polyester composites. Composit. Part A: Appl. Sci. Manufact. 35, 703–710.
Baillie, C., 2004. Green Composites: Polymer Composites and the Environment. Woodhead Publishing Ltd, Boca Raton.
Baker, D.A., Gallego, N.C., Baker, F.S., 2012. On the characterization and spinning of an organic-purified lignin toward the manufacture of low-cost carbon fiber. J. Appl. Polym. Sci. 124, 227–234.
Baltazar-Y-Jimenez, A., Bismarck, A., 2007. Wetting behaviour, moisture up-take and electrokinetic properties of lignocellulosic fibres. Cellulose 14, 115–127.
Bhardwaj, N., Rajkhowa, R., Wang, X., Devi, D., 2015. Milled non-mulberry silk fibroin microparticles as biomaterial for biomedical applications. Int. J. Biol. Macromol. 81, 31–40.
Bhattacharjee, P., Naskar, D., Kim, H.-W., Maiti, T.K., Bhattacharya, D., Kundu, S.C., 2015. Non-mulberry silk fibroin grafted PCL nanofibrous scaffold: promising ECM for bone tissue engineering. Eur. Polym. J. 71, 490–509.
Bhattacharya, M., Malinen, M.M., Lauren, P., Lou, Y.-R., Kuisma, S.W., Kanninen, L., et al., 2012. Nanofibrillar cellulose hydrogel promotes three-dimensional liver cell culture. J. Controlled Release 164, 291–298.
Bismarck, A., Baltazar-Y-Jimenez, A., Sarikakis, K., 2006. Green composites as panacea? Socio-economic aspects of green materials. Environ. Dev. Sustainab. 8, 445–463.
Bledzki, A., Gassan, J., 1999. Composites reinforced with cellulose based fibres. Progress Polym. Sci. 24, 221–274.
Bledzki, A., Fink, H.P., Specht, K., 2004. Unidirectional hemp and flax EP-and PP-composites: influence of defined fiber treatments. J. Appl. Polym. Sci. 93, 2150–2156.
Bledzki, A.K., Sperber, V., Faruk, O., 2002. Natural and wood fibre reinforcement in polymers, iSmithers Rapra Publishing.
Bolton, J., 1995. The potential of plant fibres as crops for industrial use. Outlook Agri. 24, 85–89.
Bonino, M.J., 2003. Material Properties of Spider Silk. University of Rochester, Rochester.
Bourmaud, A., Le duigou, A., Baley, C., 2011. What is the technical and environmental interest in reusing a recycled polypropylene–hemp fibre composite? Polym. Degrad. Stab. 96, 1732–1739.

Brinchi, L., Cotana, F., Fortunati, E., Kenny, J.M., 2013. Production of nanocrystalline cellulose from lignocellulosic biomass: technology and applications. Carbohyd. Polym. 94, 154–169.

Brydson, J.A., 1999. Plastics Materials. Butterworth-Heinemann, Oxford.

Buga, M.-R., Zaharia, C., BĂLan, M., Bressy, C., Ziarelli, F., Margaillan, A., 2015. Surface modification of silk fibroin fibers with poly (methyl methacrylate) and poly (tributylsilyl methacrylate) via RAFT polymerization for marine antifouling applications. Mater. Sci. Eng. C 51, 233–241.

Bunsell, A., Harris, B., 1974. Hybrid carbon and glass fibre composites. Composites 5, 157–164.

Calamak, S., Aksoy, E.A., Ertas, N., Erdogdu, C., SAGıRoglu, M., Ulubayram, K., 2015. Ag/silk fibroin nanofibers: effect of fibroin morphology on Ag + release and antibacterial activity. Europ. Polym. J. 67, 99–112.

Cantero, G., Arbelaiz, A., Llano-Ponte, R., Mondragon, I., 2003. Effects of fibre treatment on wettability and mechanical behaviour of flax/polypropylene composites. Composit. Sci. Technol. 63, 1247–1254.

Cha, R., Wang, C., Cheng, S., He, Z., Jiang, X., 2014. Using carboxylated nanocrystalline cellulose as an additive in cellulosic paper and poly (vinyl alcohol) fiber paper. Carbohyd. Polym. 110, 298–301.

Chanda, M., Roy, S.K., 2006. Plastics Technology Handbook. CRC Press, Boca Raton, FL.

Chawla, K., 1998. Fibrous Materials. Cambridge University Press, United Kingdom.

Chen, F., Porter, D., Vollrath, F., 2012a. Morphology and structure of silkworm cocoons. Mater. Sci. Eng. C 32, 772–778.

Chen, F., Porter, D., Vollrath, F., 2012b. Silk cocoon (*Bombyx mori*): multi-layer structure and mechanical properties. Acta Biomater. 8, 2620–2627.

Cherian, B.M., Leão, A.L., De souza, S.F., Costa, L.M.M., De olyveira, G.M., Kottaisamy, M., et al., 2011. Cellulose nanocomposites with nanofibres isolated from pineapple leaf fibers for medical applications. Carbohyd. Polym. 86, 1790–1798.

Ching, Y.C., Rahman, A., Ching, K.Y., Sukiman, N.L., Cheng, H.C., 2015. Preparation and characterization of polyvinyl alcohol-based composite reinforced with nanocellulose and nanosilica. BioResources 10, 3364–3377.

Chuai, C., Almdal, K., Poulsen, L., Plackett, D., 2001. Conifer fibers as reinforcing materials for polypropylene-based composites. J. Appl. Polym. Sci. 80, 2833–2841.

Cordeiro, N., Mendonça, C., Pothan, L., Varma, A., 2012. Monitoring surface properties evolution of thermochemically modified cellulose nanofibres from banana pseudo-stem. Carbohyd. Polym. 88, 125–131.

Cordero, A.I., Amalvy, J.I., Fortunati, E., Kenny, J.M., Chiacchiarelli, L.M., 2015. The role of nanocrystalline cellulose on the microstructure of foamed castor-oil polyurethane nanocomposites. Carbohyd. Polym. 134, 110–118.

Davoodi, M., Sapuan, S., Ahmad, D., Ali, A., Khalina, A., Jonoobi, M., 2010. Mechanical properties of hybrid kenaf/glass reinforced epoxy composite for passenger car bumper beam. Mater. Design 31, 4927–4932.

De Carvalho Mendes, C.A., Ferreira, N.M.S., Furtado, C.R.G., De sousa, A.M.F., 2015. Isolation and characterization of nanocrystalline cellulose from corn husk. Mater. Letters 148, 26–29.

Dewilde, B., 1983. 20 eeuwen vlas in Vlaanderen, Tielt, Bussum, Lannoo.

Dong, A., Yu, Y., Yuan, J., Wang, Q., Fan, X., 2014. Hydrophobic modification of jute fiber used for composite reinforcement via laccase-mediated grafting. Appl. Surface Sci. 301, 418–427.

Du, S., Li, J., Zhang, J., Wang, X., 2015. Microstructure and mechanical properties of silk from different components of the *Antheraea pernyi* cocoon. Mater. Design (1980-2015) 65, 766–771.

Elfehri borchani, K., Carrot, C., Jaziri, M., 2015. Biocomposites of Alfa fibers dispersed in the Mater-Bi® type bioplastic: morphology, mechanical and thermal properties. Compos. Part A: Appl. Sci. Manufact. 78, 371–379.

Elfordy, S., Lucas, F., Tancret, F., Scudeller, Y., Goudet, L., 2008. Mechanical and thermal properties of lime and hemp concrete ("hempcrete") manufactured by a projection process. Construct. Build. Mater. 22, 2116–2123.

Elsaid, A., Dawood, M., Seracino, R., Bobko, C., 2011. Mechanical properties of kenaf fiber reinforced concrete. Construct. Build. Mater. 25, 1991–2001.

Eshkoor, R., Oshkovr, S., Sulong, A.B., Zulkifli, R., Ariffin, A., Azhari, C., 2013a. Effect of trigger configuration on the crashworthiness characteristics of natural silk epoxy composite tubes. Composit. Part B: Eng. 55, 5–10.

Eshkoor, R.A., Oshkovr, S.A., Sulong, A.B., Zulkifli, R., Ariffin, A.K., Azhari, C.H., 2013b. Comparative research on the crashworthiness characteristics of woven natural silk/epoxy composite tubes. Mater. Design 47, 248–257.

Essabir, H., Bensalah, M., Bouhfid, R., Qaiss, A., 2014. Fabrication and characterization of apricot shells particles reinforced high density polyethylene based bio-composites: mechanical and thermal properties. J. Biobased Mater. Bioenergy 8, 344–351.

Etaati, A., Wang, H., Pather, S., Yan, Z., Mehdizadeh, S.A., 2013. 3D X-ray microtomography study on fibre breakage in noil hemp fibre reinforced polypropylene composites. Composit. Part B: Eng. 50, 239–246.

Etaati, A., Pather, S., Fang, Z., Wang, H., 2014. The study of fibre/matrix bond strength in short hemp polypropylene composites from dynamic mechanical analysis. Composit. Part B: Eng. 62, 19–28.

Etim, U., Umoren, S., Eduok, U., 2016. Coconut coir dust as a low cost adsorbent for the removal of cationic dye from aqueous solution. J. Saudi Chem. Soc. 20, S67–S76.

Fan, X., Zhang, T., Zhao, Z., Ren, H., Zhang, Q., Yan, Y., et al., 2013. Preparation and characterization of bacterial cellulose microfiber/goat bone apatite composites for bone repair. J. Appl. Polym. Sci. 129, 595–603.

Fatima, S., Mohanty, A., 2011. Acoustical and fire-retardant properties of jute composite materials. Appl. Acoustics 72, 108–114.

Felix, J.M., Gatenholm, P., 1991. The nature of adhesion in composites of modified cellulose fibers and polypropylene. J. Appl. Polym. Sci. 42, 609–620.

Fuentes, C., Tran, L.Q.N., Dupont-Gillain, C., Vanderlinden, W., De feyter, S., Van vuure, A., et al., 2011. Wetting behaviour and surface properties of technical bamboo fibres. Colloids Surfaces A: Physicochem. Eng. Aspects 380, 89–99.

Garcia-Jaldon, C., Dupeyre, D., Vignon, M., 1998. Fibres from semi-retted hemp bundles by steam explosion treatment. Biomass Bioenergy 14, 251–260.

Gassan, J., Bledzki, A.K., 1996. Composition of different natural fibers. Die Angewandte Makromolekulare Chemie 236, 129–138.

Godbole, V., Lakkad, S., 1986. Effect of water absorption on the mechanical properties of bamboo. J. Mater. Sci. Letters 5, 303–304.

Gong, G., Pyo, J., Mathew, A.P., Oksman, K., 2011. Tensile behavior, morphology and viscoelastic analysis of cellulose nanofiber-reinforced (CNF) polyvinyl acetate (PVAc). Composit. Part A: Appl. Sci. Manufact. 42, 1275–1282.

Gopinath, A., Kumar, M.S., Elayaperumal, A., 2014. Experimental investigations on mechanical properties of jute fiber reinforced composites with polyester and epoxy resin matrices. Proc. Eng. 97, 2052–2063.

Gratani, L., Crescente, M.F., Varone, L., Fabrini, G., Digiulio, E., 2008. Growth pattern and photosynthetic activity of different bamboo species growing in the Botanical Garden of Rome. Flora-Morphol. Distribut. Funct. Ecol. Plants 203, 77–84.

Gu, H., 2009. *Tensile be*haviours of the coir fibre and related composites after NaOH treatment. Mater. Design 30, 3931–3934.

Guo, Z., Liu, Z., Ye, L., Ge, K., Zhao, T., 2015. The production of lignin-phenol-formaldehyde resin derived carbon fibers stabilized by BN preceramic polymer. Mater. Letters 142, 49–51.

Hannukainen, K.-S., Suhonen, S., Savolainen, K., Norppa, H., 2012. Genotoxicity of nanofibrillated cellulose in vitro as measured by enzyme comet assay. Toxicol. Letters 211, S71.

Harish, S., Michael, D.P., Bensely, A., Lal, D.M., Rajadurai, A., 2009. Mechanical property evaluation of natural fiber coir composite. Mater. Character. 60, 44–49.

Hassan, A., Salema, A.A., Ani, F.N., Bakar, A.A., 2010. A review on oil palm empty fruit bunch fiber-reinforced polymer composite materials. Polym. Composit. 31, 2079–2101.

Hassan, M.S., 2015. Removal of reactive dyes from textile wastewater by immobilized chitosan upon grafted Jute fibers with acrylic acid by gamma irradiation. Radiat. Phys. Chem. 115, 55–61.

Herrera-Franco, P.J., Valadez-González, A., 2004. Mechanical properties of continuous natural fibre-reinforced polymer composites. Composit. Part A: Appl. Sci. Manufact. 35, 339–345.

Herzog, B., Goodell, B., Lopez-Anido, R., Gardner, D.J., 2005. Durability of fiber-reinforced polymer (FRP) composite-wood hybrid products fabricated using the composites pressure resin infusion system (ComPRIS). Forest Products J. 55, 54.

Ho, M.-P., Wang, H., Lau, K.-T., 2012a. Effect of degumming time on silkworm silk fibre for biodegradable polymer composites. Appl. Surface Sci. 258, 3948–3955.

Ho, M.-P., Wang, H., Lee, J.-H., Ho, C.-K., Lau, K.-T., Leng, J., et al., 2012b. Critical factors on manufacturing processes of natural fibre composites. Composit. Part B: Eng. 43, 3549–3562.

Horrocks, A.R., Price, D., 2001. Fire Retardant Materials. woodhead Publishing, Cambridge, England.

Hu, S., Zhang, S., Pan, N., Hsieh, Y.-L., 2014. High energy density supercapacitors from lignin derived submicron activated carbon fibers in aqueous electrolytes. J. Power Sources 270, 106–112.

Huda, M.S., Drzal, L.T., Mohanty, A.K., Misra, M., 2006. Chopped glass and recycled newspaper as reinforcement fibers in injection molded poly(lactic acid) (PLA) composites: a comparative study. Composit. Sci. Technol. 66, 1813–1824.

Jarman, C., 1998. Plant Fibre Processing: A Handbook. Intermediate Technology Publications, London.

Jawaid, M., Abdul khalil, H.P.S., 2011. Cellulosic/synthetic fibre reinforced polymer hybrid composites: a review. Carbohyd. Polym. 86, 1–18.

John, M.J., Thomas, S., 2008. Biofibres and biocomposites. Carbohyd. Polym. 71, 343–364.

Kabir, M., Wang, H., Lau, K., Cardona, F., 2012. Chemical treatments on plant-based natural fibre reinforced polymer composites: an overview. Composit. Part B: Eng. 43, 2883–2892.

Kamal, I., Malek, A.R.A., Yusof, M.N.M., Masseat, K., Ashaari, Z., Abood, F., 2009. Physical and mechanical properties of flame retardant-treated Hibiscus cannabinus particleboard. Modern Appl. Sci. 3, 2.

Karmaker, A., Youngquist, J., 1996. Injection molding of polypropylene reinforced with short jute fibers. J. Appl. Polym. Sci. 62, 1147–1151.

Kawahara, Y., Shioya, M., Takaku, A., 1996. Mechanical properties of silk fibers treated with methacrylamide. J. Appl. Polym. Sci. 61, 1359–1364.

Kazayawoko, M., Balatinecz, J., Woodhams, R., Law, S., 1997. Effect of ester linkages on the mechanical properties of wood fiber-polypropylene composites. J. Reinforced Plastics Composites 16, 1383–1406.

Kazemimostaghim, M., Rajkhowa, R., Wang, X., 2015. Drug loading and release studies for milled silk particles of different sizes. Powder Technol. 283, 321–327.

Kikuchi, T., Tani, Y., Takai, Y., Goto, A., Hamada, H., 2014. Mechanical Properties of Jute Composite by Spray up Fabrication Method. Energy Procedia 56, 289–297.

Kim, H.J., Um, I.C., 2014. Effect of degumming ratio on wet spinning and post drawing performance of regenerated silk. Int. J. Biol. Macromol. 67, 387–393.

Ko, J.S., Yoon, K., Ki, C.S., Kim, H.J., Bae, D.G., Lee, K.H., et al., 2013. Effect of degumming condition on the solution properties and electrospinnablity of regenerated silk solution. Int. J. Biol. Macromol. 55, 161–168.

Kostic, M., Pejic, B., Skundric, P., 2008. Quality of chemically modified hemp fibers. Bioresour. Technol. 99, 94–99.

Kozlowski, R., Wladyka-Przybylak, M., 2004. Uses of natural fiber reinforced plastics. Natural Fibers, Plastics and Composites. Springer.

Ku, H., Wang, H., Pattarachaiyakoop, N., Trada, M., 2011. A review on the tensile properties of natural fiber reinforced polymer composites. Composit. Part B: Eng. 42, 856–873.

Kuchinda, N., Ndahi, W., Lagoke, S., Ahmed, M., 2001. The effects of nitrogen and period of weed interference on the fibre yield of kenaf (Hisbiscus cannabinus L.) in the northern Guinea Savanna of Nigeria. Crop Protection 20, 229–235.

Kumar, A., Negi, Y.S., Choudhary, V., Bhardwaj, N.K., 2014. Characterization of cellulose nanocrystals produced by acid-hydrolysis from sugarcane bagasse as agro-waste. J. Mater. Phys. Chem. 2, 1–8.

Lalia, B.S., Guillen, E., Arafat, H.A., Hashaikeh, R., 2014. Nanocrystalline cellulose reinforced PVDF-HFP membranes for membrane distillation application. Desalination 332, 134–141.

Lam, T., Liyama, K., 2000. Structural details of kenaf cell walls and fixation of carbon dioxide. Proceedings of the Abstract of the 2000/International Kenaf Symposium.

Lewis, R.V., 2006. Spider silk: ancient ideas for new biomaterials. Chem. Rev. 106, 3762.

Liang, S., Gning, P.-B., Guillaumat, L., 2015. Quasi-static behaviour and damage assessment of flax/epoxy composites. Mater. Design 67, 344–353.

Lin, N., Dufresne, A., 2014. Nanocellulose in biomedicine: current status and future prospect. Eur. Polym. J. 59, 302–325.

Liu, X.-M., He, D.-Q., Fang, K.-J., 2015. Adsorption of cationic copolymer nanoparticles onto bamboo fiber surfaces measured by conductometric titration. Chinese Chem. Letters 26, 1174–1178.

Liu, Y., Wang, H., Yu, G., Yu, Q., Li, B., Mu, X., 2014. A novel approach for the preparation of nanocrystalline cellulose by using phosphotungstic acid. Carbohyd. Polym. 110, 415–422.

Lo, T.Y., Cui, H., Leung, H., 2004. The effect of fiber density on strength capacity of bamboo. Mater. Letters 58, 2595–2598.

Lu, N., Oza, S., 2013. Thermal stability and thermo-mechanical properties of hemp-high density polyethylene composites: effect of two different chemical modifications. Composit. Part B: Eng. 44, 484–490.

Ma, H., Zhuo, Q., Wang, B., 2009. Electro-catalytic degradation of methylene blue wastewater assisted by Fe 2 O 3-modified kaolin. Chem. Eng. J. 155, 248–253.

Maddahy, N., Ramezani, O., Kermanian, H., 2012. Production of nanocrystalline cellulose from sugarcane bagasse. Proceedings of the 4th International Conference on Nanostructures (ICNS4), pp. 12–14.

Mainka, H., Täger, O., Stoll, O., Körner, E., Herrmann, A.S., 2013. Alternative precursors for sustainable and cost-effective carbon fibers usable within the automotive industry. Society of Plastics Engineers (Automobile Division)–Automotive Composites Conference & Exhibition.

Mainka, H., Täger, O., Körner, E., Hilfert, L., Busse, S., Edelmann, F.T., et al., 2015. Lignin–an alternative precursor for sustainable and cost-effective automotive carbon fiber. J. Mater. Res. Technol. 4, 283–296.

Mansor, M.R., Sapuan, S., Zainudin, E.S., Nuraini, A., Hambali, A., 2014. Conceptual design of kenaf fiber polymer composite automotive parking brake lever using integrated TRIZ–Morphological Chart–Analytic Hierarchy Process method. Mater. Design (1980-2015) 54, 473–482.

Matias, M., De La Orden, M., Sánchez, C.G., Urreaga, J.M., 2000. Comparative spectroscopic study of the modification of cellulosic materials with different coupling agents. J. Appl. Polym. Sci. 75, 256–266.

Mehta, G., Mohanty, A., Misra, M., Drzal, L., 2004. Effect of novel sizing on the mechanical and morphological characteristics of natural fiber reinforced unsaturated polyester resin based bio-composites. J. Mater. Sci. 39, 2961–2964.

Melke, J., Midha, S., Ghosh, S., Ito, K., Hofmann, S., 2016. Silk fibroin as biomaterial for bone tissue engineering. Acta Biomater. 31, 1–16.

Meng, M., He, H., Xiao, J., Zhao, P., Xie, J., Lu, Z., 2016. Controllable in situ synthesis of silver nanoparticles on multilayered film-coated silk fibers for antibacterial application. J. Colloid Interface Sci. 461, 369–375.

Milanese, A.C., Cioffi, M.O.H., Voorwald, H.J.C., 2011. Mechanical behavior of natural fiber composites. Proc. Eng. 10, 2022–2027.

Miroiu, F.M., Stefan, N., Visan, A.I., Nita, C., Luculescu, C.R., Rasoga, O., et al., 2015. Composite biodegradable biopolymer coatings of silk fibroin–Poly (3-hydroxybutyric-acid-co-3-hydroxyvaleric-acid) for biomedical applications. Appl. Surface Sci. 355, 1123–1131.

Mishra, M., Yagci, Y., 2009. Handbook of Vinyl Polymers: Radical Polymerization, Process, and Technology. CRC Press, Boca Raton.

Mohanty, A., Drzal, L., Misra, M., 2002. Novel hybrid coupling agent as an adhesion promoter in natural fiber reinforced powder polypropylene composites. J. Mater. Sci. Letters 21, 1885–1888.

Mohanty, A.K., Misra, M., Hinrichsen, G., 2000. Biofibres, biodegradable polymers and biocomposites: an overview. Macromol. Mater. Eng. 276-277, 1–24.

Mohanty, A.K., Misra, M., Drzal, L.T., 2005. Natural Fibers, Biopolymers, and Biocomposites. CRC Press, Boca Raton.

Mohanty, S., Nayak, S.K., 2010. Short bamboo fibre-reinforced HDPE composites: influence of fibre content and modification on strength of the composite. J. Reinforced Plastics Composites.

Monteiro, S., Terrones, L., D'almeida, J., 2008. Mechanical performance of coir fiber/polyester composites. Polymer Testing 27, 591–595.

Mottaghitalab, F., Farokhi, M., Shokrgozar, M.A., Atyabi, F., Hosseinkhani, H., 2015. Silk fibroin nanoparticle as a novel drug delivery system. J. Controlled Release 206, 161–176.

Mougin, G., 2006. Natural-fibre composites: problems and solutions. JEC Composit. 32–35.

Mtibe, A., Linganiso, L.Z., Mathew, A.P., Oksman, K., John, M.J., Anandjiwala, R.D., 2015. A comparative study on properties of micro and nanopapers produced from cellulose and cellulose nanofibres. Carbohyd. Polym. 118, 1–8.

Muensri, P., Kunanopparat, T., Menut, P., Siriwattanayotin, S., 2011. Effect of lignin removal on the properties of coconut coir fiber/wheat gluten biocomposite. Composit. Part A: Appl. Sci. Manufact. 42, 173–179.

Mwangi, T.K., Bowles, R.D., Tainter, D.M., Bell, R.D., Kaplan, D.L., Setton, L.A., 2015. Synthesis and characterization of silk fibroin microparticles for intra-articular drug delivery. Int. J. Pharm. 485, 7–14.

Norppa, H., 2012. Nanofibrillated cellulose: results of in vitro and in vivo toxicological assays. Presentation on Sunpap conference.

Nadanathangam, V., Satyamurthy, P., 2011. Preparation of spherical nanocellulose by anaerobic microbial consortium. Proceedings of 2nd International Confer-ence on Biotechnology and Food Science. *IACSIT Press*, *Singapore*, pp. 181–183.

Nair, K., Diwan, S., Thomas, S., 1996. Tensile properties of short sisal fiber reinforced polystyrene composites. J. Appl. Polym. Sci. 60, 1483–1497.

Neto, W.P.F., Silvério, H.A., Dantas, N.O., Pasquini, D., 2013. Extraction and characterization of cellulose nanocrystals from agro-industrial residue–soy hulls. Ind. Crops Products 42, 480–488.

Newball, H.H., Brahim, S.A., 1976. Respiratory response to domestic fibrous glass exposure. Environ. Res. 12, 201–207.

Nishino, T., Hirao, K., Kotera, M., Nakamae, K., Inagaki, H., 2003. Kenaf reinforced biodegradable composite. Composit. Sci. Technol. 63, 1281–1286.

Niu, P., Liu, B., Wei, X., Wang, X., Yang, J., 2011. Study on mechanical properties and thermal stability of polypropylene/hemp fiber composites. J. Reinforced Plastics Composites, 30, 36–44.

Numata, K., Sato, R., Yazawa, K., Hikima, T., Masunaga, H., 2015. Crystal structure and physical properties of *Antheraea yamamai* silk fibers: long poly (alanine) sequences are partially in the crystalline region. Polymer 77, 87–94.

Olsen, D., 1991 Effectiveness of maleated polypropylenes as coupling agents for wood flour/polypropylene composites. Proceedings of the ANTEC Conference: Society of Plastics Engineers (Eds) Montreal, Canada.

Oshkovr, S., Eshkoor, R., Taher, S., Ariffin, A., Azhari, C., 2012. Crashworthiness characteristics investigation of silk/epoxy composite square tubes. Composit. Struct. 94, 2337–2342.

Oshkovr, S.A., Taher, S.T., Oshkour, A.A., Ariffin, A.K., Azhari, C.H., 2013. Finite element modelling of axially crushed silk/epoxy composite square tubes. Composit. Struct. 95, 411–418.

Osorio, L., Trujillo, E., Van vuure, A., Verpoest, I., 2011. Morphological aspects and mechanical properties of single bamboo fibers and flexural characterization of bamboo/epoxy composites. J. Reinforced Plastics Composites 30, 396–408.

Ott, G., Singh, M., Kazzaz, J., Briones, M., Soenawan, E., Ugozzoli, M., et al., 2002. A cationic sub-micron emulsion (MF59/DOTAP) is an effective delivery system for DNA vaccines. J. Controlled Release 79, 1–5.

Oyejobi, D., Jimoh, A.A., 2009. The Prospects, Challenges and Approach to Full Utilization of Bamboo as a Structural Material.

PÉREZ-Rigueiro, J., Viney, C., Llorca, J., Elices, M., 1998. Silkworm silk as an engineering material. J. Appl. Polym. Sci. 70, 2439–2447.

Panaitescu, D.M., Vuluga, Z., Ghiurea, M., Iorga, M., Nicolae, C., Gabor, R., 2015. Influence of compatibilizing system on morphology, thermal and mechanical properties of high flow polypropylene reinforced with short hemp fibers. Composit. Part B: Eng. 69, 286–295.

Panda, N., Bissoyi, A., Pramanik, K., Biswas, A., 2015. Development of novel electrospun nanofibrous scaffold from *P. ricini* and *A. mylitta* silk fibroin blend with improved surface and biological properties. Mater. Sci. Eng. C 48, 521–532.

Pantamanatsopa, P., Ariyawiriyanan, W., Meekeaw, T., Suthamyong, R., Arrub, K., Hamada, H., 2014. Effect of modified jute fiber on mechanical properties of green rubber composite. Energy Proc. 56, 641–647.

Paridah, M., Hafizah, A.N., Zaidon, A., Azmi, I., Nor, M.M., Yuziah, M.N., 2009. Bonding properties and performance of multi-layered kenaf board. J. Tropical Forest Sci. 113–122.

Patil, S., George, T., Mahadik, K., 2015. Green synthesized nanosilver loaded silk fibroin gel for enhanced wound healing. J. Drug Delivery Sci. Technol. 30, 30–36.

Peng, B.L., Dhar, N., Liu, H., Tam, K., 2011. Chemistry and applications of nanocrystalline cellulose and its derivatives: a nanotechnology perspective. Canadian J. Chem. Eng. 89, 1191–1206.

Phong, N.T., Fujii, T., Chuong, B., Okubo, K., 2011. Study on how to effectively extract bamboo fibers from raw bamboo and wastewater treatment. J. Mater. Sci. Res. 1, 144.

Pickering, K., 2008. Properties and Performance of Natural-Fibre Composites. Woodhead Publishing, Cambridge.

Pirani, S., Hashaikeh, R., 2013. Nanocrystalline cellulose extraction process and utilization of the byproduct for biofuels production. Carbohyd. Polym. 93, 357–363.

Pothan, L.A., Oommen, Z., Thomas, S., 2003. Dynamic mechanical analysis of banana fiber reinforced polyester composites. Composit. Sci. Technol. 63, 283–293.

Prabhakaran, S., Krishnaraj, V., Zitoune, R., 2014. Sound and vibration damping properties of flax fiber reinforced composites. Proc. Eng. 97, 573–581.

Pritchard, E.M., Kaplan, D.L., 2011. Silk fibroin biomaterials for controlled release drug delivery. Exp. Opin. Drug Delivery 8, 797–811.

Qing, Y., Sabo, R., Zhu, J., Agarwal, U., Cai, Z., Wu, Y., 2013. A comparative study of cellulose nanofibrils disintegrated via multiple processing approaches. Carbohyd. Polym. 97, 226–234.

Rafatullah, M., Sulaiman, O., Hashim, R., Ahmad, A., 2010. Adsorption of methylene blue on low-cost adsorbents: a review. J. Hazardous Mater. 177, 70–80.

Ragauskas, A.J., Beckham, G.T., Biddy, M.J., Chandra, R., Chen, F., Davis, M.F., et al., 2014. Lignin valorization: improving lignin processing in the biorefinery. Science 344, 1246843.

Rahman, M.M., Khan, M.A., 2007. Surface treatment of coir (Cocos nucifera) fibers and its influence on the fibers' physico-mechanical properties. Composit. Sci. Technol. 67, 2369–2376.

Rahman, M.M., Afrin, S., Haque, P., Islam, M.M., Islam, M.S., Gafur, M.A., 2014. Preparation and characterization of jute cellulose crystals-reinforced poly (L-lactic acid) biocomposite for biomedical applications. Int. J. Chem. Eng. 2014.

Rai, M., Yadav, A., Gade, A., 2009. Silver nanoparticles as a new generation of antimicrobials. Biotechnol. Adv. 27, 76–83.

Rathod, M., Haldar, S., Basha, S., 2015. Nanocrystalline cellulose for removal of tetracycline hydrochloride from water via biosorption: equilibrium, kinetic and thermodynamic studies. Ecol. Eng. 84, 240–249.

Ray, A.K., Das, S.K., Mondal, S., Ramachandrarao, P., 2004. Microstructural characterization of bamboo. J. Mater. Sci. 39, 1055–1060.

Riaño, N., Londoño, X., López, Y., Gómez, J., 2002. Plant growth and biomass distribution on Guadua angustifolia Kunth in relation to ageing in the Valle del Cauca–Colombia. J. Am. Bamboo Soc. 16, 43–51.

Rouison, D., Sain, M., Couturier, M., 2004. Resin transfer molding of natural fiber reinforced composites: cure simulation. Composit. Sci. Technol. 64, 629–644.

Rouison, D., Sain, M., Couturier, M., 2006. Resin transfer molding of hemp fiber composites: optimization of the process and mechanical properties of the materials. Composit. Sci. Technol. 66, 895–906.

Rout, J., Misra, M., Tripathy, S., Nayak, S., Mohanty, A., 2001. The influence of fibre treatment on the performance of coir-polyester composites. Composit. Sci. Technol. 61, 1303–1310.

Saikia, P., Dutta, D., Kalita, D., Bora, J.J., Goswami, T., 2015. Improvement of mechanochemical properties of bamboo by bio-chemical treatment. Construct. Build. Mater. 101, 1031–1036.

Sajab, M.S., Chia, C.H., Zakaria, S., Jani, S.M., Ayob, M.K., Chee, K.L., et al., 2011. Citric acid modified kenaf core fibres for removal of methylene blue from aqueous solution. Bioresour. Technol. 102, 7237–7243.

Salentijn, E.M., Zhang, Q., Amaducci, S., Yang, M., Trindade, L.M., 2015. New developments in fiber hemp (*Cannabis sativa* L.) breeding. Ind. Crops Products 68, 32–41.

Salit, M.S., 2009. Research on Natural Fibre Reinforced Polymer Composites. Universiti Putra Malaysia Press, Serdang.

Sanadi, A.R., Caulfield, D.F., Jacobson, R.E., Rowell, R.M., 1995. Renewable agricultural fibers as reinforcing fillers in plastics: mechanical properties of kenaf fiber-polypropylene composites. Ind. Eng. Chem. Res. 34, 1889.

Saska, S., Barud, H., Gaspar, A., Marchetto, R., Ribeiro, S.J.L., Messaddeq, Y., 2011. Bacterial cellulose-hydroxyapatite nanocomposites for bone regeneration. Int. J. Biomater. 2011.

Satyamurthy, P., Vigneshwaran, N., 2013. A novel process for synthesis of spherical nanocellulose by controlled hydrolysis of microcrystalline cellulose using anaerobic microbial consortium. Enzyme Microbial Technol. 52, 20–25.

Satyamurthy, P., Jain, P., Balasubramanya, R.H., Vigneshwaran, N., 2011. Preparation and characterization of cellulose nanowhiskers from cotton fibres by controlled microbial hydrolysis. Carbohyd. Polym. 83, 122–129.

Satyanarayana, K.G., Arizaga, G.G.C., Wypych, F., 2009. Biodegradable composites based on lignocellulosic fibers—an overview. Progress Polym. Sci. 34, 982–1021.

Sawsen, C., Fouzia, K., Mohamed, B., Moussa, G., 2014. Optimizing the formulation of flax fiber-reinforced cement composites. Construct. Build. Mater. 54, 659–664.

Sawsen, C., Fouzia, K., Mohamed, B., Moussa, G., 2015. Effect of flax fibers treatments on the rheological and the mechanical behavior of a cement composite. Construct. Build. Mater. 79, 229–235.

Scarponi, C., Messano, M., 2015. Comparative evaluation between E-Glass and hemp fiber composites application in rotorcraft interiors. Composit. Part B: Eng. 69, 542–549.

Shahzad, A., 2012. Hemp fiber and its composites–a review. J. Composit. Mater. 46, 973–986.

Shinoj, S., Visvanathan, R., Panigrahi, S., Kochubabu, M., 2011. Oil palm fiber (OPF) and its composites: a review. Ind. Crops Products 33, 7–22.

Shubhra, Q.T., Alam, A., Gafur, M., Shamsuddin, S.M., Khan, M.A., Saha, M., et al., 2010. Characterization of plant and animal based natural fibers reinforced polypropylene composites and their comparative study. Fibers Polym. 11, 725–731.

Shukla, S., Pai, R.S., Shendarkar, A.D., 2006. Adsorption of Ni (II), Zn (II) and Fe (II) on modified coir fibres. Sep. Purif. Technol. 47, 141–147.

Solanas, C., Herrero, S., Dasari, A., Plaza, G.R., Llorca, J., PÉREZ-Rigueiro, J., et al., 2014. Insights into the production and characterization of electrospun fibers from regenerated silk fibroin. Eur. Polym. J. 60, 123–134.

Somvipart, S., Kanokpanont, S., Rangkupan, R., Ratanavaraporn, J., Damrongsakkul, S., 2013. Development of electrospun beaded fibers from Thai silk fibroin and gelatin for controlled release application. Int. J. Biol. Macromol. 55, 176–184.

Song, Y.S., Lee, J.T., Ji, D.S., Kim, M.W., Lee, S.H., Youn, J.R., 2012. Viscoelastic and thermal behavior of woven hemp fiber reinforced poly (lactic acid) composites. Composit. Part B: Eng. 43, 856–860.

Summerscales, J., Short, D., 1978. Carbon fibre and glass fibre hybrid reinforced plastics. Composites 9, 157–166.

Summerscales, J., Dissanayake, N.P.J., Virk, A.S., Hall, W., 2010. A review of bast fibres and their composites. Part 1 – Fibres as reinforcements. Composit. Part A: Appl. Sci. Manufact. 41, 1329–1335.

Syverud, K., Chinga-Carrasco, G., Toledo, J., Toledo, P.G., 2011. A comparative study of Eucalyptus and Pinus radiata pulp fibres as raw materials for production of cellulose nanofibrils. Carbohyd. Polym. 84, 1033–1038.

Tamizi, M., 2010. Fundamental and characteristic study of cultivated Malaysia bamboo-Selective genus Gigantochloa. PhD. Thesis, Universiti Sains Malaysia, 211p.

Tazi, N., Zhang, Z., Messaddeq, Y., Almeida-Lopes, L., Zanardi, L.M., Levinson, D., et al., 2012. Hydroxyapatite bioactivated bacterial cellulose promotes osteoblast growth and the formation of bone nodules. Amb Express 2, 61.

Teimouri, A., Azadi, M., Emadi, R., Lari, J., Chermahini, A.N., 2015. Preparation, characterization, degradation and biocompatibility of different silk fibroin based composite scaffolds prepared by freeze-drying method for tissue engineering application. Polym. Degrad. Stab. 121, 18–29.

Thomas, S., Pothan, L.A., 2009. Natural Fibre Reinforced Polymer Composites: From Macro to Nanoscale. Old City Publishing, Philadelphia.

Thwe, M.M., Liao, K., 2003. Environmental effects on bamboo-glass/polypropylene hybrid composites. J. Mater. Sci. 38, 363–376.

Tibolla, H., Pelissari, F.M., Menegalli, F.C., 2014. Cellulose nanofibers produced from banana peel by chemical and enzymatic treatment. LWT-Food Sci. Technol. 59, 1311–1318.

Tiryaki, B., Yagmur, E., Banford, A., Aktas, Z., 2014. Comparison of activated carbon produced from natural biomass and equivalent chemical compositions. J. Analyt. Appl. Pyrolysis 105, 276–283.

Tokareva, O., Jacobsen, M., Buehler, M., Wong, J., Kaplan, D.L., 2014. Structure–function–property–design interplay in biopolymers: spider silk. Acta Biomater. 10, 1612–1626.

Tonoli, G., Teixeira, E., Corrêa, A., Marconcini, J., Caixeta, L., Pereira-Da-Silva, M., et al., 2012. Cellulose micro/nanofibres from Eucalyptus kraft pulp: preparation and properties. Carbohyd. Polym. 89, 80–88.

Tsukada, M., Obo, M., Kato, H., Freddi, G., Zanetti, F., 1996. Structure and dyeability of *Bombyx mori* silk fibers with different filament sizes. J. Appl. Polym. Sci. 60, 1619–1627.

Ude, A., Ariffin, A., Azhari, C., 2013a. An experimental investigation on the response of woven natural silk fiber/epoxy sandwich composite panels under low velocity impact. Fibers Polym. 14, 127–132.

Ude, A., Ariffin, A., Azhari, C., 2013b. Impact damage characteristics in reinforced woven natural silk/epoxy composite face-sheet and sandwich foam, coremat and honeycomb materials. Int. J. Impact Eng. 58, 31–38.

Ude, A., Eshkoor, R., Zulkifili, R., Ariffin, A., Dzuraidah, A., Azhari, C., 2014. *Bombyx mori* silk fibre and its composite: a review of contemporary developments. Mater. Design 57, 298–305.

Väänänen, V., Rydman, E., Ilves, M., Hannukainen, K., Norppa, H., Von wright, A., et al., 2012 Evaluation of the suitability of the developed methodology for nanoparticle health and safety studies. SUNPAP Conference.

Venkateshwaran, N., Elayaperumal, A., 2010. Banana fiber reinforced polymer composites - a review. J. Reinforced Plastics Composites 29, 2387–2396.

Voronova, M., Surov, O., Zakharov, A., 2013. Nanocrystalline cellulose with various contents of sulfate groups. Carbohyd. Polym. 98, 465–469.

Voronova, M.I., Surov, O.V., Guseinov, S.S., Barannikov, V.P., Zakharov, A.G., 2015. Thermal stability of polyvinyl alcohol/nanocrystalline cellulose composites. Carbohyd. Polym. 130, 440–447.

Wahyuni, A.S., Supriani, F., Gunawan, A., 2014. The performance of concrete with rice husk ash, sea shell ash and bamboo fibre addition. Proc. Eng. 95, 473–478.

Wang, B., Sain, M., Oksman, K., 2007. Study of structural morphology of hemp fiber from the micro to the nanoscale. Appl. Compos. Mater. 14, 89–103.

Wang, C., Huang, H., Jia, M., Jin, S., Zhao, W., Cha, R., 2015a. Formulation and evaluation of nanocrystalline cellulose as a potential disintegrant. Carbohyd. Polym. 130, 275–279.

Wang, C.-S., Ashton, N.N., Weiss, R.B., Stewart, R.J., 2014a. Peroxinectin catalyzed dityrosine crosslinking in the adhesive underwater silk of a casemaker caddisfly larvae, *Hysperophylax occidentalis*. Insect Biochem. Mol. Biol. 54, 69–79.

Wang, F., Cao, T.-T., Zhang, Y.-Q., 2015b. Effect of silk protein surfactant on silk degumming and its properties. Mater. Sci. Eng. C 55, 131–136.

Wang, J., Zhang, S., Xing, T., Kundu, B., Li, M., Kundu, S.C., et al., 2015c. Ion-induced fabrication of silk fibroin nanoparticles from Chinese oak tasar Antheraea pernyi. Int. J. Biol. Macromol. 79, 316–325.

Wang, J.-T., Li, L.-L., Feng, L., Li, J.-F., Jiang, L.-H., Shen, Q., 2014b. Directly obtaining pristine magnetic silk fibers from silkworm. Int. J. Biol. Macromol. 63, 205–209.

Wang, K., Addiego, F., Laachachi, A., Kaouache, B., Bahlouli, N., Toniazzo, V., et al., 2014c. Dynamic behavior and flame retardancy of HDPE/hemp short fiber composites: effect of coupling agent and fiber loading. Composit. Struct. 113, 74–82.

Wang, S., Li, Y., Xiang, H., Zhou, Z., Chang, T., Zhu, M., 2015d. Low cost carbon fibers from bio-renewable Lignin/Poly (lactic acid)(PLA) blends. Composit. Sci. Technol. 119, 20–25.

Wenk, E., Merkle, H.P., Meinel, L., 2011. Silk fibroin as a vehicle for drug delivery applications. J. Controlled Release 150, 128–141.

Wever, D.-A.Z., Heeres, H., Broekhuis, A.A., 2012. Characterization of *Physic nut* (*Jatropha curcas* L.) shells. Biomass Bioenergy 37, 177–187.

Xiong, R., Lu, C., Zhang, W., Zhou, Z., Zhang, X., 2013. Facile synthesis of tunable silver nanostructures for antibacterial application using cellulose nanocrystals. Carbohyd. Polym. 95, 214–219.

Xu, Q., Gao, Y., Qin, M., Wu, K., Fu, Y., Zhao, J., 2013a. Nanocrystalline cellulose from aspen kraft pulp and its application in deinked pulp. Int. J. Biol. Macromol. 60, 241–247.

Xu, X., Zhou, J., Jiang, L., Lubineau, G., Chen, Y., Wu, X.-F., et al., 2013b. Porous core-shell carbon fibers derived from lignin and cellulose nanofibrils. Mater. Letters 109, 175–178.

Xu, Y., Salmi, J., Kloser, E., Perrin, F., Grosse, S., Denault, J., et al., 2013c. Feasibility of nanocrystalline cellulose production by endoglucanase treatment of natural bast fibers. Ind. Crops Products 51, 381–384.

Yallew, T.B., Kumar, P., Singh, I., 2014. Sliding wear properties of jute fabric reinforced polypropylene composites. Proc. Eng. 97, 402–411.

Yoon, K., Lee, H.N., Ki, C.S., Fang, D., Hsiao, B.S., Chu, B., et al., 2013. Effects of degumming conditions on electro-spinning rate of regenerated silk. Int. J. Biol. Macromol. 61, 50–57.

Yousefi, H., Faezipour, M., Hedjazi, S., Mousavi, M.M., Azusa, Y., Heidari, A.H., 2013. Comparative study of paper and nanopaper properties prepared from bacterial cellulose nanofibers and fibers/ground cellulose nanofibers of canola straw. Ind. Crops Products 43, 732–737.

Yu, D., Kang, G., Tian, W., Lin, L., Wang, W., 2015. Preparation of conductive silk fabric with antibacterial properties by electroless silver plating. Appl. Surface Sci. 357, 1157–1162.

Yue, X., Zhang, F., Wu, H., Ming, J., Fan, Z., Zuo, B., 2014. A novel route to prepare dry-spun silk fibers from CaCl 2–formic acid solution. Mater. Letters 128, 175–178.

Zhang, H., Liu, X., Yang, M., Zhu, L., 2015a. Silk fibroin/sodium alginate composite nanofibrous scaffold prepared through thermally induced phase-separation (TIPS) method for biomedical applications. Mater. Sci. Eng. C 55, 8–13.

Zhang, H., Wang, K., Xing, Y., Yu, Q., 2015b. Lysine-doped polypyrrole/spider silk protein/poly (l-lactic) acid containing nerve growth factor composite fibers for neural application. Mater. Sci. Eng. C 56, 564–573.

Zhang, Q., Wang, N., Hu, R., Pi, Y., Feng, J., Wang, H., et al., 2015c. Wet spinning of Bletilla striata polysaccharide/silk fibroin hybrid fibers. Mater. Letters 161, 576–579.

Zhang, W., Yin, J., Lin, Z., Lin, H., Lu, H., Wang, Y., et al., 2015d. Facile preparation of 3D hierarchical porous carbon from lignin for the anode material in lithium ion battery with high rate performance. Electrochim. Acta 176, 1136–1142.

Zhang, Y., Lu, X.-B., Gao, C., Lv, W.-J., Yao, J.-M., 2012. Preparation and characterization of nano crystalline cellulose from bamboo fibers by controlled cellulase hydrolysis. J. Fiber Bioeng. Inform. 5, 263−271.

Zhao, J., Zhang, W., Zhang, X., Zhang, X., Lu, C., Deng, Y., 2013. Extraction of cellulose nanofibrils from dry softwood pulp using high shear homogenization. Carbohyd. Polym. 97, 695−702.

Zhou, M.-S., Sun, Z.-J., Yang, D.-J., Huang, J.-H., Qiu, X.-Q., 2014. The effect of plasticizer on the properties of alkali lignin/HDPE composites. Acta Polymer. Sinica 2, 210−217.

Zhu, J.Y., Sabo, R., Luo, X., 2011. Integrated production of nano-fibrillated cellulose and cellulosic biofuel (ethanol) by enzymatic fractionation of wood fibers. Green Chem. 13, 1339−1344.