**NECESSITY AND EMERGING APPLICATIONS OF BIOCOMPOSITES BASED ON SUSTAINABLE POLYMERIC MATERIALS**

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**Abstract**

Recently bio-based polymers and their composite materials have become of great interest for industrialists, and scientific researchers to deal with non-biodegradable synthetic plastic materials. The growing amount of synthetic polymeric materials is a great environmental problem that has to be solved as soon as possible. Plant-based natural fibre; a high potential field of the reinforced polymer composite material is considered as lightweight and economical products as they possess lower density, significant material characteristics, and extraordinary molding flexibility. In this chapter, we consider the possibility of solving this problem by the development of biodegradable compositions based on polyolefins and elastomers. The plant-fiber-based composites have widespread usage in construction, automotive, packaging, sports, biomedical, and defence sectors for their superior characteristics. Therefore, this chapter would demonstrate an overview regarding the background of natural fibre composites, factors influencing the composite properties, chemical interaction between the fiber and matrices, future potentiality, and marketing perspectives for triggering new research works in the field of biocomposite (BC) materials. Their future wide scopes are in commercial market to design them as biodegradable, compostable, environment friendly, cheaper, low waste, pollution less, and sustainable in end use performance.

**Keywords:** Biopolymer, Bio-composites, Sustainable, Biodegradable plastic

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***1.1. Biocomposites***

Biocomposites Composites are materials consisting of at least two identifiable constituents: in most cases, a matrix reinforced with fibers. The functions of the matrix are to hold the reinforcement in place, to deform and distribute the stress to the reinforcement under applied loads, and to provide a chemical protection, whereas the main function of the fibers is to provide mechanical properties to the material. When at least one of the two constituents is bio-based, such materials are called biocomposites [1]. Biocomposite materials are of great interest because they combine material properties in ways not found in nature, often resulting in lightweight structures with tailored properties for specific applications, thereby saving weight and reducing energy needs. The necessity of replacing petroleum-based products in our everyday life makes biocomposites a good alternative and this is the reason why a more and more significant amount of research is carried out on related topics [2-6]. Starch-based composites Various reinforcing natural fibre, such as caraua [3], hardwood (HW) and softwood (SW) [4], silk protein [5], pehuen cellulosic husk [6], date palm and flax [7], and newspaper fibre [8] are mixed with starch-based polymers to increase the modulus or impact toughness and reduced water uptake of the thermoplastic composites. Such composites are completely biodegradable and compostable and therefore considered “environmental friendly” since, at the end of their useful life, they can be discarded without causing any damage to the environment [9]. The improvement of the tensile properties of starch-based biocomposites varies depending on the type and nature of the fibre, its orientation (random or unidirectional), content and form (fibre or fabric) and type of blending/plasticizer used (Critical factors on manufacturing processes of natural fibre composites). Teixeira et al. [10] used plasticized cassava starch as a matrix and combined it with cassava bagasse cellulose nanofibrils as reinforcing materials. Expansion of corn starch based biocomposites reinforced with graft copolymers of Saccharum spontaneum L. (Ss) fibre was achieved by Kaith et al. [11]. Recently, Vallejos et al. [12] applied corn and cassava starches plasticized with 30% glycerin as biometrics and using a fibrous material obtained from ethanol-water fractionation of bagasse as reinforcement. Another relevant research based on starch materials was carried out by Duanmu et al. [13]. They determined moisture absorption, dimensional stability, and mechanical properties of wood fibre reinforced allyl glycidyl ether modified (AGE)–potato starch composites. However, no study has been carried out to evaluate the potential use of sugar palm starch (SPS) as a matrix. Raw woody hemp core (WHC) was recently used in a thermoplastic composite as a reinforcement agent for a starch based biopolymer [14]. The mechanical properties reported for WHC previously may fulfil some technical requirements. A final illustration of the applications of WHC in composites is the styrene-butadiene rubber composites presented by Wang et al. [15]. The authors stated that the mechanical properties of the composite improved with an increasing content of WHC powder, and interestingly, the short WHC fibre has a positive effect on the elongation at break compared with results obtained for long fibre.

Sahari et al. [16] demonstrates the effect of fibre content on mechanical properties, water absorption behaviour and thermal properties of sugar palm fibre reinforced plasticized sugar palm starch (SPF/SPS) biocomposites. The biocomposites were made with various amounts of fibre (10%, 20% and 30% by weight percent) by utilizing glycerol as plasticizer for the starch. It was declared that the mechanical properties of plasticized SPS enhanced with the inclusion of SPF fibre. The tensile strength and modulus of SPF/SPS biocomposites showed increasing trend with increasing SPF while the addition of the SPF made the elongation fall from 8.03% to 3.32%. This was due to the remarkable intrinsic adhesion of the fibre-matrix interface caused by the chemical similarity of starch (SPS) and the cellulose fibre. Water uptake and moisture content of SPF/SPS composites decreased with the characters of fibre, which is due to better interfacial bonding between the matrix and fibre as well as the consumption to absorption caused by the fibre. On the other hand, fractographic studies through scanning electron microscopy (SEM) showed homogeneous distribution of fibre in the matrix with excellent adhesion that play a significant role in improving the mechanical properties of biocomposites. López et al. [17] investigated fully biodegradable composites which were obtained by melt blending of TPS and was blended with PBAT and/or reinforced with cellulosic fibre obtained from recovered newspaper up to 30%. Fibre from recovered newspaper acted as a reinforcing agent, increasing the tensile strength and the Young’s modulus of TPS up to 260%. By opposite, although it helped reducing the water uptake of composites and increased the rigidity of the materials, the incorporation of the highly hydrophobic biodegradable PBAT to the biocomposite formulations caused a sharp decrease in the water uptake of the biocomposites. Nevertheless, even though PBAT has a higher tensile strength than TPS, its incorporation failed to recover the materials. Prachayawarakorn et al. [18] have explored the thermoplastic rice starch (TPRS) reinforcing it with natural silk protein fibre. The effect of silk contents and lengths of properties of the TPRS/silk composites were examined. Internal mixer and compression moulding machine were applied to process and shaped the TPRS/silk composites. It was determined that stress at maximum load and Young’s modulus of the TPRS/silk biocomposites significantly increased with the incorporation of silk fibre. Furthermore, mechanical and thermal properties including water absorption of the TPRS/silk and TPRS/cotton composites were comparable.

**1.2. Poly(hydroxyalkanoate)-based composites**

A variety of fibre materials has been explored to make biocomposites with PHB. It has been shown that the addition of natural fibre increased modulus, Tg, and heat distortion temperature (HDT) of PHB (or PHBV) composites. Nevertheless, the improvements in tensile strength and toughness were found to be difficult and to depend on many factors, such as fibre length and aspect ratio, interfacial bonding, fibre sources, fibre treatments, and fibre forms (single fibre/fabrics). Various co-polymers of PHB have been combined with PALF [19], recycled wood fibre (RWF) [20], empty fruit bunch (EFB) [21], and cellulose nanowhisker (CNW) [22] to prepare biocomposites. These studies have shown that the natural fibre can be embedded in the PHBV matrix as an excellent reinforcer to improve mechanical properties. Table 8 gathers the reported tensile, mechanical properties of PHA based biocomposites. One of the main advantages of PHA for biocomposite applications is their polar character since PHA show better adhesion to lingo−cellulose fibre compared to conventional polyolefins [23].

Taha and Ziegman [24] prepared randomly oriented hemp/Biomer P226 composite specimens by extrusion and tested them in tension and flexure. The highest material properties were measured at 20% volume fraction. The tensile modulus, strength, and elongation were 2.4 GPa, 21.4 MPa, and 4.20%, respectively. The addition of hemp fibre increased the modulus but lowered the strength 40 and elongation relative to the pure polymer. At 20% volume fraction, the flexural modulus and strength were 3.7 GPa and 45.8 MPa, respectively. Wong et al. [25] investigated the use of plasticizers in flax-PHB composites. A weighed amount of flax fibre was added to neat plasticizer followed by heating to 120oC for eight hours. After heating, excess plasticizer was removed by washing the fibre with acetone and the residual solvent was removed under vacuum. Flax−PHB composites with 1:1 fibre/polymer volume ratios were then made by dissolving PHB in chloroform and mixing plasticizer-modified fibre into the solution. Sample bars were annealed at 70oC for three hours to allow PHB to crystallise and interact with the fibre. As a control material, PHB containing 4% v/v plasticizer without added flax was prepared and subjected to the same treatment as the PHB-flax composites. The mechanical properties biodegradability of wood fibre reinforced BiopolTM composites were investigated by Peterson et al. [26].

A significant finding was that the wood fibre-Biopol composites were highly biodegradable, often degrading faster than pure Biopol specimens. Statistical analysis of composite tensile results showed which conditions of pressure, temperature, heating time and time of pressing gave the best strength and modulus results. Process temperature had the greatest influence on composite tensile strength with values varying from 23 MPa at 219oC to 17 MPa at 240oC. The tensile modulus varied from 2.9 GPa to 2.15 GPa over the same temperature range. Javadi et al. [27] incorporated recycled wood fibre (RWF) and synthetic filler like nanoclay into the PHBV/PBAT blend to provide hybrid biocomposites with excellent material properties. To provide good interfacial adhesion between hydrophilic wood fibre and hydrophobic PHBV, the RWF were pre-treated with silane. Salim et al. [28] evaluated properties of biocomposites from oil palm empty fruit bunch (EFB) fibre blended with poly(3-hydroxybutyrate-co-38 mol%-3- 41 hydroxyvalerate) [P(3HB-co-38 mol%-3HV)]. The authors used maleic anhydride (MAH), and benzoyl peroxide (DBPO) to enhance the miscibility between PHBV and EFB fibre. On the basis of this study, the possibility of utilizing biodegradable PHBV/EFB blends showed fewer chemicals leached compared to commercial packaging. Wu and Liao [29] investigated the mechanical and biological properties of composite materials produced from chestnut shell fibre (CSF) and PHA, as well as CSF and glycidyl methacrylate grafted PHA (PHA-g-GMA). Biocomposites developed from PHA-g-GMA/CSF were found to have noticeable features of mechanical properties compared with those of PHA/CSF. CSF could be homogeneously dispersed in the PHA-g-GMA matrix as a result of condensation reactions. Moreover, the PHA/CSF and PHA-g-GMA/CSF biocomposites were more biodegradable than pure PHA. Ten et al. [30] successfully demonstrated for the successful orientation cellulose nanowhisker (CNW) in the PHBV matrix using an electric field. PHBV films with 1.5-7% of CNWs were produced using solution casting. A high degree of fibre orientation was found under the electric field of 2 kV/cm and above. Rheological results confirmed the enhance restraints. In another study by the same authors, the reason was attributed to the PHBV/CNW network structure in the rheological tests which were formed without physical touching and geometrical overlapping between nanotubes [31].

***1.3. Poly(lactic acid)-based composites***

Several researches have been performed to examine the possibility of realizing PLA-based composites reinforced with natural fibre with enhanced final properties. The fillers used include cotton linters maple hardwood fibre [32], sweet sorghum fibre [33], bamboo fibre [34], ramie fibre [35], and lyocell fibre [36]. Most of these studies show that it is possible to achieve various degrees of improvement in tensile and flexural strength and significant increases in modulus with the creation of the respective composites. A number of biocomposites systems prepared by different techniques based on PLA as the matrix are listed in Table 1.



It has been reported that tensile and flexural properties improved by increasing the content of cellulose fibre reinforcements in PLA-based composites. More recently, Liu et al. [37] proved that the basalt fibre with the adequate sizing can significantly increase tensile, bending and impact strength of PLA. By using 20% of basalt fibre and 20% of ethylene-acrylate-glycidyl methacrylate copolymer (EAGMA) it was possible to further increase the unnotched impact strength of PLA from 19 kJ/m2 to 34 kJ/m2. Finally, it was stated that there is a strong connection between the phases, due to the absence of gaps at the fibre-matrix interface; however, the surface of the fibre was still poorly wetted by the matrix. Kurniawan et al. [38] treated the surface of the basalt fibre by using atmospheric pressure glow discharge plasma polymerization and analyzed the adhesion between the basalt fibre/PLA in hot pressed composites. It was demonstrated that by increasing the plasma polymerization time above 3 min it was possible to increase the tensile strength of the composites above the own strength of PLA. The plasma polymerized fibre was well wetted by the PLA on the electron microscopic micrographs. Our previous study by Jandas et al. [39] examined that the PLA/banana fibre (BF) biocomposites by melt mixing and compression molding. Preliminary results showed that the properties of PLA and BF fibre composites are promising. Before composites fabrication, the BF fibre was chemically modified using acetic anhydride, (3-aminopropyl)triethoxysilane (APS), bis-(3-triethoxysilylpropyl)tetrasulfide (Si-69), and mercerized using NaOH solution to improve the compatibility with the PLA matrix. This study concluded that the morphological analysis highlighted a very significant enhancement of the surface wetting as well as on the adhesion level. Song et al. [40] focused on the physical behaviour of hemp/PLA composites, particularly the thermal properties and viscoelastic behaviour. They used twill and plain woven hemp fabrics as reinforcements by stacking film method. The impact and tensile properties of PLA resin reinforced with twill structure fabric were found 15% and 10% higher than the plain woven respectively. Increment of fibre volume fraction from 6 to 20% was the reason why the coefficient of thermal expansion of the fabricated hemp fabric composites decreased considerably (from 70 × 10-6 m/oC to 10 × 10-6 m/oC), which indicates that the composites have great potential for parts experiencing a wide range of temperatures, such as automobile and aerospace applications.

Goriparthi et al. [41] developed a biodegradable composite using jute fibre and PLA, but their focus was to improve the adhesion of jute fibre by surface modification using alkali, permanganate, peroxide and silane treatments. Combination of the special prepreg fabrication method along with surface treatment on their composite sample exhibits some enhancement at least 45% on the mechanical properties. Ma and Joo [42] also studied the effect under different fabrication conditions, such as fibre content (5, 10 and 15%), processing temperatures (200, 210, and 220 oC) and alkali treatment (6, 8, 10, and 12%) on the mechanical properties and structure of jute fibre with PLA using film stacking method. In the designed experiment, the optimum mechanical properties of composites (improvements up to 76%) can be obtained by using 15% of jute fibre treated with 12% of NaOH for 8 h with a processing temperature of 2010 oC. The modified rule of mixture equation was also developed and confirmed. It is more suitable to predict the tensile modulus of jute fibre reinforced biocomposites. Huda et al. [43] have studied PLA/recycled cellulose composites prepared by 44 extrusion and injection moulding, finding that the filler (up to 30%) significantly improved the rigidity without affecting the crystallinity degree and thermal stability.

***1.4. Soy resin-based composites***

The soy-based biocomposites reinforced with natural fibre were found to have better properties than PP composites with natural fibre [44]. Liu et al. [45] reported the kenaf fibre reinforced soy-based biocomposites were fabricated by extrusion/injection moulding and compression moulding. The compression-moulded samples showed high thermal and mechanical properties, and the modulus, impact strength, and heat deflection temperature values of the biocomposites increased as the fibre length, fibre content, and fibre orientation increased. Researchers recently investigated the effect of stearic acid on tensile, and thermal properties of ramie fibre reinforced SPI resin green composites [46]. It was noted that part of the stearic acid crystallized in SPI resin and that the crystallizability was transformed by the addition of glycerol as a plasticizer. The fabricated green composite was found to have enormous potential for certain indoor applications. Jute fabric was used by Huang and Netravali [47] to reinforce concentrated soy protein (SPC) modified with glutaraldehyde and nonocaly. During composite fabrication, they used metal frame to wind the hemp yarns and applied small tension to minimize the yarn shrinkage and misalignment during the drying of the resin. The unidirectional flax yarn strengthened SPC composites proved longitudinal tensile failure stress of 298 MPa and Young’s modulus of 4.3 GPa. The flexural stress was 117 MPa, and the flexural modulus was 7.6 GPa. All those results show that the flax yarn reinforced SPC has the potential to replace non-biodegradable materials in many fields. Unlike reference [48], Behera et al. [49] used woven and a non-woven jute in the different weight percentage (40−80%) to reinforce soy resin (soy milk) based resin. They found that both woven and non-woven jute composites, composite having 60% jute felt showed highest mechanical properties. Tensile strength and tensile modulus were recorded 35.6 MPa and 0.972 GPa for woven, 37.1 MPa and 1.04 GPa for the non-woven jute composites respectively. Flexural strength and modulus were documented 33.5 MPa and 1.02 GPa for woven, 38.4 MPa and 1.12 GPa for the non-woven jute composites respectively. Furthermore, both kinds of fabric are different in structure, and their comparison cannot be justified. Reddy and Yang [50] fabricated environment-friendly biocomposites, used jute fibre to reinforce soy protein resin using water without any chemical as plasticizer. Their composite was fabricated using the prepreg method and they found that biocomposites have excellent flexural strength, tensile strength and modulus in comparison to jute/PP composite. Hydrogen bonding interaction is often insufficient to ensure adequate mixing of lignin with protein. Huang et al. [51] blended kraft lignin with soy protein using methylene diphenyl diisocyanate (MDI) as a compatibilizer. The compatibilization occurred through MDI and hydroxyl groups of lignin and protein molecules. Only a slight reduction in water absorption was observed, but the addition of 2% MDI caused a simultaneous enhancement of tensile strength, modulus, and elongation at break of the polymer blends, which was attributed to graft copolymerization and crosslinking. The characterization of Phytagel® modified SPI resin and unidirectional flax yarn reinforced biocomposites was undertaken by Lodha and Netravali [52]. The incorporation of Phytagel® in SPI resin led to an overall ten-fold increase in the tensile fracture stress and a nine-fold increase in Young’s modulus of the soy protein resin along with a seven-7-fold decrease in fracture strain. The dynamic mechanical properties, such as loss and storage modulus of the modified resin increased and the Tg also increased by about 56oC. Table 8 shows the mechanical properties of soy oil based natural fibre composites manufactured using different processing techniques.

***1.5. Wood-Plastic Composites (WPCs)***

In recent years, wood fibre/flour (WF) reinforced biocomposites, named WPCs, play a vital role in the social, economic and environmental growth of human history [53-56]. The WPCs show characteristics, such as moderate strength, high durability, and light weight. Besides, they are inexpensive and sustainable, which give them attraction for innovative design [57-60]. WPCs refer to materials or products consisting of one or more lignocellulosic fibre/flours and one or a mixture of polymers. Various wood and natural fibre have been used in the processing of WPCs, for example, hemp [61-63], cellulosic fibre [64] and flax [65]. Recently, there has been a commercial interest in the use of thermo-mechanical pulp (TMP) fibre for manufacturing of WPCs [66-70]. According to Ashori et al. [71], WPCs are based on plant fibre (wood and nonwood) and thermosets (epoxy and phenolic resins) or thermoplastics, such as PE, PP, PS, PVC, and PLA [72-75]. Additionally, additives, such as coupling agents, colorants, stabilizers, blowing agents, reinforced agents, and lubricants could be applied to improve properties [76]. The characteristics of wood fibre depend primarily on their source. The average lengths of soft and hard wood fibre are 3.3 and 1.0 mm, diameters 33 and 20 µm, and strength 100−170 and 90−180 MPa, respectively [77]. The main applications of WPCs are initially used for construction applications (decking, docks, landscaping timbers, fencing) and non-structural applications, but now they have been widely used for a broader range of applications, including automotive, gardening and outdoor products [78]. According to the study by Markarian et al. [79], the WPCs market, including thermoplastics and thermosets, has been estimated globally at 900,000 t, in which, 70% of this volume was consumed by North America, 20% in Europe, and 10% from Asia. In Europe, the market showed growth rates averaging 23% per year from 2003 to 2007 and predicted to continue at 26% per year from 2012. Properties of WPCs depend on many factors like, interaction between wood filler and matrix, including matrix characteristics, 47 chemical and physical characteristics of the wood filler and processing conditions. On the other hand, the components of WPCs, contain cellulose, hemicellulose and lignin, in which the hydroxyl groups build plenty of hydrogen bonds between the macromolecules of the wood polymers [80]. The hydroxyl groups could form new hydrogen bonds with water molecules, which induce the water absorption, creation of micro-cracks in the sample, resulting in reduced mechanical property because stress could not be transferred efficiently from the matrix to the fibre [81,82]. In order to overcome this disadvantage, different methods are used for the enhancement of mechanical properties and water resistance of WPCs. Some of them are focused on processing the WF, such as MAH grafted copolymer of the matrix polymer [83], silane treatment [84,85], heat treatment [86,87], and treatment with sodium hydroxide [88]. Besides, the inclusion of nanoparticles as reinforcing filler is another technique for improving the overall characteristics of lignocellulosic-thermoplastic composites, such as silica nanopowder [89] and montmorillonite clay [90]. However, these methods have their weaknesses, such as complicated processing processes and easy aggregation of nanoparticles that caused poor dispersion in the polymer matrix and a limited increase in overall properties of composites. The reinforcement capability of wood fibre in various matrix polymers has been studied earlier. Maldas et al. [91] investigated the effect of different wood species on mechanical properties of PS composites. They concluded that the softwood pulps provided better reinforcement for the polymer than hardwood pulps, which is due to the difference in morphology, density and aspect ratios of the wood fibre types.

Bhattacharyya et al. [92] showed that wood fibre with PP composites is indeed formable. The sheets they manufactured used pinus radiate fibre along with PP powder, for a total through thickness of 1.3 mm. Two types of composites were produced, 48 layered and homogeneous with PP and wood fibre mixed during formation. Results exposed a tensile modulus increase of upto 250% with a 25-30% fibre volume fraction. This study found that the ability to form these composites into not only two-dimensional shapes, but also simple three-dimensional shapes [93]. However, the impact performance of PLA-based WPCs is not sufficient due to the inherent brittleness of PLA. After being toughened with the impact strength of the resulting PLA-based WPCs is significantly improved. However, environmental impacts of the PLA-based WPCs must be evaluated before they are used as engineering materials in the near future. Bolin and Smith [94] found that the environmental impacts from lumber treated with alkaline copper quaternary (ACQ) were 14 times less for fossil fuel use, potential smog emissions, almost 3 times less for greenhouse gas emissions, and water use, 4 times less for acidification, and since half for ecological toxicity compared with those from WPCs decking. For eutrophication, the environmental impacts were approximately equal. Stark and Rowlands [95] used TMP fibre in their investigations, however, without varying the processing conditions during refining. Wood chips were processed in a laboratory-scale of 300 mm single-disk Sprout-Bauer pressurized refiner with incoming steam pressure of 414 kPa and constant plate gap wide of 0.127 mm. As a result, mechanical properties of the composites were well developed when using wood fibre instead of wood flour. Migneault et al. [96] compared injection moulding (IM) and extruded WPCs made with different fibre length-to-diameter (L/D) ratio of chemo-thermo-mechanical pulp (CTMP) with a high-density polyethylene (HDPE) matrix. The IM process appeared in greater composite physical and mechanical properties than the extrusion process, but higher density was achieved with the extrusion process. This study provides a good understanding of the connections between processing method, fibre characteristics, and composite performance. In the same manner, the 49 author also investigated the effect of white birch pulp fibre with different aspect ratios (length-todiameter), HDPE, and using two standard processes: extrusion or injection moulding [97].

***1.6. Applications***

***1.6.1. Natural fibre for biocomposites***

Lignocellulosic natural fibre Natural fibre is subdivided, based on their origins, viz. plants, animals, or minerals. All plant fibres are composed of cellulose while animal fibres consist of proteins (hair, silk and wool). Plant fibres includes bast (or stem or soft sclerenchyma) fibre, leaf or hard fibre, seed, fruit, wood, cereal straw and other grass fibres. A diagram with a classification of the various fibres is shows in Fig. 1.



**Fig. 1 Classification of Fibers**

Over the last few years, a number of researchers have been involved in studying the exploitation of natural fibre as composite materials [97]. Lignocellulosic fibres have three primary chemical constituents, namely cellulose (α-cellulose), hemicelluloses and lignin. In addition, lignocellulosic fibres also contains minor as amount of pectin, waxes, and water-soluble substances [29]. The general chemical structures of cellulose, hemicelluloses, lignin and pectin are shown in Fig. 2.



**Fig. 2 General chemical structures of cellulose, hemicelluloses, lignin and pectin**

The lignocellulose fibres compositions (Table 2) and structures vary greatly, depending on plant species, age, climate, and soil conditions. The information on the chemical composition of lignocellulosic fillers and fibres is important because it determines their properties and thus their applications. An essential part of all lignocellulosic fibres is cellulose.

**Table 2 The chemical composition of several lignocellulosic fibers**



Cellulose itself is a linear macromolecule consisting of β−D-glucose units, linked together by β−1,4−glycosidic linkages at C1 and C4 position to generate a linear polymer chain [29]. Cellulose is a polysaccharide (C6H12O5)n which can be degraded to yield glucose (C6H12O6). The shortest repeat unit is cellobiose (C6H11O5)2O developed by the condensation of two glucose units and hence is also known as anhydrocellobiose as shown in Fig. 7a. Each repeat unit contains six hydroxyl groups, which form intermolecular and intramolecular hydrogen bonds with the macromolecule itself and also with other cellulose macromolecules or polar molecules [2]. Therefore, all lignocellulosic fibres are hydrophilic in nature. The degree of polymerization of cellulose varies according to the plant species (7000-15000). Cellulose macromolecules exist in tightly packed; slender rod-like crystalline structures called microfibrils. The remaining portion that has a lower packing density is referred to as amorphous cellulose [98]. The cellulose chains agglomerate to ~3-5 nm thick (microfibrils) with crystalline and non-crystalline regions. It is water insoluble but recalcitrant to hydrolysis because of its densely packed highly crystalline structure with straight, stable supra-molecular fibre of high tensile strength and its low accessibility [2,20]. The hydroxyl groups of the glucose residues from one chain form hydrogen bonds with the oxygen 22 molecules on the same or a neighbor chain, thereby holding the chains firmly together and forming microfibrils that make the recalcitrant compact structure [20,98]. Hemicellulose the second most abundant organic material on earth after cellulose, it was assumed that the moisture of the hemicellulose is 2.6 times higher than lignin [29]. Hemicellulose is structurally more complex than cellulose, which contains only 1,4-β-Dglucopyranose linkages (Fig. 7b) [97]. It is bound to the cellulose and lignin component by covalent and non-covalent bonds in the cell wall, the interactions between these components increase the rigidity and flexibility of fibre cell wall. Hemicelluloses are a group of homo- and heteropolymers consisting mainly of anhydro-β-(1-4)-D-xylopyranose, glucopyranose, mannopyranose, and galactopyranose main chains with a number of substituent [97]. In addition, the degree of polymerization of native cellulose is about 10–100 times greater than that of hemicellulose. Although the chemical bonds between cellulose and hemicellulose are not covalent, they are still very strong and difficult to separate. The major hemicellulose components in softwood are mannan-based and those in hardwood xylan-based. Hemicelluloses constitute on average about 26% of hardwood, 22% of softwood, and 30% of various agricultural residues [19,97].

Lignin is defined as polymeric natural products arising from an enzyme-initiated dehydrogenative polymerization of three primary precursors: trans-coniferyl, trans-sinapyl, and trans-p-coumaryl shown in Fig. 7c [19]. The only difference between the precursors is the number of methoxyl groups (−OCH3) present on the aromatic ring. Lignin fills the spaces in the cell wall between pectin, hemicellulose, and cellulose. Lignin is a three-dimensional (highly branched) polymer composed of aromatic units with strong intramolecular bonding [19]. Lignin 23 chemical substituents of the backbone of the hemicellulose, such as arabinose, galactose, and 4- O-methylglucronic acid, are covalently linked with lignin. Due to its insolubility in water, amorphous, hydrophobic binding capacity, and cannot be broken down into monomeric units. The hydrophobic lignin system affects the properties of another network, in a way, that it acts as a coupling agent and improves the stiffness of the cellulose/hemicellulose in the plant [99,100]. It has been shown that lignin possesses a softening temperature of about 90oC and a Tm of 170oC. Pectin plays an important role in the natural fibres as a component that binds the fibres into bundles and also determines the luster and touch of the fibres. Pectins are present in plant tissue to varying degrees, being found predominantly in fruit peel and gums [19]. Unretted flax contains between 3 to 4% pectins [29]. In flax, pectins are to be found in the cells surrounding fibre bundles, especially those separating from core tissue of plant. The high concentration of pectins occurs in the primary cell wall and middle lamella of fibre. Here, in conjunction with hemicelluloses, their function is that of a cementing material. Because of their importance as a binding material, removal of pectins during the retting process is particularly important in the production of technical fibre. Pectins are a family of compounds that in turn make up a larger family of pectic substances including pectins, pectic acids, and pectinic acids. Pectin is both a dietary fibre and functional fibre. Pectinic acids represent polygalacturonic acids that either are partly esterified with methanol or have no or negligible amounts of methyl esters [29]. Pectins typically represent a complex group of heteropolysaccharides called galacturonoglycans, which also vary in methyl ester content. The simplest pectin is homogalacturonan (HG), an unbranched polymer of α-(1→4) linked D-galacturonic acid as shown in Fig. 7d. The predominant galacturonan backbone of pectin is, however, modified by the insertion, at intervals, of 24 α−L−rhamnopyranose units [101]. Rhamnogalacturonan I (RGI) is a complex pectic polysaccharide with a backbone composed of alternating α-(1→2) linked L-rhamnose-α-(1→4) linked D-galacturonic acid. Rhamnogalacturonan II (RGII) is a highly branched pectic polysaccharide with a backbone made of (1→4) linked galacturonic acid residues. RGI has been suggested to function as a scaffold to which other pectins, such as RGII and HG, are covalently attached [101]. Waxes are also important in terms of technological parameters. They determine soft touch, low friction and thus the case of moving the fibre [2,19]. In flax fibre, waxes are present mainly in the outer part of the stem, in the epidermis, and in smaller amounts in fibre cells. Waxes contain different types of alcohols that are soluble in water as well as in acids, such as phenolic, oleaginous and stearic acid. Globally, the specific tensile strength of plant fibre is between 1600 and 2950 MPa, during their specific strength modulus varies from 10 to 130 GPa. The Young’s modulus of cellulose is about 140 GPa while that of hemicelluloses has been estimated at 8 GPa [100], both varying according to the relative humidity. It is also well known that lignin increases the stiffness of the fibre structure by acting as a compatibilizer between cellulose and hemicellulose [28,2].

Surface modification considering that fibre usually does not have good adhesion with polymer matrices due to their hydrophilic nature. In order to overcome this issue, a number of surface modification methodologies have been attempted. Such modifications not only improve their wettability with the polymer matrices, but also reduce the moisture absorption, sometimes impart unique 25 properties and ease of processing. Surface modification typically involves one of four methods, chemical (mercerization, acetylation, silane treatment, etc.), physic-chemical (solvent extraction), physical (use of different rays or plasma, steam explosion) and mechanical methods (rolling, swaging) attempted by many researchers.

Physical modification of natural fibre Physical methods involve treatment by corona-discharge treatment [102], physic-chemical ones, such as steam explosion treatment [103], high energy ray radiation processing [104] and autoclave treatment [105]. All these techniques aim to improve the fibre-matrix adhesion by reducing the difference between hydrophilic/hydrophobic characters of fibre and the matrix [102]. It is noteworthy that physical treatments appear as the most eco-friendly ones. Plasma treatment offers a unique approach to modifying the chemical and physical structures of both fibre and polymeric surfaces without altering the bulk structures and characteristics of resulting materials [106]. Plasma treatment is mainly applied for the cleaning, sterilization and surface etching of the films in food packaging application. In fact, surface hydrophilicity and adhesion ability of the films increase dramatically after plasma treatment because polar groups are formed on film surfaces. Therefore, further modification is possible through assembling hydrophilic substances with antimicrobial abilities onto the surface of the treated films. On the other hand, steam explosion results in improved properties of lignocellulosic materials, which include reduced stiffness, smoother surface, improved bending properties, and better distribution [107,108]. Steam explosion process, a high-pressure steaming involves heating of lignocellulosic materials at high temperatures and pressures followed by mechanical disruption of the pretreated material by violent discharge (explosion) into a collecting tank. This process has been applied to many lignocellulosic materials to enhance dispersibility and adhesion with the polymer matrix [108]. In an innovative study, interfacial properties and microfailure degradation mechanisms of the oxygen plasma treated biodegradable poly(p-dioxanone) (PPDO) fibre/PLA composites have been reported by Park et al. [26]. The interfacial shear strength (IFSS) between plasma-treated PPDO fiber and PLA was improved by induced polar functional group and showed twice the improvement with respect to the untreated case. Scheme 1 shows the possible chemical interaction between plasma−treated PPDO fibre and PLA matrix. Oxygen-based functional groups were produced on the surface of PPDO fibre by oxygen plasma treatment. Primary covalent and secondary hydrogen bondings enhancing interfacial adhesion could occur between hydroxyl and carboxyl groups on the surface of PPDO fibre and hydrogen in PLA matrix. Yuan et al. [109] investigated the effect of air plasma and argon treatment on IFSS between sisal fibre and PP by means of single fibre pull-out test. They found that optimum treatment parameters were shortest plasma treatment time, medium power level, and medium chamber pressure. Under these conditions, the IFSS of air plasma treated sisal fibre were found to be higher than argon plasma treated fibre. Sinha and Panigrahi [110] have studied a low-pressure argon plasma treated jute fibres. The fibres were treated in argon plasma for 5 min, 10 min and 15 min, respectively. With longer plasma treatment, the fibre became rougher, and the formation of pits on the fibre surface was observed when the fibre were plasma treated for 15 min. Ragoubi et al. [102] have applied a corona discharge on miscanthus fibre and they have observed a chemical oxidation as well as physical etching inducing an increase of mechanical properties of miscanthus/PLA and miscanthus/PP composites.

Chemical modification of natural fibre Chemical modifications was applied to natural fibre in order to improve the matrix–fibre adhesion. Some chemical modification could lead to reduced moisture absorption of natural fibres and their composites. Most of the chemical modifications of natural fibre involve silanization, alkalization (mercerization), acetylation, cyanoethylation, benzoylation, isocyanation, dewaxing, esterification, etherification, and graft copolymerization. Other modifications of natural fibres include crosslinking with formaldehyde, p-phenylenediamine and phthalic anhydride; nitration; dinitrophenylation and transesterification [112,113]. Their chemical composition allows them to react with the fibre surface, which forms a bridge of chemical bonds between the fibre and matrix. Schematic presentations of some of these surface chemical modifications of natural fibre are shown in Scheme 2. Some of the most effective chemical modification strategies are briefly described as follows. Alkaline treatment or mercerization is one of the oldest, cost effective and most used chemical methods for natural fibre when they used to reinforce thermoplastics and thermosets. Its efficiency depends on the type and concentration of the alkaline solution, time of treatment, and the temperature used for modification [112]. If the alkali concentration is higher than the optimum condition, the excess delignification of the fibre can take place, which results in weakening or damaging the fibre. This treatment removes lignin, hemicellulose, wax and oils covering the surface of the fibre. Addition of aqueous sodium hydroxide (NaOH) to natural fibre promotes the ionization of the hydroxyl group to the alkoxide natural fibres (fibre−cell−O−Na) [22]. The chemical reaction of the fibre−cell and NaOH is represented in Scheme 2. The development of a rough surface and the enhanced aspect ratio result in a better mechanical interlocking that induces as improved the fibre−matrix interfacial adhesion in the resulting 28 composites. Several studies conducted on alkali treatment of natural fibres and they reported that the mercerization led to an increase in the amount of amorphous cellulose while reduce the hydrogen bonding intensity [22].

The effects of alkali treatment of pineapple leaf fibre on the performance of pineapple leaf fibre/PLA biocomposites have been shown by Huda et al. [21]. It was found that the alkali-treated fibre reinforced biocomposites offered superior mechanical properties compared to untreated fibre biocomposites. This study also suggested that the appropriate modification of natural fibre surface significantly contributes to improving the interfacial properties of the resulting biocomposites. Cao et al. [115] investigated the effect of NaOH treatment of bagasse fibre on the mechanical properties of bagasse fibre reinforced polyester biocomposites. Among, the various concentration of NaOH used, superior properties were obtained for the biocomposites made from 1% NaOH treated bagasse fibre. Mishra et al. [116] employed sisal fibre with 5% and 10% NaOH, cyanoethylated and acetylated fibre surface treatment for fabricating of hybrid glass-sisal composite and the result showed that the 5% NaOH solution treated sisal fibres has the best effects on the interface bonding of composite because of higher alkali concentration. Silane is a multifunctional molecule which is used as a coupling agent to modify fibre surfaces. Silane was found to be the most effective among many coupling agents for the natural fibre surface treatment. The uptake of silane is very much dependent on a number of factors including hydrolysis time, organofunctionalized of silane, temperature, and pH. It undergoes several 29 stages of hydrolysis, condensation and bond formation during the treatment process of the natural fibre. After hydrolysis, one end of silanol reacts with the cellulose hydroxyl groups (Si−O−cell−fibre) and the other end reacts (bond formation) with the matrix (Si−matrix) functional groups [15]. After silane modification, hydrocarbon chains allow the fibre to absorb more water, which means that its chemical affinity to the polymer matrix is improved [118]. Goriparthi et al. [119] also studied jute fibre to reinforced PLA composites, but their focus was to improve the adhesion between the fibre-matrix by surface modification of jute fibre in the presence of alkali, permanganate, peroxide and silane treatments. Combination of the special prepreg fabrication method along with surface treatment on their composite sample exhibits enhancement at least 45% on the tensile and flexural modulus. Other authors investigated the effect of silane treatments on non-woven kenaf/PLA composites [120] and the effect of a combined alkali and silane treatment on ramie fibre/PLA composites [121]. They found a significant improvement of the overall mechanical properties, the alkali and combined alkali/silane treatments showed the best results. Acetylation treatment is known as esterification methods for plasticizing of natural fibre. Acetyl group (−CH3COO) reacts with the hydrophilic hydroxyl groups of the fibre and takes out the existed moisture. As a result, hydrophilic nature of the fibre is reduced while improves the dimensional stability as well as dispersion of fibre into polymeric matrices. After acetylation, the moisture regains considerably reduced as the fibre became more hydrophobic due to the substitution of hydroxyl groups with acetyl groups [122]. Natural fibres are acetylated with and without an acid catalyst to graft acetyl groups onto the cellulose structure. In general, acetic acid and acetic anhydride individually do not react sufficiently with the natural fibre. To accelerate the reaction, the natural fibre is initially soaked in acetic acid followed by treated with acetic anhydride between the time periods of 1-3 h with higher temperature. In benzoylation of natural fibres, benzoyl chloride is used for fibre treatment. The benzoyl (C6H5CO) group interacts with the hydroxyl group of natural fibre and thus decreases the hydrophilic nature of the fibre. The introduced benzoyl groups of the natural fibres improve the interfacial adhesion between the fibre-matrix, thereby increasing the performance of the composites.

Manikandan et al. [124] reported that the benzoylation of the short sisal fibre improved fibre–PS adhesion, thereby, considerably increasing the strength of the composite. Among several strategies based on the preparation of effective coupling agents, the additions of a maleic anhydride (MHA) grafted coupling agent provide efficient interaction with the functional surface of the fibre-matrix. The maleic units induce the formation of covalent bonds (via condensation reactions) and physical interactions (via hydrogen bonds) with hydroxyl groups of natural fibre surface. These interactions allow both the reduction of the hydrophilic nature of fibre and the improvement of fibre−polymer compatibility. There are numerous published studies in which the effect of MAH grafting on the mechanical properties of natural fibre has been investigated, and it is impossible to list all of them here. Hence, only a few representative studies are discussed here. Mohanty et al. [125] used MAPP as coupling agent for the surface modification of sisal fibres. It has been found that a 30% fibre loading with 1% MAPP concentration in toluene and 5 min impregnation time with 6 mm average fibre lengths gave the best results. The composite a significant increase in flexural and impact strength was observed with the treated composites. In recent publications, it was revealed 31 that anhydride grafting onto PHB-HV can be achieved by reactive blending and is an effective coupling agent in natural fibre biocomposites [126]. An alternative to chemical and physical methods is represented by the rapidly expanding use of biological agents, such as fungi [127] and enzymes [128]. Biological modifications offer several advantages over chemical and physical methods. They can selectively remove hydrophilic pectin and the hemicellulosic material while requires less energy input. Kardas et al. [129] investigated the effect of four type of enzyme treatments (Lipase A, Lipase AK, Lipozyme, and Esterase) on the micro-topography of polyester fabric. They found that the esterase preparation was the most effective method for producing a more uniform and homogeneous texture of the fabric material. Li and Pickering [130] used chelators and enzymes to separate hemp fibre into individual bundles. They found that the crystallinity and thermal properties improved after separating the bundles. In another study, Pietak et al. [131] investigated the surface wettability of natural fibre using atomic force microscopy and contact angle measurements. They measured an increase in adhesion force for treated samples because enzymatic and chemical treatments (steam explosion) removed the primary cell wall exposing the secondary wall. The same study provides a clear evidence of the hemicellulose removal from hemp fibre after enzymatic treatment. A recent study conducted on bamboo fibre treatment using a number of enzymes (xylanase, cellulose, pectin lyase and laccase) and it revealed that the different systems were effective in improving fibre fineness, presumably as a result of the removal of the more polar hemicellulose fraction as shown in Table 3.. Enzymatic treatments were shown to remove surface impurities of surface the fibre and to improve the division of fibre bundles and increase the fineness as shown by Saleem et al. [132,134] on hemp.

**Table 3 Recent works regarding treated natural fibre reinforced biocomposites**



**1.6. 2. *Manufacturing/processing aspects of biocomposites***

The manufacturing of natural fibre reinforced biocomposites is based, in general, on the technologies used for conventional synthetic fibre composites. Typical processing techniques include extrusion, injection moulding, compression moulding, pultrusion and filament winding, direct long-fibre thermoplastic (D-LFT) is suitable and investigated for natural fibre reinforced thermoplastic biocomposites [28,136]. On the other side, resin transfer moulding (RTM) and sheet moulding compound (SMC) are implemented with thermosets matrices. Processing methods and suitable processing conditions have significant influence on the parameters (moderate temperatures (below 200oC), dispersion, orientation and aspect ratio) that determine the mechanical properties of a natural fibre reinforced biocomposites [137]. Therefore to choose a suitable process to fabricate natural fibre reinforced biocomposites, design, and manufacturing engineers would mainly focus on a number of criteria including desired properties, size, and shape of resultant composites, the production rate, processing qualities of raw materials and the manufacturing cost [138]. Drying of the fibre before processing is necessary because moisture on the fibre surface acts as a debonding agent at the fibre-matrix interface. Additionally, because of water evaporation during the reaction, voids appear in the matrix (thermosets have a reaction temperature above 100oC). Both the aspects lead to a decrease in the mechanical properties of biocomposites. Many studies have been conducted on the potential of using natural fibre as reinforcement for renewable polymers to make biocomposites through injection moulding [139,140]. It is possible to produce complex geometric components with functional elements fast and also in great numbers by injection moulding. There are a number of advantages (minimal warping and shrinkage, economics of scale, high function integration) as compared to compression moulding [140]. The influence of injection moulding process on the mechanical properties of the hemp fibre reinforced PP composites was investigated [140]. Their investigations included fibre treatments and modifications, model predictions of micro-mechanics and strengths, the optimization of hemp fibre quality. Compression moulding is used to produce components at high production volume, and it is suitable to produce automotive components [141]. Thermoset bulk moulding compound (BMC) and SMC are two moulding materials traditionally used in compression moulding [121]. For thermoplastic materials, glass mat thermoplastic is normally a typical moulding compound for compression moulding. High strength manila hemp fibre reinforced starch based biodegradable resin composites have been produced with a hot pressing technique [113]. These preliminary composites were put in a metal mould and heated to 130oC for 5 min with a flexible heater shaped to fit around the metallic mould and hot pressed at 10 MPa for 10 min. The volume fraction of manila hemp fibre in the specimens was varied from 30% to 70% during the processing [142]. The fibre attrition is significant in most extrusion and compounding processes, which often precede injection moulding. The degree of fibre attrition is dependent on initial fibre length, fibre volume fraction and several process variables including screw design, shear rate and melt viscosity. In order to optimize fibre dispersion, there is often some form of mixing required before the final processing of specimens. However, the lack of compatibility between natural fibre and matrix often has more to do with differences in the polarity [138]. Some methods of processing natural fibre biocomposites result in significant fibre attrition. In particular, specimens that undergo mixing followed by a granulation step before being injection moulded shows a significant fibre length reductions [143].

Gatenholm et al. [144] found that in processing PHB with cellulose, the fibre size reduction of cellulose was related to the reduction in molecular 34 weight of the PHB. They proposed that chain scission of PHB was occurring during processing; this resulted in the formation of crotonic acid, which hydrolyzed the cellulose. The result was significant fibre attrition, but also improved fibre dispersion. Karmaker and Youngquist [145] have noted fibre attrition during high shear mixing, such as thermo-kinetic processes used by extrusion and injection moulding. Furthermore, true fibre can have orientations and the ability to overlap and interlock, giving better mechanical properties. However, these factors significantly affect the formability of the composite sheets and cannot be addressed by examining woodflour reinforced thermoplastic composites. In addition to this work, Bhattacharyya et al. [146] have developed thermoforming of woodfibre and PP composite sheets produced without any modification of the fibre or the polymer, the emphasis being on their formability and the associated issues. The authors investigated four thermoforming processes, such as V-bending, die-match forming, air pressure forming and deep drawing, to examine both single-curvature and double-curvature deformation conditions. They also used the technique of grid strain analysis (GSA) which has been applied to quantify the difference in strain distributions during sheet deformation. It was concluded that the properties of woodfibre-PP composite sheets possessed better mechanical, and stiffness properties compared to those of the unreinforced PP. Liquid composite moulding (LCM) is a generic name for similar processes where a liquid resin is transferred into a closed mould cavity filled with dry reinforcement. LCM processes encompass RTM, vacuum assisted resin transfer moulding (VARTM), structural reaction injection moulding (S-RIM), resin infusion under flexible tooling (RIFT) and other subsets where the basic approach is to separately inject the liquid resin into a bed of stationary performs [147]. The successful implementation of RTM involves optimization of numerous parameters. Of these, the mould filling process is related to the perform architecture and permeability, resin 35 viscosity and temperature, gate location and configuration, vent control, and perform placement techniques [148]. The RTM process has become a popular composite manufacturing process due to its capability for high volume production and cost effectiveness. RTM and RIFT are often reported as the manufacturing route for natural fibre reinforced biocomposites [149]. The hemp fibre-unsaturated polyester composites manufactured with RTM process were found to have a uniformity with no noticeable defects [150]. The tensile, flexural and impact properties of these materials were found to increase linearly with increasing fibre content. It was observed that the optimum properties were not reached in this study and that fibre content higher than 35 vol% should yield better mechanical properties. O’Donnell et al. [151] showed how VARTM was able to cure large-volume natural fibre/soybean oil composites at room temperature. Pultrusion is a popular manufacturing method for synthetic fibre and, while it has only recently become a more popular area of exploration for use with natural fibre, it has high potential for use with natural fibre and biodegradable polymers [152]. Angelov et al. [153], and Linganiso et al. [154] reported recently on the pultrusion of natural fibre and different matrices. Depending on the natural fibre used, the profiles exhibited specific properties equivalent to those of glass fibre reinforced composites. This makes natural fibre reinforced biocomposites appropriate candidates for potential applications in the automotive industry [155]. D-LFT processes essentially combine compounding and moulding into an integrated process. In D-LFT systems the moulder can introduce a resin, reinforcement and additives at moulding prior to feeding, directly, to injection or compression moulding for part processing [28]. D-LFT is relatively capital intensive and less common than pre-compounded long fibre granules (LFG). The most common approach of compounded LFGs is polyolefin and polyamide plastics reinforced with long glass (primarily) or carbon (or other) fibre [156,157]. 36 A more elegant method involves spinning of the fibre with the compatibilizer. This avoids metering and handling problems for the mat or composite manufacturer [158]. In addition, MAH grafting onto polypropylene chains (MAH-PP) is the possible way to increase the adhesion between fibre-matrix and later in the composite and, it probably results in completely bonded MAH. The processing of natural long fibre granules by injection moulding (LFG/LFG-IM) results in an easier increase of the fibre-matrix adhesion since compatibilizers can simply be added to the granules. At the same time, the MAH-PP products could be spun into the PP fibre to produce granules that already contain the bonding agent. When natural long fibre granules are used in injection moulding and extrusion-compression moulding (ECM) processes, the fibre length is set by the granule and will decrease in the screw equipment [159]. The degree of the fibre length drop depends on the processing conditions and the screw design. The same is valid for the D-LFT extrusion-compression moulding (D-LFT-ECM). Table 3 shows the processing conditions and the respective fibre lengths resulting from the LFG-IM, LFG-ECM, D-LFT, and CM processes. Injection molded and plasticized made with natural LFGs hardly differ in their tensile properties. The direct process results in lower tensile parameters. The highest tensile and stiffness values are found in composites manufactured by compression moulding of non-wovens or mats. As referred to above, fibre length in a reinforced polymer matrix influences the final performance of a part in terms of stiffness, strength and impact strength [160].

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