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Lignocellulosic Materials for Green Packaging: The Prospects and Challenges

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ABSTRACT: The use of lignocellulosic materials as green packaging materials has garnered attention due to their abundance and renewability. This paper reviews the recent advances in the use of lignocellulosic materials for green packaging by examining the relevant literature published mainly in the last 10 years. Literature search was performed by entering keywords such as biopolymers, lignocellulosic materials and green packaging into scholarly databases, namely Scopus, the Web of Science and Science Direct. This review shows that numerous lignocellulosic materials such as wheat straw, rice straw, pineapple crown and palm fibers have desirable properties as biomaterials. Lignocellulosic materials are versatile and can be used as films, filler in composites, coating and reinforcements in biodegradable foam. They can be converted to cellulose nanofibers which are basically cellulose fibrils incorporated into a learning matrix to provide tensile and flexural properties, as well as cellulose nanocrystals via treatment of cellulosic fiber with acid followed by sonification. Nonetheless, other inherent properties of lignocellulosic materials such as high moisture absorption and incompatibility with other biomaterials limit their use as packaging materials. These limitations prompt the reinforcement of lignocellulosic materials through adding bio-reinforcing agents such as nanoparticles and nanoclay.

KEYWORDS: Biocomposites; biopolymers; cellulose; green packaging; lignocellulosic

INTRODUCTION

The increasing use of conventional non-biodegradable packaging materials particularly plastics has resulted in far-reaching environmental problems especially those related to the mismanagement of these materials (Choong et al., 2020; Wong et al., 2020). Large plastic items for instance, have found their ways into surface water, causing entanglement of aquatic animals and blockage of their alimentary canals upon accidental ingestion (Tang, 2020a). These items could degrade in the environment as a result of photo-oxidative reactions, thermal reactions, mechanical actions or biological actions, producing small fragments of plastics of micro- and nano-scales called microplastics and nanoplastics respectively (Tang, 2020a). These micro- and nano-scale plastics are not well characterized in terms of their ecotoxicology. Studies have pointed to the leaching of a wide range of plastics additives from these materials as well as the adsorption and concentration of other pollutants on their surfaces, which complicate the understanding of their ecotoxicological impacts (Tang, 2020b).

Besides, conventional packaging materials are commonly disposed to landfills where they take up significant land space. Owing to the extremely long time taken for these materials to degrade, they tend to remain in the environment and occupy land space which could otherwise be more productively used, in addition to causing potential land contamination (Tang, 2019; Tang & Angela, 2019; Tang, 2020d). In view of the limited degradability of conventional packaging materials and increasing emphasis on sustainability, the search into biodegradable green packaging materials has garnered attention and much progress has been made in this area (Tang & Al Qahtani, 2020). Green packaging materials are considered sustainable and eco-friendly as they can be readily reused and recycle and their manufacturing utilizes renewable resources (Hao et al., 2019). In fact, a packaging material that meets the requirements of green design and manufacturing principles whose lifecycle creates minimal impacts on the environment could be called a green packaging material (Hao et al., 2019). Green packaging can be manufactured from a wide range of biodegradable materials comprising bioplastics, biomass-based materials, recycled cardboard and paper, and organic fabrics (Karan et al., 2019). Biomass-based materials are derived from plant or animal biomass such as cellulose, starch and protein, or from microbial processes. The non-starch component of the plant biomass is collectively called lignocellulose, comprising cellulose, hemicellulose and lignin (Su et al., 2018) (see Figure 1). These materials invariably contain biopolymers. Biopolymers can be of natural, synthetic or microbial origin and not all of them are biodegradable (see Figure 2) (Moustafa et al., 2019). Examples of natural biopolymers are starch, cellulose, chitin and chitosan while synthetic biopolymers include polylactic acid and polyvinyl alcohols. Microbial biopolymers are derived from microorganisms and are typified by

extracellular polymeric substances and polyhydroxyalkanoates. Bioplastics which have come into the limelight as a potential substitute for conventional plastics could be made of biomass-based materials or other biopolymers. Figure 2 shows the classification of bioplastics (Moustafa et al., 2019).

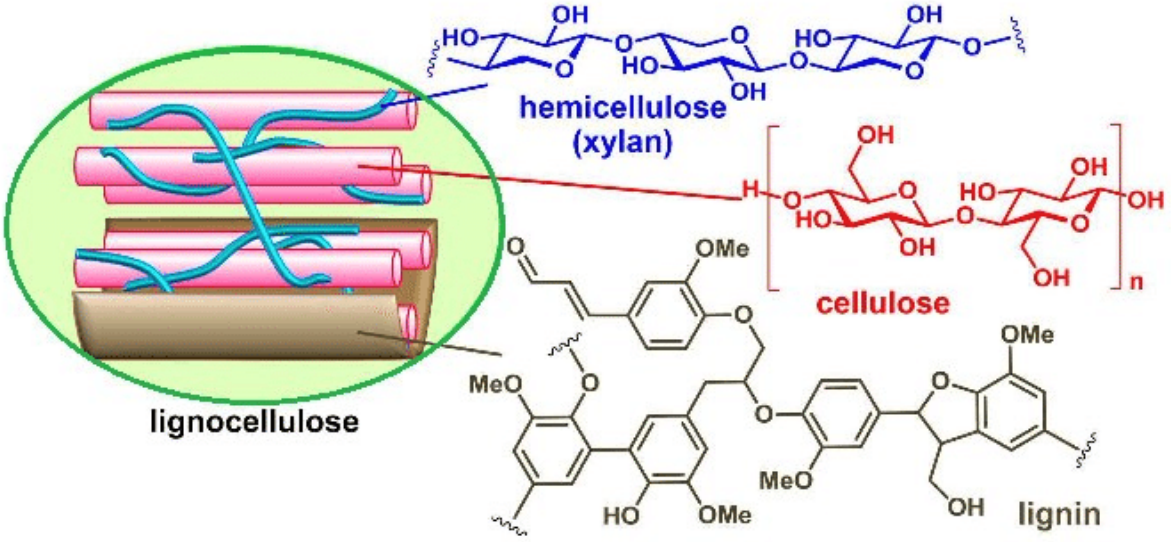

Figure 1. The Structure of Lignocellulosic Materials

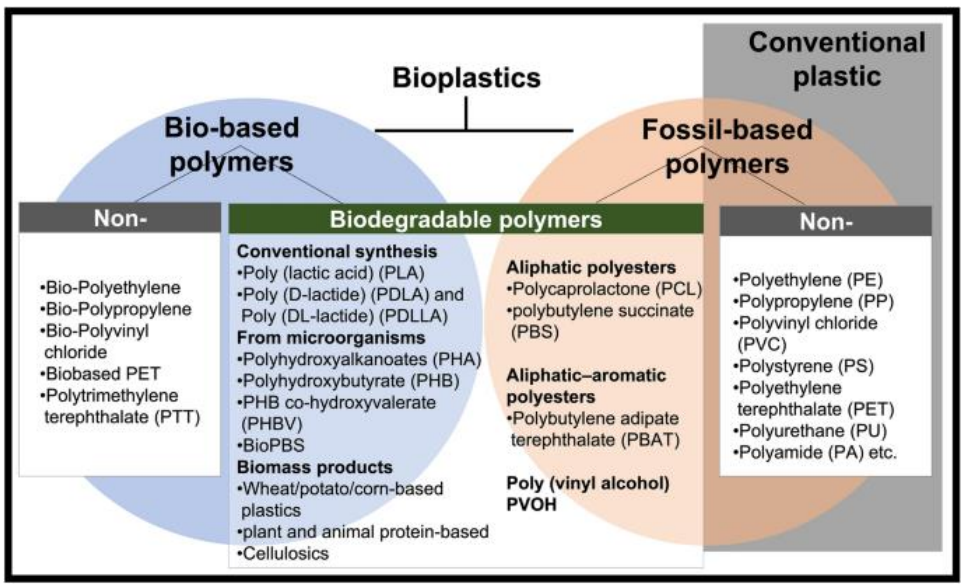

Figure 2. Classifications of Polymers (Moustafa et al., 2019)

In view of the shortcomings of starch-based biomaterials particularly weak mechanical strength and high affinity for moisture, there has been increasing interest in the use of lignocellulosic materials for green packaging. Cellulosic biomass has been manufactured into semi-synthetic films and fibers to replace plastic materials such as polyethylene, polystyrene and polypropylene. As the manufacturing processes require chemical treatment, the processed materials are often considered semi-synthetic (Ferreira et al., 2016). Pure cellulosic polymers are brittle and highly crystalline with poor mechanical properties and moisture barrier (Ferreira et al., 2016). They are mixed with other materials to improve their properties as packaging materials. For instance, cellulose is combined with peptide to form peptidopolysaccharide films which are significantly more effective than pure cellulose film as antimicrobial packaging (Wu et al., 2019). The mixing of lignocellulosic materials with other materials produces three overarching groups of potential green packaing materials, namely biocomposite materials, bionanocomposites and nanopapers (Bhagwat et al., 2020).

Biocomposites comprise biodegradable and often fibrous materials such as cellulose fibers and coffee grounds while nanocomposites are essentially a mixture of biopolymers and nano-fillers of synthetic or inorganic origin, for instance a

mixture of polylactic acid and carbon nanotubes (Bhagwat et al., 2020). These nanocomposites are called bionanocomposites if they satisfy the requirement of being naturally biodegradable (Bhagwat et al., 2020). Nanopapers are formed when nanomaterials such as carbon nanotubes, carbon nanofibers, cellulose nanofibrils and nanoclays are packed into thin sheets, conferring them certain desirable properties like high electrical and thermal conductivity (Huang, 2018). The use of biomaterials particularly cellulose nanofibrils in the synthesis of nanopapers imparts renewability and biodegradability to nanopapers, qualifying them as green packaging materials (Su et al., 2018).

Where advances of green packaging materials are concerned, biocomposites are considered to be ahead of other materials in that there are already commercialized biocomposite packaging materials. Automotive interior components and sports equipment made of Flax composites have already been tested (Park & Park, 2018). Biocomposites could be classified as non-wood and wood fibers, all containing cellulose and lignin. The natural non-wood fibers have greater attraction due to their desirable physical and mechanical properties, long fibers and high cellulose content, which confer them high tensile strength and cellulose crystallinity (Van Hai et al., 2015). With the optimistic potential of lignocellulosic materials for green packaging, this review aims to present the advances made, as well as their challenges and prospects.

METHODS

This paper reviews more than 30 scholarly articles on the use of lignocellulosic materials for the production of green packaging. The articles were searched from journal databases namely Scopus, Web of Science, ScienceDirect and ProQuest with keywords comprising lignocellulosic, green packaging, biocomposites, biofibers, wood fibers and non-wood fibers. The search was limited to literature published in the last 10 years to give an updated overview of the area (Tang, 2020c).

DISCUSSION

Lignocellulosic materials compose of cellulose, hemicellulose and lignin (Su et al., 2018). They have great potential to be converted into green packaging materials. However, on their own, lignocellulosic materials may not have the desirable features of packaging materials such as good mechanical properties and heat resistance. Lignocellulosic materials are processed to convert them into forms that facilitate their further modifications or manufacturing. Examples of the processed lignocellulosic materials are biofibers, nanocrystalline cellulose, micro- and nano-fibrillated cellulose and bacterial cellulose nanofibers. These materials are subsequently modified by addition of other substances to acquire the desired properties for packaging. This gives rise to biocomposites, bionanocomposites and nanopapers (Bhagwat et al., 2020). Taking bionanocomposites for instance, they can be prepared through solvent casting method which involves dispersing nanocellulose within an aqueous or organic medium to form a homogenous suspension at room temperature prior to mixing with polymer solutions. The resultant composites are then dried in a vacuum oven (Bandyopadhyay-Ghosh et al., 2015). Another method of synthesizing bionanocomposites is called melt-intercalation process which involves the use of extruders or mixers. In melt-intercalation, nanoparticles are mixed with a fused polymer and as the temperature of the mixture rises above the glass transition temperature, nanocomposites are formed. Penetration of polymer chains through the reinforcement produces intercalation (Bharimalla et al., 2017). In-situ polymerization has also been employed to produce nanocomposites. It minimizes agglomeration of nanofillers and permits their even distribution in the polymer. This method improves moisture barrier and biodegradability of nanocomposites but is limited to polymerization in liquid phase (Bharimalla et al., 2017). Another method of synthesizing nanocomposites is coating which involve adsorbing composite layers to a substrate or packing them inside two substrates, yielding a thin sheet with thickness from nanometer to micrometer (Li et al., 2019). The thin bionanocomposites are potential substitutes for conventional oil-based plastics films

Table 1. Methods of Synthesizing Bionanocomposites, Their Advantages and Disadvantages

Method	Description	Advantage	Disadvantage
Solvent casting	Mixing of homogenous nanocellulose suspension with polymer solution, followed by drying	• Better dispersion of nanoparticles • Require only small amount of nanofiller to produce nanocomposite films of uniform thickness	• Time- and energy-consuming • Pilot-scale production
Melt-intercalation	Annealing polymer matrix at high temperature, adding nanocellulose and kneading the composite to	• Environmentally friendly as it uses minimal solvent • Lower interfacial	• High temperature used can damage surface modification of nanocellulose

	achieve uniform distribution	tension • Economical	
In-situ polymerization	Swelling of nanocellulose in liquid monomer or monomer solution as monomer penetrates the interlayers	• Better exfoliation • Improved moisture barrier and biodegradability	• Time constraint in executing polymerization process • Expensive
Coating	Synthesis of composites in an aqueous solution containing the polymer and the filler building blocks. The polymer promotes the growth of the filler crystals. The polymer is trapped within layers as the crystals grow.	• Enhanced mechanical strength • Weight reduction • Improved barrier properties	• High temperatures used degrade polymers and promote aggregation of the growing crystals

Cellulose acetate is made of purified cellulose which is treated with acetic anhydride and acetic acid, and subsequently dissolved in acetone before being shaped into fibers through spinnerets (Liu et al., 2021). Cellulose acetate has conventionally been used in the manufacturing of synthetic fibers (M. Gilbert, 2017) and has been classified as 'generally recognized as safe' by the US Food and Drug Administration. It is heat tolerant and is a material found in certain disposable packaging and wrapping (Van den Oever et al., 2017). Plasticizers such as diethyl phthalate and triacetin have been added to cellulose acetate to improve its printability, rigidity and clarity. Cellulose acetate films generally have low tensile strength though it could resist puncturing to a certain extent. They have relatively poor water and gas barrier properties and could readily be hydrolyzed to yield acetic acid (El-Rehim et al., 2018). These shortfalls of cellulose acetate constrain their wider use as food packaging and require combination of other polymers to acquire certain desirable properties such as durability and flexibility. Mixing cellulose acetate with other materials give rise to composites. Its modification with graphene oxide or hydroxide nanoplatelets double layers significantly improves oxygen barrier properties (Helanto et al., 2019). The clay-like nanocomposite cellulose acetate films exhibit better water and oxygen impermeability (El-Rehim et al., 2018). Attempts have been made to mix cellulose acetate, thymol, modified montmorillonite and plasticizers to produce antimicrobial food packaging (Rydz et al., 2018). Melt-intercalation of cellulose acetate and montmorillonite nanoparticles in the absence of plasticizers demonstrates remarkable mechanical property (Ramos et al., 2018). Besides, zinc oxide has been embedded into cellulose acetate nanofibers to confer antimicrobial activity against bacteria such as *S. aureus* and *E. coli*.

Cellophane is a thin transparent film derived from cellulose through viscose method, with glycerin added to improve flexibility (Helanto et al., 2019). Cellophane has been commonly used for bread and cheese packaging or as flower packaging. Folding of cellophane films cannot be reversed and they have low heat-tolerance, which often prompt a separate sealing layer for materials packaged with the films. For instance, amorphous polylactic or starch-based sealing layers are incorporated to cellophane to yield transparent sealable films (Muller et al., 2017). When impregnated with a thin aluminium oxide barrier, cellophane becomes highly compostable. Besides, cellophane is also coated with nitrocellulose wax or polyvinylidene chloride to impart moisture resistance and sealability. The coated cellophane is used as packaging for confectionery and fresh produce (Piergiovanni & Limbo, 2016).

Coffee grounds byproducts are rising green materials with potential uses as biofuels and sources of polysaccharides (Karmee, 2018). They could potentially be used to reinforce biopolymers in the synthesis of affordable green composites but their hydrophilicity is a major constraint to their incorporation into polymer matrices, resulting in their incompatibility with hydrophobic polymers and limitation for bio-reinforcement (Karmee, 2018). To improve their compatibility to polymer matrix, various methods including treating coffee grounds with antimicrobial rosin, adding compatibilizer to polymer matrix and torrefaction involving treatment of coffee grounds at 230-310ºC in inert gas to improve hydrophobicity have been proposed (de Castro et al., 2016). Besides coffee grounds, an array of other lignocellulosic fibers has been investigated. Al-Oqla et al. (2019) studied the mechanical, thermal and fiber-polymer interfacial adhesion characteristics of lignocellulosic fibers derived from olive trees, lemons, loquats and palms. Their study revealed promising use of these fibers as bio-based materials with palm fibers exhibiting the best tensile strength and the elongation to break while lemon and loquat have superior thermal characteristics. Palm and olive fibers possess good thermal stabilities which implicate their potentials for mechanical and electrical applications for

instance as materials for furniture, insulations and circuit boards. Among the fibers tested, loquat fiber was found to have the best interfacial bonding with various types of polymers (Al-Oqla et al., 2019).

Nanocellulosic materials have received much attention recently. A star of nanocellulosic materials is the cellulose microfibrils. Cellulose microfibrils have crystalline and amorphous domains randomly scattered along their length (Su et al., 2015). The crystalline domains are characterized by closely packed cellulose chains while the amorphous domains are auxiliary parts of the microfibrils which are susceptible to chemical or enzymatic attack. Fibrillation of native cellulose fibers is achieved by grinding and homogenization to form microfibrillated cellulose or nanofibrillated cellulose (Su et al., 2015). On the other hand, nanocrystalline cellulose or cellulose nanowhiskers are produced from biomass by enzyme hydrolysis or acid hydrolysis, which removes amorphous domains to form powder, liquid or gel containing pure crystalline cellulose of nanoscale (Eichhorn, 2011). The nanocrystalline cellulose formed has a rigid rod shape with diameters between 1 and 100 nm and lengths ranging from tens to hundreds of nanometers. It exhibits high tensile strength (7,500 MPa), high stiffness (Young's modulus of 100-140 GPa), high aspect ratio of 70 and large surface area (150 – 250 m^2/g) (Eichhorn, 2011). Furthermore, its electrical and optical properties are enhanced. Studies had demonstrated feasible extraction of nanocrystalline cellulose from biomass such as *Phormium tenax* leaf natural fibers and okra bast fibers and cellulose nanocrystals extracted from the latter were successfully incorporated into polyvinyl alcohol (PVA) biodegradable matrix (Fortunati et al., 2012). In a separate study, PVA bionanocomposites were synthesized using cellulose nanocrystals obtained from commercial microcrystalline cellulose originated from *Phormium tenax* and flax as reinforcing materials. Thermal analysis of the bionanocomposites showed that cellulose nanocrystals enhanced the plastic properties (Fortunati et al., 2013).

Unmodified biopolymers usually have poor mechanical, thermal and barrier properties which limit their uses. These shortcomings of synthetic biopolymers such as PVA and polybutylene adipate terephthalate could be overcome with addition of reinforcing materials to attain desirable properties and reduce the production cost. Moustafa et al. managed to enhance the thermo-mechanical and barrier properties of propylene-based elastomer/ polybutylene adipate terephthalate/ ethylene-octene organoclay by adding < 5 wt% of expanded organoclay (Bhagwat et al., 2020). Xie et al. (2018) synthesized polybutylene adipate terephthalate/ organomodified layered double hydroxide films with significantly better water barrier properties than unfilled polybutylene adipate terephthalate films, thus, indicating the potential of the films as food packaging. However, the addition of certain nanomaterials and the modification of synthetic biopolymers as food packaging could warrant safety concerns (Tang, 2020e). Therefore, lignocellulosic nanofibers such as cellulose nanocrystals or cellulose nanofibers derived from natural biopolymers are better options for green packaging. They are made from renewable biomass materials and have good reinforcing capability as well as better mechanical properties compared to inorganic fillers (Su et al., 2018). There has also been mounting interest to look into the antimicrobial properties of green packaging particularly green packaging of natural biopolymers where microorganisms could grow. Attempts have been made to embed antimicrobial peptides, nisin or pediocin into starch-based packaging materials such as starch-halloysite nanocomposite via a casting method (Abreu et al., 2015)(Liu et al., 2017). It was found that the effectiveness of the antimicrobial agents incorporated was affected by the dose of antimicrobial agents and their optimal release over time. Similarly, antimicrobial agents such as thymol and metal oxides has been incorporated into lignocellulosic materials to impart antimicrobial properties. For food packaging, it is crucial that the antimicrobial agents sorbed are not harmful to human and essential oils from natural sources were found to be suitable for such purpose (Liu et al., 2017). Essential oils have been added to thermostarch/ layered silicates bionanocomposites as food packaging (Campos-Requena et al., 2017).

In addition to water barrier, packaging materials for food containing oil or fat should ideally demonstrate grease barrier properties. Studies have been launched into the development of grease-resistant additives for green food packaging. Cellulose fibers with high cohesive energy density, molecular size and ability to form a homogenous and continuous film were identified as a potential candidate (Su et al., 2018). Micro-fibrillated cellulose suspensions were used to coat paper base-stocks exhibiting various permeability to air in producing packaging materials with good oil resistance. The coating was found to lower air permeability and increase oil resistance. Lavoine et al. also found micro-fibrillated cellulose coating was able to increase oil resistance of paper though not as good as polyethylene (Lavoine et al., 2014). A composite coating produced from nanofibrillated cellulose and O-acetyl-galactoglucomannan from spruce wood was found to have excellent grease resistance (Su et al., 2018). Similarly, films developed from nanofibrillated cellulose and chitosan nanoparticles also displayed great grease resistance (Su et al., 2018).

CONCLUSIONS

The review shows optimism in the use of lignocellulosic materials as green packaging materials due to their renewability, biodegradability, abundance and versatility as films, filler in composites, coating and reinforcements in biodegradable foam. However, their inferiority in hydrophobicity, water barrier properties, antimicrobial properties and grease resistance may warrant more studies

into the development of eco-friendly additives to achieve the desirable properties. To increase the potential of lignocellulosic materials as green packaging materials, improvement of the physical, mechanical and antimicrobial properties of the materials while maintaining or improving their biodegradability and compostability is crucial. Besides, cost-effectiveness of manufacturing green packaging materials poses a major barrier to the widespread use of these materials and there is an urgent need for innovation in their processing and manufacturing.

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