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# Antimicrobial poly(lactic acid)/cellulose bionanocomposite for food packaging application: A review



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A R T I C L E I N F O	A B S T R A C T
<i>Keywords:</i> Poly(lactic acid) Nanocellulose Antimicrobial Packaging	Conventional food packaging materials are derived from nonrenewable fossil resources and face difficulties in disposal and recycling. The development of green and ecologically balanced food packaging materials has received much attention as a prospective solution that can partially substitute for the imperishable fossil fuel-derived plastic. Polylactic acid (PLA) is known as a promising biopolymer, and the incorporation of nano-cellulose can further enhance the properties of this biopolymer. The combination of PLA and nanocellulose enables the development of a sustainable eco-friendly food packaging as PLA can be obtained from agricultural resources and nanocellulose can be extracted from agricultural waste. This critical review demonstrates the current developments, and the advantages of PLA and nanocellulose for the purpose of food packaging are introduced. The nanocellulose produced from agricultural waste is summarized along with specific references to food packaging applications. In addition, the antimicrobial agents used in PLA/nanocellulose-based packaging are discussed. Overall, greener food packaging with enhanced antimicrobial properties with a suitable combination of bio-based nanocellulose is highlighted in this review.

#### 1. Introduction

The huge consumption of fossil fuel-derived plastic material has created consistent waste production issues that cause environmental pollution (Han, Yu, & Wang, 2017). In 2015, the global plastic market reached 322 M tons, and approximately 49 M tons of plastics are utilized for packaging purposes. Owing to the inappropriate disposal and nonbiodegradability of these petroleum plastics, 4.8-12.7 M tons of plastics ended up in ocean in the year of 2010, and more than 30% of the plastic waste was land-filled in the year 2014. Consequently, the recycling of plastic has become an encouraging option to abate the accumulation of plastic waste in our environment. However, this recycling system does not work on food packaging systems because of the contamination of organic substances that is found on the food packaging plastics (Ingrao, Gigli, & Siracusa, 2017). Hence, food packaging producers should not only consider the factor of maintaining the effectiveness of the quality of food but also the environmental impact with regards to the disposal of packaging materials. For this reason, biopolymers have received widespread attention due to their biodegradability, and the biopolymers are able to replace the fossil fuel-based plastic materials and address the waste disposal problem (Adilah, Jamilah, Noranizan, & Hanani, 2018; Rhim, Park, & Ha, 2013).

Biopolymers can be divided into four categories: [i] polymers that

have D-glycopyranoside as their building block or repeating unit (e.g., starches, cellulosic and ligno-cellulosic); [ii] polymers acquired through microbial sources (e.g., polyhydroxyalkanoates (PHAs) that include the polymer of poly(hydroxybutyrate) (PHB) and polyglycolic acid (PGA)); [iii] polymers that are chemically synthesized from agroresources (e.g., polylactic acid (PLA)) and [iv] polymers that are chemically synthesized from fossil fuel resources (e.g., polycaprolactones (PCLs) and poly (butylene succinate adipate) (PBAT)) (Elsawy, Kim, Park, & Deep, 2017; Souza & Fernando, 2016).

Among all these biopolymers, poly(lactic acid) (PLA) has been employed for packaging purposes attributable to the ability of this aliphatic polyester to be derived from renewable agricultural resources in the way that its monomer can be generated by the fermentation of agricultural resources (Tiimob et al., 2018). The bio-based character of PLA would contribute to the diminution of ecological impacts correlating with the extensive utilization of conventional plastics derived from nonrenewable fuel sources (Lizundia et al., 2016).

#### 2. Food packaging application

#### 2.1. PLA for food packaging applications

The life cycle production of PLA begins with starch that is extracted

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from plants such as corn. During the growth of the corn, the carbon dioxide is harnessed from the environment for photosynthesis. Throughout the photosynthesis process, consumption of free energy originating from solar energy and carbon dioxide will take place in the corn plant to convert glucose into starch, contributing to chain building for PLA. Therefore, the benefit of PLA production compared with other commercial polymers is the ability to reduce carbon dioxide ( $CO_2$ ) emissions, suggesting that PLA also has the potential to reduce environmental pollution compared to the other conventional plastics (Vink, Glassner, Kolstad, Wooley, & O'Connor, 2007).

The building block for PLA is lactic acid, which can be obtained through the fermentation of maize or other sustainable agricultural raw resources (Abdulkhani, Hosseinzadeh, Ashori, Dadashi, & Takzare, 2014). A few appealing things about PLA are biocompatibility, processability, renewability and less energy consumption (Rasal, Janorkar, & Hirt, 2010). In comparison with biopolymers such as poly(hydroxyalkanoates) (PHAs) and poly(ethylene glycol) (PEG), the good thermal processability of PLA means it can be manufactured through various processing methods such as extrusion, film casting and fiber spinning (Rhim, Mohanty, Singh, & Ng, 2006). Because of the biodegradable and biocompatible characteristics of PLA, it received approval from the Food and Drug Administration (FDA) in the applications with food contact and became an adequate candidate for the application of packaging for fresh food or short service life goods, including overwrapping, lamination filming and blister packaging (Ingrao et al., 2017).

PLA can be available in different molecular weights. However, only PLA with high molecular weight will be selected for use in the packaging industry. There are two main PLA industrial manufacturers, which are NatureWorks LLC and Corbion, and both of these manufacturers are using ring opening polymerization to produce PLA. The ring opening polymerization with rapid reaction time and comparatively mild reaction conditions was able to yield PLA with high molecular weight while having a smaller molecular weight distribution (Hamdan & Sonomoto, 2011). In 2003, massive production of PLA was recorded under the tradename of Ingeo. Approximately 150 M tons of PLA (Ingeo) were manufactured by NatureWorks LLC every year via the process of ring open polymerization (Ingrao et al., 2017).

According to Lovett and de Bie (2016), the Corbion manufacturer was employing the highest yielding feedstock locally available to produce PLA, in such a way that raw sugar originating from cane in Brazil and Thailand, and raw sugar originating from the sources of sugar beets in Spain and the Netherlands are used as a feedstock to generate the monomer of PLA. Instead of using raw sugar, USA is using dextrose from corn to produce lactic acid. By taking a closer look on the feedstock efficiency, PLA can be considered the most efficient biopolymer because the yield of 1 kg of PLA only required approximately 1.6 kg of fermentable sugar feedstock. In comparison with the other biopolymers, approximately 2.5-3 times more sugar feedstock was needed to produce 1 kg of plastic. In addition, by considering the factor of land use, the land use per ton of bio-based polymer from five crops (i.e., maize, wheat, sugar beet, sugarcane and miscanthus) for current agricultural practices was compared in the Lovett and de Bie (2016) study. PLA produced from either sugarcane or sugar beet is an ideal biopolymer that can be used in food packaging applications as it only occupies approximately 0.12 ha/ton (land use per ton), compared to biobased polyethylene (PE), which needs approximately 0.32 ha/ton.

In addition, the unique properties of PLA are comparable to polyolefin and poly(ethylene terephthalate) (PET), and these compounds can be used in a broad range of applications. Overall, the properties of PLA were almost similar to polystyrene (PS) as PLA has a low value for elongation at break of approximately 2–5% and a high Young's Modulus, which is approximately 3 GPa (Spinella et al., 2015). In summary, PLA can be recommended as a good candidate for the application of food packaging, including in its functional properties, for example, high transparency, good barrier properties, good sealability, good oil and grease resistance and excellent organoleptic characteristics (Gruber, 2001).

#### 2.2. Nanocellulose for food packaging applications

#### 2.2.1. Nanocellulose

Nanocellulose can be depicted as a biodegradable nanomaterial because cellulose is sustainable, renewable, recyclable, nontoxic and produces a low carbon footprint (Dufresne, 2017). Cellulose accounts for approximately 35%-50% of the total composition of natural fibers and thus significantly influences the chemical properties of natural plant fibers. Plants produce approximately 75 billion tons of cellulose per year, making cellulose biopolymers inexhaustible (Feng et al., 2018). The general formula for cellulose is  $(C_6H_{10}O_5)_n$ , and cellulose represents the major component in vegetable tissue. In nature, cellulose is seldom available in its pure state. Nonetheless, cellulose is mostly intimately mingled with coloring matter, fats, gums, hemicellulose, lignin, tannins, etc. The 1,4-glycosidic bonds linking the cellulose and  $\beta$ -glucopyranoside residues were used to express the prime structure of cellulose. The cellulose consists of a linear polymer which is known as an anhydroglucose unit. The  $\beta$ -glucosidic bond plays an important role in linking the anhydroglucose units together at carbon atoms one and four. In general, the degree of polymerization of cellulose depends on the extraction source and can be very high (up to 20,000 residues within a chain). Cellobiose residue is recognized as a cellulose repeating unit rather than glucose residue, as the residues within the cellulose have a rotation of 180° relative to each other (Candido, Godoy, & Gonçalves, 2017).

Intramolecular and intermolecular hydrogen bonding is responsible for the stabilization of the ordering of cellulose chains, which in turn leads to the formation of compact microfibrils. The hydrogen bonding is actually responsible for the stabilization of the thermal properties of cellulose since the existence of this bonding prevents cellulose from melting before thermal degradation. Moreover, these bonds cause the cellulose to become insoluble in almost all of the solvents by reason of considerable tensile strength that is imparted by the bonds on the cellulose. In addition, the cellulose can form thin and long fibrils by grouping approximately 100 cellulose chains together, and these fibrils are made up of two parts that are amorphous and crystalline, respectively (de Oliveira, Bras, Pimenta, da Silva Curvelo, & Belgacem, 2016; Hoi & Martincigh, 2013; Moubarik, Grimi, & Boussetta, 2013). The connections for both regions of crystalline and amorphous parts execute important roles as their relationship determines the dimensional characteristics of the crystalline domains as well as the degree of crystallinity of the cellulose (Leão, Miléo, Maia, & Luz, 2017).

Three types of nanocellulose have been utilized as key components for the application of cellulose-derived packaging. These types are cellulose nanocrystals (CNCs), bacterial nanocellulose (BNC) and nanofibrillated cellulose (NFC). Division of these nanocelluloses depends on their size, synthesis method, function and source. The CNCs are the archetypal cellulose that has a low aspect ratio, with diameters of approximately 5 nm and a length of 100-250 nm. The degree of crystallinity in CNCs can be high, and sometimes this degree of crystallinity can achieve 90%. The CNCs mostly can be synthesized by acid hydrolysis. Commonly, sulfuric hydrolysis of cellulose results in cellulose nanocrystals (CNCs) with needle-shaped crystals with diameters within the range of 10-20 nm and the length of approximately 100 nm. CNF usually has a higher aspect ratio and a web-like structure. The diameter of CNF is usually in the range of 5-60 nm, and its synthesis methods include grinding, high pressure homogenizing, high pressure microfluidization and high intensity ultrasonication. For the preparation of BNC, the molecule is normally synthesized by microorganisms, resulting in highly crystalline cellulose with a high degree of polymerization. However, this kind of synthesis method is usually very expensive (Feng et al., 2018).

In comparison with CNFs, CNCs tends to offer better elongation

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