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Article · December 2021

DOI: 10.1016/j.jobab.2021.11.004

Jiaxiu Wang
Anhui University

Kolja Ostendorf
Holzmühle Westerkamp GmbH

Kai Zhang
Georg-August-Universität Göttingen

Journal of Bioresources and Bioproducts xxx (xxxx) xxx

Contents lists available at ScienceDirect

Journal of Bioresources and Bioproducts

journal homepage: http://www.keaipublishing.com/en/journals/journal-of-bioresources-and-bioproducts/

Biobased materials for food packaging

Jiaxiu Wang[a,b,c], Markus Euring[a], Kolja Ostendorf[a], Kai Zhang[a,*]

[a] Department of Wood Technology and Wood-based Composites, Georg-August-University of Göttingen, Büsgenweg 4, D-37077 Göttingen, Germany
[b] School of Life Sciences, Anhui University, 230601 Hefei, China
[c] Anhui Key Laboratory of Modern Biomanufacturing, 230601 Hefei, China

ARTICLE INFO

Keywords:
Food packaging
Nanomaterial
Fiber
Biobased material

ABSTRACT

Consumers prefer foods that are healthier with high quality and safety. Food packaging are demanded to effectively extend the shelf-life, preserve the nutrients and decrease the microbial contamination during the transport and storage of food. With the increasing concern on the environmental impacts caused by food packaging wastes, sustainable and green packaging are highly demanded to minimize the harmful effects of food packaging waste on the environment. Bio-based materials are derived from sustainable and renewable biomass, instead of finite petrochemicals. The applications of bio-based materials for food packaging are highlighted in this review. The emphasis is placed on the categories of related biobased materials, their characteristics and advantages for food packaging, as well as the strategies used to improve their performances. Though a lot of trials have been done on biobased materials for food packaging, further attempts to improve their performances, understand the functioning mechanisms and develop greener methods for the production, processing and destiny of these bio-based materials are still highly needed for the future research.

1. Introduction

The deterioration of food is usually caused by oxidation, microbial spoilage and metabolism, which can be influenced by environmental contamination and other factors, such as temperature, humidity, light, physical damage, microorganism, odors, shocks, dust (Han et al., 2018). Food packaging is used to preserve the quality of food, ensure the food safety, and extend their shelf-life. Different categories of foods have distinct requirements regarding to their storage and transport, for example, the preservation of fruits and vegetables require the reduction of respiration and transpiration rate, which usually can be achieved by the control of humidity, temperature, light, gas (O_2, CO_2, ethylene) environment and so on. Dairy products, such as milk, cheese and cream, should avoid oxidation and microbial growth, and thus the external conditions such as oxygen, light and moisture must be carefully considered. Meat products suffer from the problem of discoloration, which can be prevented via vacuum packaging or modified atmosphere packaging. To guarantee the safety and quality of meat products, the biopolymer packaging, that is environmentally friendly, have been widely used (Chen et al., 2019).

Regarding to the objectives and functioning mechanisms, diverse technologies for food packaging have been developed, ranging from passive packaging, active packaging (AP), through intelligent or smart packaging (IOSP) to sustainable or green packaging (SOGP). Passive packaging paid more attention on the mechanical strength, barrier performance and thermal stabilities (Robertson, 2013). The AP usually involves the integration of oxygen/radical/ethylene scavengers, moisture absorbers, carbon dioxide emitters

* Corresponding author at: Department of Wood Technology and Wood-based Composites, Georg-August-University of Göttingen, Büsgenweg 4, D-37077 Göttingen, Germany
E-mail address: kai.zhang@uni-goettingen.de (K. Zhang).

https://doi.org/10.1016/j.jobab.2021.11.004
Received 17 July 2021; Received in revised form 26 September 2021; Accepted 30 September 2021
Available online xxx

Please cite this article as: J. Wang, M. Euring, K. Ostendorf et al., Biobased materials for food packaging, Journal of Bioresources and Bioproducts, https://doi.org/10.1016/j.jobab.2021.11.004

and antimicrobial compounds in the packaging (Vilela et al., 2018). The IOSP have been realized by using time-temperature indicators, gas indicators, microwave doneness indicator and radiofrequency identification et al (Han et al., 2018). The SOGP are becoming increasingly urgent and significant due to the constant concerns on environmental influences of food packaging wastes. The SOGP is aiming to develop materials for food packaging with minimum environmental impact. The impact usually depends on the way in which the materials are produced and processed, and the end-of-life state of the packaging material, including recycling, incineration, landfill disposal and compost. The SOGP involves three aspects: 1) the raw material, 2) the production process and 3) the waste management (Han et al., 2018). To be specific, SOGP prefers 1) materials from renewable resources or recycled materials to eliminate the CO_2 emission and reduce the usage of petrochemicals, 2) lighter and thinner packaging with economically and energetically efficient processes, 3) materials that are biodegradable, compostable, recyclable or reusable to minimize the harmful effect on the environment. Though various techniques have been applied for food packaging, the safety issues are widely concerned, especially active packaging and packaging techniques involving nanotechnology. Some regulations, such as The EU regulation 1935/2004, EU regulation 10/2011 and the Chapter 21 of the Code of Federal Regulations in U.S., set concrete regulations, specific details and requirements on food packaging materials (Dainelli et al., 2008, Welle, 2014). The migration of nanosized substances and other hazardous food additives are the main concern.

The objective of this review is to highlight the bio-based materials for food packaging. It presents an overview of the properties of biobased materials and the techniques used for food packaging and demonstrates diverse biobased materials categorized by their size, including biobased polymers, nanomaterials, natural fibers and their composites. Biobased polymers are chemically singular polymer molecules, and the sizes of polymers are within molecular level, which can vary with the category and the origin of the polymers. Most biobased polymers should possess the length in the range from tens to thousands of nanometers. The biobased nanomaterials introduced here include mainly nanocrystals and nanofibers, which contain many polymer chains, and the sizes of nanocrystals usually have diameter with sizes of several nanometers and lengths ranging from hundreds of nanometers to several micrometers. Natural fibers are larger fibers with one or more chemical components. The practical food packaging applications of these bio-based materials involve various types of packaging and the properties such as mechanical strength, barrier properties, antimicrobial activities and antioxidant functions are discussed. The primary advantages of each material and the related functioning mechanisms are also briefly summarized in this review. We finally discuss the perspectives and challenges of biobased materials for food packaging for future research.

2. Biobased materials and their composites

Biobased materials in this review include biobased polymers, biobased nanomaterials, biobased fibers and their composites (Fig. 1). Biobased polymers can generally be classified into four types: 1) polymers extracted from biomass, such as cellulose, hemicellulose, chitin, 2) synthetic polymers from biomass monomers, such as polylactic acid (PLA) and biopolyethylene (BioPE), 3) polymers produced by microorganisms, such as polyhydroxyalkanoates (PHAs), bacterial cellulose, and 4) biodegradable polymers synthesized from petrochemical mononers, such as poly(caprolactone) (PCL), poly(butylene succinate-co-adipate) (PBSA), polybutylene adipate terephthalate (PBAT), poly(glycolic acid) (PGA), polybutylene succinate (PBS) and polypropylene carbonate (PPC). Biobased nanomaterials include mainly cellulose nanocrystals, cellulose nanofibers, chitin nanocrystals and other nanomaterials from biomass. Biobased fibers are referred to natural fibers produced by animals, plants as well as geological processes. As food packaging materials usually need to satisfy several requirements, such as mechanical properties, permeability, and antibacterial properties, the real applications of biobased food packaging materials usually involve their composites. This review presents diverse categories of biobased materials and their composites for their applications in food packaging.

2.1. Biobased polymers

2.1.1. Cellulose

Cellulose is the most abundant biopolymer in nature, and it is a linear semi-rigid polymer with D-glucose as the repeating unit. With the existence of intermolecular and intramolecular hydrogen bonds, cellulose is usually not easy to be dissolved directly for use. Cellophane, made from regenerated cellulose, has been widely used for food packaging in real life. Cellophane films are transparent and mechanically stiff with excellent stability of dimensions. They are well known as candy wrappings, and also the packaging for cheese, cookies, coffee and chocolates. Cellophane incorporated with nisin has also been reported with antimicrobial properties for meat packaging (Guerra et al., 2005). Cellulose/lignin composite transparent films were reported with UV-shielding and antimicrobial properties, with potential to be used as food packaging materials (Guo et al., 2019). There are many hydroxyl groups on the cellulose chain, allowing various reactions to obtain cellulose derivatives. Cellulose derivatives were also reported to be used for food packaging. Carboxymethyl cellulose (CMC) was blended with polyvinyl alcohol to achieve packaging films with adequate mechanical properties. With additions of some extracts, such as oleic acid containing rosemary extract oil, the resultant composite film demonstrated excellent properties for active packaging, with no fungal observed after storage for 60 d at 25 °C (Nasibi et al., 2020). It was also found that lay-by-layer edible coating of CMC and chitosan was able to preserve the freshness and quality of strawberries by lowering the metabolites contents (Yan et al., 2019). Acetate cellulose with the incorporation of bacteriophages were also reported with antimicrobial activities potential for food packaging (Gouvêa et al., 2015). Other cellulose derivatives, such as cellulose cinnamate has also been reported as effective packaging plastics for fruits, such as strawberries and mini-date vine tomatoes due to their excellent mechanical properties, thermal stability as well as low permeability to water vapor, oxygen and oil (Wang et al., 2021a; Wang et al., 2021b). It should be

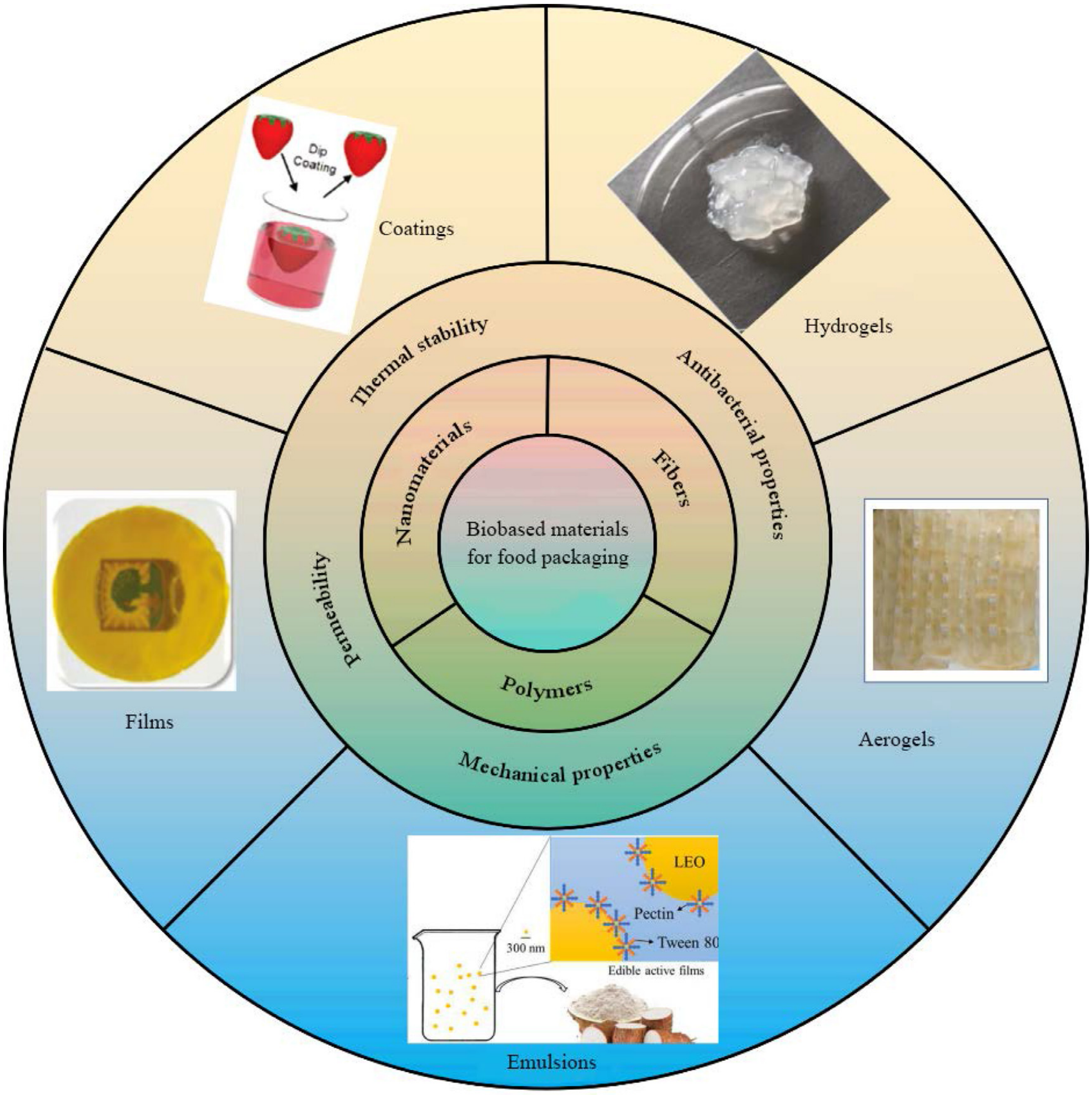

Fig. 1. An overview of biobased materials for food packaging. Reproduced with the permission. (Jung et al., 2020; Lu et al., 2020; Mendes et al., 2020; Abral et al., 2021; Zhou et al., 2021).

noted that food packaging materials made from cellulose polymer and paper are distinct in their properties and performance for their food packaging applications though the chemical composition is all cellulose.

2.1.2. Hemicellulose

Hemicelluloses are branched polymers, consisting of pentosans and hexosans. Hemicelluloses with many side chains usually are highly water-soluble and have good film-forming properties. For instance, Xylans are the most abundant type of hemicellulose in the cell wall of hardwoods. Xylan films have been reported with good barrier properties against oxygen, grease. Galactomannan and xyloglucan have been extracted from *Caesalpina pulcherrima* and *Tamarindus indica,* without using chemical reactions. Their blend films were found with good barrier properties and thermal stability, enable them as potential environmental-friendly, edible and biodegradable food packaging materials (Mendes et al., 2017). However, hemicellulose films usually suffer from sensitivity to moistures and low mechanical strength, which can be improved via either chemical modification or composition with other polymers. With chemical modifications, hemicellulose can be prepared into materials with diverse unique properties. For example, quaternized hemicellulose (QH) and chitosan were intercalated in montmorillonite (MMT), resulting in compact and mechanically robust films with improved oxygen and water vapor barrier properties for food packaging. The improvement of mechanical properties was mainly due to the compact nacre-like structure while the enhancement of barrier properties was mainly due to the high viscosity of chitosan (Chen et al., 2016). Acetylated hemicellulose (AH) mixed with nanocellulose, followed by coating with polycaprolactone films were reported with enhanced mechanical properties and hydrophobicity. The films were incorporated with polyphenols and the antioxidant release was evaluated. This hemicellulose based composite film can be used as active food packaging materials (Mugwagwa and Chimphango, 2020).

2.1.3. Chitosan/Chitin

Chitin is also an abundant biopolymer on earth after cellulose. It mainly originates from the exoskeleton of marine invertebrates and insects or the cell wall of some fungi. Chitosan is a cationic biopolymer, which can be produced by deacetylation of chitin. Chitosan has amino and hydroxyl group in its structure, which enabled the antimicrobial activities against gram-positive and gram-negative bacteria. Chitosan films showed good antimicrobial and antioxidant activities for food packaging (Kumar et al., 2020b). Chitosan has also been blended with 1) other biopolymers including polysaccharides, proteins, 2) some synthetic polymers, e.g., poly(vinyl alcohol), poly(lactic acid), 3) some functional extracts, such as beeswax, honeysuckle flower extract, and 4) nanomaterials, such as metal and metal oxide nanomaterials, graphene oxide, montmorillonite, silica, CNF to adjust the mechanical, thermal and barrier properties for food packaging (Wang et al., 2018; Kumar et al., 2020b). Furthermore, chitosan has three reactive functional groups: one amino group at C-2 position and two hydroxyl groups at C-3 and C-6 positions. These reactive groups enabled chitosan to be further functionalized for their diverse derivatives. These chitosan derivatives proved to demonstrate improved properties for food packaging (Wang et al., 2018). Water soluble chitosan derivatives, including alkyl chitosan, quaternary chitosan and carboxymethyl chitosan were used as effective additives for food packaging materials with improved barrier properties (Nechita and Roman, 2020). Various methods including solution casting, layer by layer assembly, extrusion and spraying/coating/ dipping have been applied to obtain chitosan-based films/coatings for shelf-life extension of fruits, vegetables, fish and meat (Kumar et al., 2020b). By integrating with some specific indicators, such as anthocyanin, chitosan films can be used for sensing films for meat freshness (Zhang et al., 2014c). Instead of conventional dry films, chitosan hydrogel films have also been developed for the sensing of microbials for the food quality and safety monitoring (Sadat Ebrahimi et al., 2015).

2.1.4. Lignin

Lignin is also an abundant biopolymer on the earth. It exists mainly in the cell wall of plants, and it is a complex polymer with cross-linked structure. It usually linked with hemicellulose with ester bonds. Lignin has three kinds of precursors, including coniferyl alcohol, p-coumaryl alcohol and sinapyl alcohol. Lignin contains various active groups, including hydroxyl, methoxy, carbonyl, carboxyl and benzene. Due to its specific structures and groups, lignin has been widely used in food packaging with its antimicrobial, UV-shielding and anti-oxidation properties. The hydroxypropylmethylcellulose (HPMC)-lignin and HPMC-lignin-chitosan composite films were investigated to evaluate the antimicrobial properties of lignin and found the concentration of lignin is critical for their antimicrobial activities (Alzagameem et al., 2019). Lignin was also mixed in PVA/gelatin composites for obtaining antibacterial properties. The hydroxyl groups of phenolic compounds in lignin interact with the cell membrane and cause the final cell lysis (El-Nemr et al., 2020). Lignin can be integrated into many polymer matrixes, including polyvinyl alcohol (PVA), latex, PLA, chitosan, gelatin and alginate, for achieving enhancement of mechanical, wettability of the materials. Lignin has also been applied for active food packaging as an antioxidant agent by various methods (Gaikwad et al., 2018). It was found that lignin with low molecular weight, narrow polydispersity, more phenolic hydroxyl groups, less aliphatic hydroxyl groups demonstrated higher antioxidant activity (Pan et al., 2006). Comparing lignin from diverse sources, such as alkali, lignosulfonate, kraft or steam explosion lignin, alkali lignin was found with higher radical scavenging activity because it contains free phenolic monomers (Domenek et al., 2013). Lignin with syringyl groups were found with the highest antioxidant activities and UV-shielding properties due to the additional methoxyl groups (Guo et al., 2019). By adding lignin in (3-hydroxybutyrate) (PHB) and polyhydroxyalkanoate (PHA) films, lignin acted as both antioxidant and nucleating agent. As a result, the films showed increased stiffness, lower oxygen and carbon dioxide permeability for food packaging (Vostrejs et al., 2020).

2.1.5. Starch

Starch consists of linear and helical amylose and branched amylopectin. Starch is semi-crystalline polymers and thus starch films are not adequately flexible for food packaging. In order to increase the flexibility of starch films, chemical or physical modifications, plasticization and enzymatic treatment were used. Citric acid and gelatin are proved to be effective for improving the properties and structure of starch films. Some starch derivatives such as acetylated starch have been applied as coatings on paper and showed good water vapor barrier properties and barrier properties against other gases for food packaging. To overcome the hydrophilicity of starch, the sulphur hexafluoride plasma treatment was applied and the contact angle reached 140°. Moreover, some additives such as CNF, ZnO, MgO and nanoclay can be blended with starch to achieve coating materials with antimicrobial properties, UV-shielding properties and better barrier properties (Nechita and Roman, 2020). Starch composite films incorporated with emulsions of lemongrass essential oil and pectin demonstrated good thermal stability, excellent barrier to water vapor and mechanical properties for food packaging (Mendes et al., 2020).

2.1.6. Pectin

Pectin is a complex heteropolysaccharide and is a component in cell wall of plants, enabling the primary cell wall extension and plant growth. Pectin has been widely used in food packaging materials. Pectin films originating from casting or thermo-compression moulding were successfully prepared for food packaging. Pectin also possesses good compatibility with other biopolymers, including proteins, lipids and other polysaccharides. Glycerol was usually used as the plasticizers for pectin films. Pectin in composite coatings with corn flour and beetroot powder demonstrate the properties of modifying the atmosphere of food, changing the oxygen level

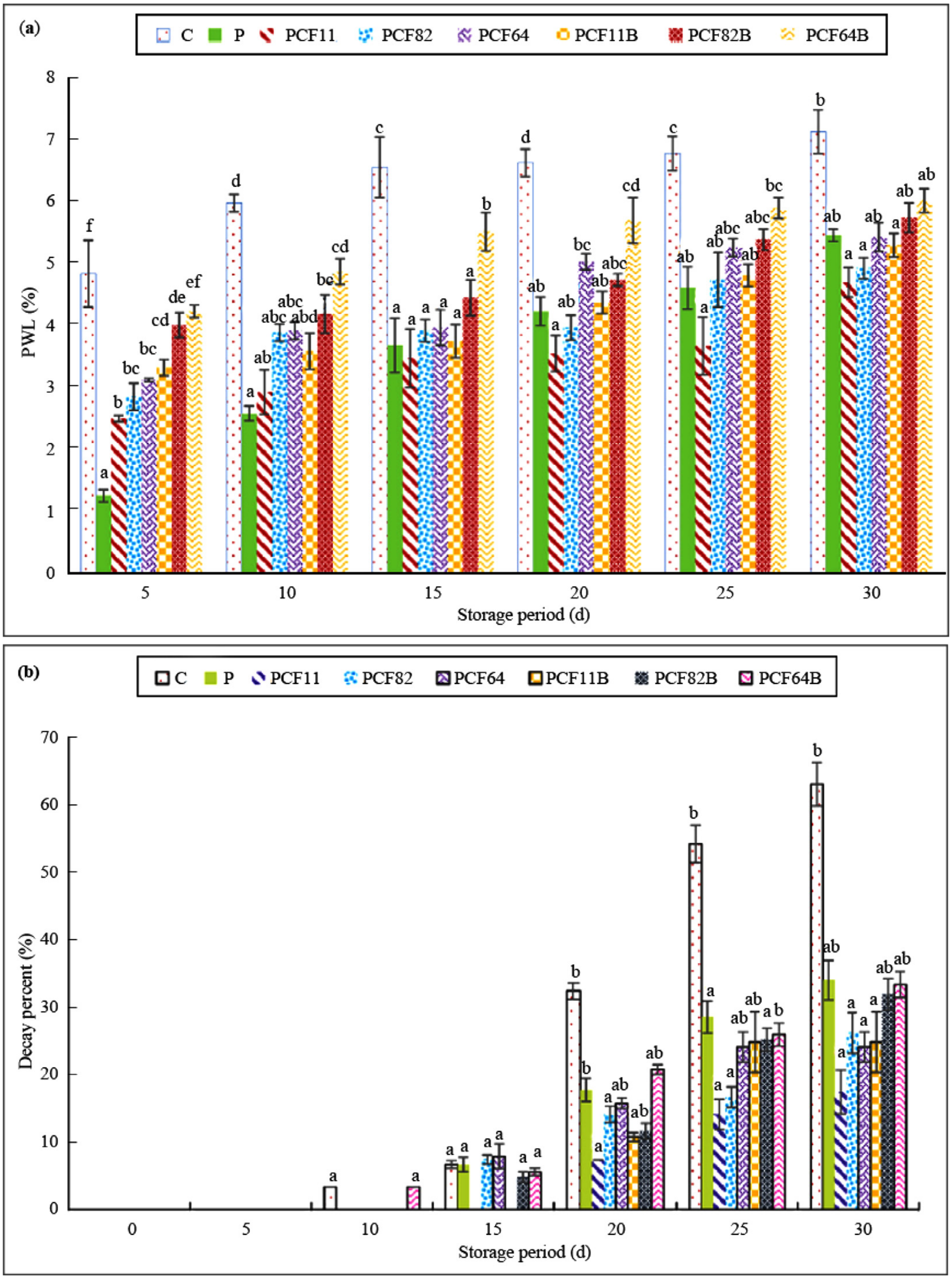

Fig. 2. (a) Physiological weight loss (PWL) and (b) decay percent of bared and coated tomatoes. The coating materials involves pectin, corn flour and beetroot powder. Reproduced with permission (Sucheta et al., 2019).

and refrain the emission of ethylene and thus delaying the food decay (Fig. 2) (Sucheta et al., 2019). Pectin was also used as coating directly on cut fruits and vegetables, or as encapsulating material for active packaging with controlled release of active compounds. For example, the active packaging using pectin film result in the delaying of soybean oil oxidation for 30 d. It was also found that the pectin/corn flour composites using as coating materials on the surface of tomatoes effectively help maintaining the freshness of tomatoes (Mellinas et al., 2020). Integrated with clove essential oil, pectin films were enhanced with barrier, mechanical, antimicrobial and antioxidant properties (Nisar et al., 2018). By encapsulating marjoram essential oil in pectin matrix films via Pickering emulsion, the release of marjoram essential oil was effectively slowed down for increasing the shelf-life of food (Almasi et al., 2020). With the application of mint and rosemary essential oil and nisin, the pectin/starch/chitosan films showed improved antimicrobial properties, barrier properties, tensile strength and thermal stabilities (Akhter et al., 2019). Tea extracts were also proved as an effective in enhancement of the mechanical, antioxidant and antimicrobial properties of pectin (Lei et al., 2019).

In particular, red cabbage extracts were used in pectin film as smart packaging for fish and meat. The film changes colors with different pH value when the proteins degrade with increasing pH, anthocyanins from red cabbage extracts change the color as the sensor (Dudnyk et al., 2018). Composite films of pectin and nanomaterials including Ag and TiO_2, Au, Cu nanoparticles were also effective as enhancement with both the mechanical, barrier, and the antimicrobial and UV-screening properties for the food packaging (Kumar et al., 2020a, Mellinas et al., 2020). Furthermore, some phenolic compounds and free fatty acids were also effective additives to pectin films for food preservation with their antimicrobial activities (Kumar et al., 2020a).

Apart from films and coatings, pectin was also prepared into hydrogels, aerogels and emulsions for food-packaging. Pectin-chitosan hydrogels encapsulated with garlic and holy basil essential oil were found with excellent antimicrobial properties and inhibition of enzymatic browning and water loss for effective food packaging (Torpol et al., 2019). On the other hand, aerogels with high porosity, high specific surface area, low density and good thermal insulating properties, have attracted attention for food packaging materials. As an example, pectin-based aerogels have been applied for the storage of food with sensitivity to temperature (Nešić et al., 2018). In addition to encapsulating various active compounds, such as essential oils and extracts in pectin via emulsions as coating for food packaging, the biodegradation rate of pectin emulsions with lemongrass essential oil in cassava starch film were also investigated (Mendes et al., 2020).

2.1.7. Alginate

Alginate is an anionic biopolymer originating mainly from brown algae. Alginate are considered safe and can be used as food-contact materials. Alginate and calcium chloride were applied on the surface of paperboard to improve the barrier properties and oil resistance. Composites of sodium alginate/sodium carboxymethylcellulose/and sodium alginate/propylene glycol alginate were proved to be excellent coating materials for paper with improved water resistance, oil barrier properties and also modification of mechanical properties (Nechita and Roman, 2020).

2.1.8. Proteins

Some proteins possess good film-forming properties and can form self-standing films for food packaging. Most reported proteins that have been applied as food packaging material include whey, soy, and wheat gluten protein-based films. In addition, other proteins such as fish gelatin, corn zein, milk protein and myofibrillar proteins have also been reported for their utilization in food packaging applications. Proteins inherently have some drawbacks for their application as food packaging materials, such as inferior mechanical properties, barrier properties, thermal properties and physicochemical properties. In order to improve these properties, proteins have been incorporated with nanofillers for food packaging. Nanofillers including inorganic nanofillers such as layered silicates, silica, carbon nanotubes, zinc oxide, titanium dioxide and silver nanoparticles, and organic nanofillers such as starch nanocrystals, nanocellulose, nanochitin have been applied to improve the mechanical and barrier properties of protein-based films for food packaging applications (Zubair and Ullah, 2020). For example, the soy protein isolate/montmorillonite (SPI/MMT) nanocomposite films were reported as food packaging materials, at optimum MMT concentrations, the mechanical properties were enhanced with the intercalated and exfoliated MMT structures (Lee and Kim, 2010). The application of plasticizers and some active compounds also contributed to the improvement of water barrier properties and delay of microorganism growth for protein-based food packaging materials (Chen et al., 2019). Moreover, proteins can be modified via chemical, physical and enzymatic methods to obtain films with enhanced mechanical strength and water resistance. The mechanical strength of protein-based films was usually low and thus proteins were also used as coatings on the surfaces of cellulose-based packaging materials for their real applications (Coltelli et al., 2015).

2.2. Biobased nanomaterials

2.2.1. Cellulose nanofibers

Cellulose nanofibers (CNF) have been fabricated into films and their water vapor transport properties have been investigated in comparison with paper (Bedane et al., 2015a). In order to improve the water vapor barrier properties of cellulose nanofibers (CNFs) films, soybean-oil based polymers were coated on the surface (Lu et al., 2014). Some methods including sol-gel method, layer by layer assembly, electrospinning, and composite extrusion have also been used to improve the water vapor barrier and water resistance of CNF films (Nechita and Roman, 2020). The CNFs were also dispersed with polymers, including polylactic acid (PLA), and polyethylene glycol (PEG) to obtain composites for food packaging. Cellulose nanofibers were chemically modified with hydrophobic characters to realize good compatibility with PLA. The biodegradable composite was surface-coated on packaging paper for obtaining lower water vapor permeability (Song et al., 2014). The CNFs were coated on paper substrates and their properties, such as the release of active compounds through the nanoporous networks of CNFs, water retention value, air resistance, and tensile strength have been investigated (Lavoine et al., 2016; Jin et al., 2021). Furthermore, CNFs were coupled with clay minerals, such as montmorillonite or kaolinite, to form composites with the aim to reduce the cost and improve the mechanical, gas barrier and fire-retardant properties (Alves et al., 2019). Moreover, by using carboxymethyl cellulose nanofibers as shell and the chitosan/silver nanoparticles as core in 3D-printing inks, antimicrobial and cushioning bifunctional aerogels were produced for food packaging (Zhou et al., 2021).

2.2.2. Cellulose nanocrystals

Cellulose nanocrystals (CNC) can be isolated from various resources via diverse methods. The CNC were widely employed as reinforcing agent for many materials, sometimes with chemical modifications with the aim to improve the compatibilities. It has been blended with polyethylene, polypropylene, polylactic acid (PLA), PVA, PET and carboxymethyl cellulose (CMC) to prepare food packaging materials (Stark, 2016; Huang et al., 2020). As an example, the resultant composite films from PVA/CNC/CMC were proved to be enhanced with mechanical, barrier and thermal properties as well as improved transparency for food packaging (El Achaby et al., 2017). Incorporated with some antimicrobial materials, the PLA/nanocellulose composite materials could also have antimicrobial properties. The antimicrobial materials include two categories, organic materials and inorganic materials. Organic antimicrobial material includes organic acids, polymers or enzymes, while inorganic antimicrobial agents include metal and metal oxide nanoparticles (Gan and Chow, 2018). By employing the cellulose nanocrystals from sugarcane baggage, hydrogels were formed with the assistance of zinc ion. The resultant hydrogels were mixed with some colour indicators, which respond to pH and the concentration of CO_2. As an example, the responsive hydrogels were applied as indicators for the freshness of chicken breast for intelligent packaging (Fig. 3) (Lu et al., 2020). Incorporated with egg-derived polymers, cellulose nanocrystals suspensions with the assistance of glycerol and curcumin, are reported as excellent coatings for the preservation of perishable fruits (Fig. 3). The coated fruits including banana, avocado, papaya and strawberry were well preserved with delayed ripening, dehydration and microbial inhibition (Jung et al., 2020).

2.2.3. Bacterial cellulose

Bacterial cellulose is produced by certain types of bacteria, instead of plants. Bacterial cellulose has characteristics of high purity, large degree of polymerization, high strength and high water-holding capacity. Bacterial cellulose functionalized with bovine lactoferrin were used as edible packaging with antimicrobial functions (Padrão et al., 2016). Antimicrobial peptides have been immobilized in bacterial cellulose to realize food packaging (Malheiros et al., 2018). Some composite films of bacterial cellulose and polymers were also developed for food packaging (Bandyopadhyay et al., 2018). The PVA/gelatin reinforced with bacterial cellulose nanowhiskers were also investigated for food packaging materials (Haghighi et al., 2021). Corn starch/bacterial cellulose nanowhiskers composite films coated with electrospun polyhydroxyalkanoates/cellulose nanowhiskers were investigated for food packaging applications with improved mechanical properties and barrier properties (Fabra et al., 2016). Bacterial cellulose nanofibers blended with starch and chitosan gave rise to eco-friendly edible food packaging materials with good mechanical and antimicrobial properties (Abral et al., 2021). Incorporation of bacterial cellulose nanocrystals and silver nanoparticles in chitosan matrix result in active packaging materials with antimicrobial properties. In addition, the sensitivity of water vapor and mechanical properties were also improved (Salari et al., 2018). Incorporating with ZnO nanoparticles, bacterial cellulose films were found to be improved with mechanical, barrier and antimicrobial properties. Moreover, the release of ZnO can be controlled, promising for active packaging (Shahmohammadi Jebel and Almasi, 2016). Blending some essential oil in bacterial cellulose films decrease the tensile strength and increase the elongation at break for the edible films as essential oils act as plasticizers (Indrarti, 2017). Fruit purees and nanofibrillated bacterial cellulose were blended with pectin to prepare food packaging materials. It was found that the incorporation of fruit purees was disadvantageous for food packaging, with decrease of barrier properties, mechanical strength and modulus while the incorporation of nanofibrillated bacterial cellulose is advantageous for food packaging applications with the improvement both in mechanical properties and barrier properties (Viana et al., 2018). With the combination of bacterial cellulose, tapioca starch and chitosan, the resultant edible composite film demonstrate good mechanical properties, high thermal stability and also antibacterial properties for food packaging applications (Abral et al., 2021). Apart from CNC, CNF and bacterial cellulose, other cellulose, including microcrystalline cellulose, microfibrillated cellulose and cellulose acetate have also been used as additives in PLA for food packaging materials with improved mechanical, barrier, transparency (Khosravi et al., 2020).

2.3. Biobased fibers

2.3.1. Paper

Paper is produced from cellulose fibers. It has advantages including recyclability, biodegradability and compostability as food packaging materials, in comparison with some synthetic polymers. There is packaging made of paper and card-/paperboard and well-known and long-established examples for paper-based food packaging are tea bags, egg trays or juice boxes. Those contain either softwood, hardwood, and/or agro-residual fibers obtained from chemical and mechanical pulping process. Consequently, their composition and fiber morphology are crucial for the desired end use and demands given by the customers. For example, tea bags showed a high level of fiber coarseness (1.2 mg/m) to provide a smoother texture (Sood and Sharma, 2021). It was shown, that agro-residual fibers cannot replace wood-based fibers in total, as they were shorter and contain certain amount of non-fibrous components. However, these can be used in part as substitutes, up to a ratio of 85% for softwood fibers (Bhardwaj et al., 2019). Another lignocellulosic source for papermaking was successfully investigated for pine needles, which was used together with various amounts of nano zeolite powder (up to 30%) as an ethylene scavenger (Kumar et al., 2021). When using recycled fibers, e.g., from newspaper, the migration of mineral oil due to inks or other contaminants poses a challenge, suggesting the use of mineral oil-free inks, or bio-based inks respectively (Buist et al. 2020).

The hydrophilic nature and barrier properties of paper are inferior. For example, a high relative humidity (RH) can negatively alter the product properties such as compressive strength and dimensional stability due to hygroexpansion (Niini et al., 2021). To overcome these challenges, coatings such as synthetic polymers and lamination with aluminum foil were used, which belong to the group of Food Contact Materials (FCM) (Simoneau, 2008). The addition of coatings or inks as FCM can be considered as

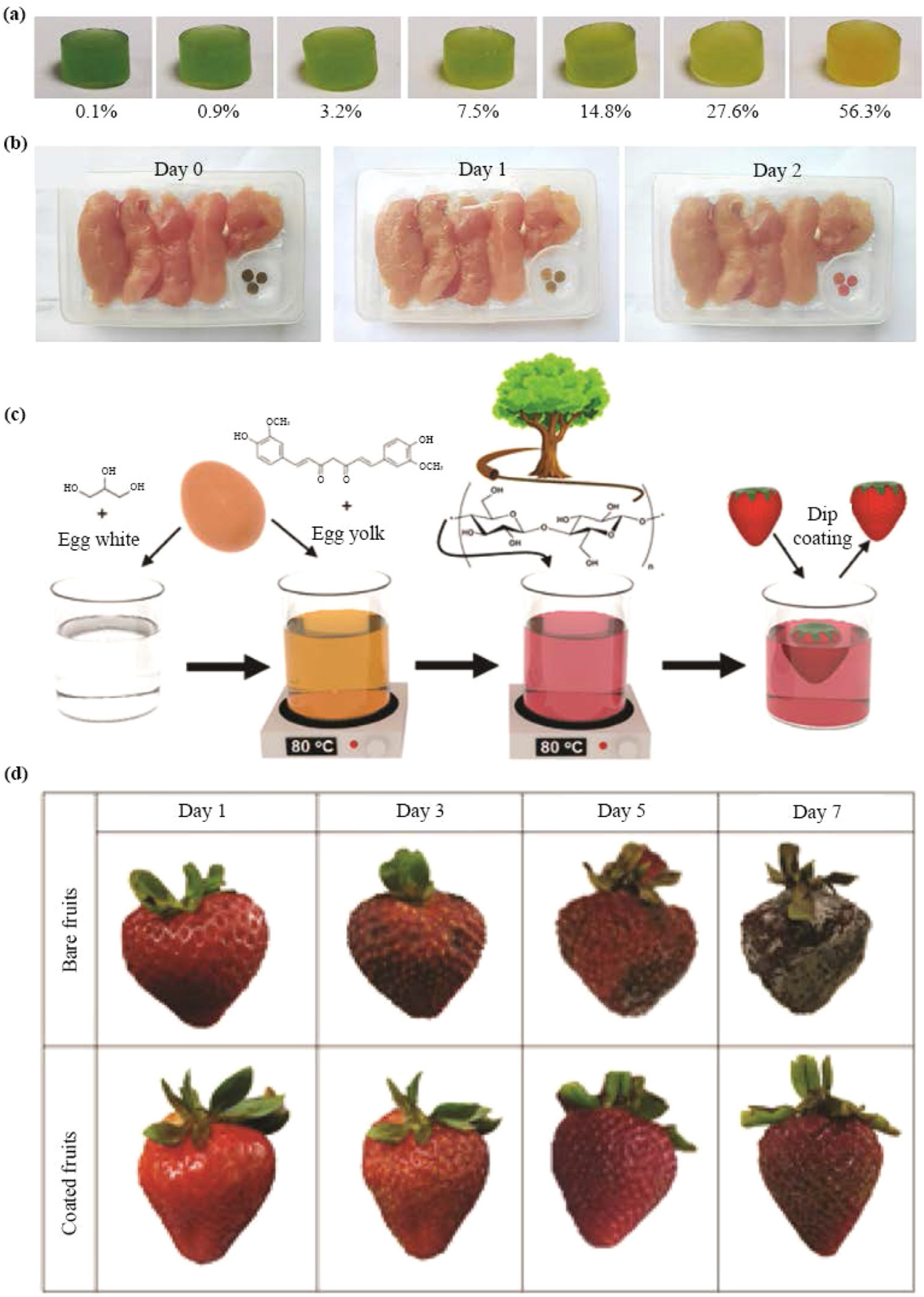

Fig. 3. Application of cellulose nanocrystals for food preservation. (a, b) Nanocellulose-based hydrogel used as indicators for intelligent food packaging. (a) Color response of nanocellulose-based hydrogel to the concentration of CO_2 concentration; (b) Nanocellulose-based hydrogel used as indicator for the freshness of chicken breast. Reproduced with permission (Lu et al., 2020). (c, d) Egg-derived polymers reinforced by nanocellulose as coatings for the preservation of perishable fruits. (c) Schematic illustration of the synthesis of the nanocellulose-based nanocomposite suspension and the coating process of fruit; (d) Time-lapse photos of bared and coated strawberries (Jung et al., 2020).

problematic for paper based food packaging due to the occurrence of hazardous chemicals, which can migrate and contaminate the food within (Selin et al., 2021). Among other things, impacts on genotoxicity or antagonistic effects on the oestrogen as well as androgen receptors were detected especially from inks (Selin et al., 2021). Additionally, these methods still have concerns regarding sustainability, recyclability and biodegradability (Nechita and Roman, 2020). Thus, the paper surfaces were coated with bio-based polymers, including PLA, poly(3-hydroxybutyrate-co-4-hydroxybutyrate (PHBV), or chemically grafted with some proteins (Bedane et al., 2015b). Antimicrobial paper has also been reported by dip-coating or mixing with guanidine-based antimicrobial polymers (Wei et al., 2017). By grafting the guanidine-based polymer on the surface of beeswaxes particles, the modified beeswaxes were shown with dual functions of improving both the antimicrobial and water barrier properties of paper for packaging (Zhang and Xiao, 2013). Beeswaxes-chitosan emulsions and beeswax mixtures emulsions were also coated on the surface of paper to improve the water vapor barrier properties and water resistance of packaging paper (Zhang et al., 2014a; Zhang et al., 2014b). Cellulase-assisted refining of paper gave rise to improved barrier and grease-resistance of paper (Lu et al., 2016). Polysaccharides have also emerged as effective coatings for paper packaging due to their advantages including wide availability, good film-forming properties. The resultant coated paper showed improvements in barrier and mechanical properties (Nechita and Roman, 2020). Cellulose itself has difficulties to be used directly as coatings for paper packaging because its high crystallinity, high hydrophilicity, difficulty of being dissolved, and bad film-forming properties. However, cellulose derivatives including cellulose ethers, such as carboxymethyl cellulose (CMC), methyl cellulose (MC), hydroxypropyl cellulose (HPC) and hydroxypropyl methyl cellulose (HPMC) and cellulose esters, e.g., cellulose acetate can be used as foods packaging materials with good mechanical, barrier and film-forming properties. The HPC and cellulose acetate are also beneficial due to their nontoxicity, edibility and thus direct contact with food (Nechita and Roman, 2020). Another possibility is to use nanofibrillated cellulose (NFC) obtained from microcrystalline cellulose for coating papers with high-performance (Jin et al., 2021). Moreover, the PHBV and the modified clay nanocomposites were proved to be very effective to improve water barrier properties to tens or hundreds of times for food packaging paper (Farmahini-Farahani et al., 2015). When using recycled fibers, e.g., from newspaper, the contamination due to inks once again comes to mind, suggesting the use of mineral oil-free inks (Buist et al. 2020).

2.3.2. Other natural fibers and their composites

Natural fibers can be produced by 1) animals, such as silk and wool, 2) plants, such as vegetable fibers, including flax, hemp, cotton lint, or sisal fibers and 3) geological processes, such as asbestos. Natural fibers, especially of lignocellulosic origin, are used as reinforcing fillers in biocomposites, embedded in a matrix of a full-bioplastic such as polysaccharides and proteins (Berthet et al., 2016). They can be produced as conventional plastics, i. e., extrusion, injection/compression molding or resin transfer molding (Faruk et al., 2012; Berthet et al., 2016). Besides the main benefits of renewability and degradability, natural fiber for food packaging provides a low density per unit, sufficient strength properties, low tool wear and above all else, they are comparatively low-cost ones (Majeed et al., 2013; Berthet et al., 2016). However, a serious problem, as with paper-based packaging, is the hydrophilic nature of natural fiber composites (Majeed et al., 2013, Berthet et al., 2016).

Several studies were performed investigating fibers of different origin and/or processing techniques for the production of food packaging. For example, coconut fibers up to a ratio of 10% together with a type of polyhydroxyalkanoate (poly(3-hydroxybutyrate-co-3-hydroxyvalerate) were used as biodegradable aliphatic polyester (bioplastic) and oregano essential oil (OEO) for impregnating coconut fibers for antibacterial activity (Torres-Giner et al., 2018). It was shown that the optimal fibers size was > 1500 μm with corresponds to an aspect ratio of 7. There was no crucial drawback concerning processing or material properties when using the highest amount of coconut fibers of 10%, which would save amounts of costly bioplastics. The addition of oregano essential oil resulted in successful bacteriostatic effect using 3% of OEO treated coconut fibers (Torres-Giner et al., 2018).

The same bioplastic was used together with 20% of wheat straw fibers (WSF) as reinforcing agent (Martino et al., 2015). Additionally, different percentages of biodegradable substances, i.e., acetyltributyl citrate (ATBC) or glycerol triacetate (GTA) were added to enhance external plasticization. It was shown, that a content of 10% of these bio-plasticizers led to an increase of the elongation at break. This effect was neutralized when adding WSF to the formulation due to interfacial gaps and therefore decreased adhesion strength between fiber and matrix, evoked by the more hydrophilic nature of the WSF against the hydrophobic nature of the matrix substance (Martino et al., 2015). A further study tested, besides wheat straw fibers, fibers from brewing spent grains and olive pomace, and concluded that WSF are most promising regarding material requirements for food packaging. Though, by adding more lignin containing olive pomace fibers to the matrix, the water vapor permeability was less pronounced than for more cellulose-rich fibers originated from wheat straw or fiber spent grains (Berthet et al., 2015).

The use of fibers from almond shell, rice husk, and seagrass as lignocellulosic wastes were investigated to reinforce biodegradable plastics for food packaging (Sánchez-Safont et al., 2018). Here, it was shown that thermal stability and water barrier performance was lowered by adding these kinds of fibers to the PHB matrix. A high water permeability consequently leads to a reduced material strength, shortened life span and increased susceptibility towards microbial growth (Chong et al., 2021). However, fibers originated from almond shells resulted in most balanced material properties in comparison to the other types of fibers (Sánchez-Safont et al., 2018). The use of waste fibers from betel nut was investigated as well (Das et al., 2020). Their work showed superior material properties up to fiber content of 10% and even a lower water vapor permeability than comparable food packaging made of cardboard. Other sources of lignocellulosic fibers, such as fibers from rice straw or husks, sugarcane bagasse, barley straw or husks, or maize (corn cob, corn husk), for biocomposites were listed in a review article (Bhardwaj et al., 2020).

One possibility to avoid enhanced water permeability is to add nanoclay to the lignocellulosic fiber reinforced bioplastics. The incorporation of nanoclay leads to intercalated and exfoliated structures, providing higher barrier properties (Majeed et al., 2013). To further enhance stability towards microbial growth and, thus, prolonged shelf life and food safety as well as quality, it is possible

Fig. 4. Selected demonstration of plant fiber-based containers for the packaging of (a) vegetable and fruit (b) vegetables (c) meat and (d) coffee. Reproduced with permission (Bonito Packaging Co. Ltd., 2021).

to add plant extracts, especially containing phenolic and flavonoid compounds (Valdés et al., 2014; Munteanu and Vasile, 2019) or the oregano essential oil presented above (Torres-Giner et al., 2018).

A Chinese manufacturer (Bonito Packaging Co., Ltd., Nantang Road, Chashan Town, Dongguan City, Guangdong) already offers bicomposites made of bagasse or bamboo fibers, without further specification of the matrix substance (Fig. 4) (Bonito Packaging Co. Ltd., 2021, https://www.bonitopak.com/molded-fiber-for-food-packaging). A bioplastic film containing only of cellulose extracted from cacao pod husks, reinforced with sugarcane bagasse fibers at different ratios was introduced in a recent study (Azmin et al., 2020). Thereby, a ratio of 75% cellulose and 25% fibers were found to be best according to lowest water absorption and water vapor permeability, which is crucial as discussed above (Azmin et al., 2020).

3. Conclusion and perspectives

This review highlights the lasted findings of bio-based materials for food packaging, emphasizing their properties and performances. Biobased materials including biobased polymers, biobased nanomaterials, biobased fibers and their composites, are proved to be effective materials for food preservation. The biobased materials were applied for food packaging as coatings, films, hydrogels, aerogels and emulsions. Diverse properties including mechanical properties, gas barrier performance and antimicrobial functions of these bio-based materials were discussed for their food packaging applications. Characteristics of these biobased materials including the performance of active packaging and smart packaging were also concluded.

The future research of biobased materials for food packaging will need to further improve the performance of these materials as the globalization leading to higher requests for food preservation and transportation. Multiple factors need to be considered, when referring to the real application of these biobased materials. Suitable biobased materials should be chosen and the properties need to be further designed with physical or chemical pathways according to the storage condition requirements of the particular foods for their packaging.

Though biobased materials for food packaging have achieved great progress, there are still some obstacles that impede these biobased materials from commercialization. One of the vital factors is how to decrease the economic and energy for the production and processing of these biobased materials, which include the extraction of biopolymers, the isolation of biobased nanomaterials and natural fibers as well as the preparation of the bio-based materials for food packaging. More adaptable commercial techniques and equipment that could match the production of biobased materials in large scale are highly demanded.

Another future trend will be focused on how to obtain more understandings on the biodegradation and sustainability of these materials for minimizing their environmental impact after using. More efforts are demanded to develop greener processes that should involve less or none toxic organic solvents. Real eco-friendly, sustainable and biodegradable biobased materials for food packaging are required for the protection of the environment.

The food safety issue is also of great importance regarding to the development of some biobased materials for food packaging. One challenge is those food packaging materials involving toxic solvents or processes. Particular attention needs to be paid to the active and intelligent packaging as well as the food packaging involving nanotechnology. The migration of substances into the food needs to be quantitatively evaluated and should fulfill the requirements of the particular regulations for food safety. More advanced technologies to determine the traces of substances that could migrated to the food are needed.

Declaration of Competing Interest

There are no conflicts to declare.

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