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The novel trend of bacterial cellulose as biodegradable and oxygen scavenging films for food packaging application : An integrative review

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Abstract. Excessive use of petroleum-based plastic packaging impacts environmental damage, so the development of biodegradable food packaging can be the solution. Bacterial Cellulose (BC) is an exopolysaccharide synthesized by several bacteria from the Acetobacteraceae family, which has the advantage of being a material in the blinding of biodegradable packaging films because of the high level of purity compared to cellulose from plants. This review aims to provide an overview of the potential for the development of BC as a primary material for producing biodegradable packaging films and expanding its application through the incorporation of oxygen scavenging agents to increase the dual function of food packaging. This study is expected to be able to encourage the increase in the use of sustainable packaging as a response to the issue of environmental damage, provide alternative technologies for increasing the shelf life of food through active scavenging systems, and expand the application of BC as raw material for food packaging.

1. Introduction

The needs and desires of consumers for food packaging have recently changed preferences, and society demands the addition of functional values in food packaging [1]. In general, food packaging must fulfil four main functions, namely storage, protection, convenience, communication [1–4] The people's preference for increasing the functional values of food packaging is related to environmental issues and packaging technology. In terms of environmental issues, conventional food packaging, especially plastic, is widely used today [5] However, the material for making plastic generally comes from non-renewable natural resources, namely petroleum and is difficult to harvest by microorganisms in nature because of its long carbon chains that contribute to environmental pollution [6].

The majority of food plastic packaging is generally single-use designed for immediate disposal after use [5,7]. Its increasing use has a significant impact on global plastic waste production [8,9]. In 2016, humans globally produced 242 million tons of plastic waste or 12% of all existing waste types, and it is predicted that in 2050 the world's waste will increase by 70% to 3.4 billion tons [10], with an increase in plastic waste around 12,000 million tons if no preventive action is taken [11]. In response to the issue of environmental pollution by non-biodegradable packaging polymers and consumer demands for the use of sustainable food packaging, research on food packaging in the form of bioplastics or films that are biodegradable from renewable natural resources continues to grow rapidly [7,12–14].

One of the renewable materials that can be developed as a bioplastic is Bacterial Cellulose (BC), a microbial polymer in the form of exopolysaccharides derived from microorganisms [7,12,13,15].

Acetobacter xylinum is the most studied bacteria because it has the best potential in exopolysaccharide synthesis. This bacterium also has an excellent ability to assimilate various sources of different types of Carbon and Nitrogen [16–18]. The structure of BC can be easily modified as needed during the synthesis process carried out in the fermentation process by controlling the addition of substrate, type of fermentation media, and other sources of nutrition [18,19]. Bacterial cellulose (BC) has advantages in producing biodegradable films because it has a unique nanofibrillar structure with good mechanical properties [7,14,17,20].

In terms of packaging technology, there has been an expansion of the actualized packaging function by developing smart and active packaging. Both types of packaging provide convenience to consumers; smart packaging is designed to provide information on changes in food quality in packaging or changes in the internal and external environment of packaging [2,16,21–23]. This information is identified through indicators (time-temperature indicators and freshness indicators), sensors (Biosensors and Gas Sensors), and traceability technology (Barcodes and Radio-Frequency Identification Systems) [2,16,21,24–30]. Meanwhile, active packaging is designed by deliberately adding bioactive compounds to the packaging system classified into 2, namely active scavenging systems or absorbers to absorb the presence of unwanted compounds in food products or the environment in the packaging. The second active packaging is active-releasing systems or emitters to release bioactive compounds into food products and the internal packaging environment (Figure 1) [1,24,31,32]. The purpose of active packaging is to extend shelf life or increase sensory properties by maintaining or increasing the safety and quality of food products [24,31,33].

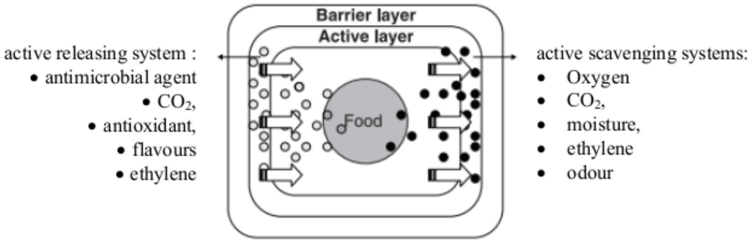

Figure 1. Active packaging systems [32]

Several types of food products have a negative response to oxygen, which affects the quality and shelf life of a decline. The oxidation of foodstuffs promotes the growth of aerobic microorganisms, decreased sensory properties, discolouration, and loss of some nutrients [2,31,32]. Oxygen scavenging can improve the oxygen barrier function of packaging [34,35]. Oxygen scavenging can take the form of sachets, labels, or the use of oxygen scavenging agents, which are directly incorporated into packaged polymers [24,31,32,34]. According to Roberta [34] the use of oxygen scavenging in the form of sachets allows the leakage of sachets which will contaminate the product or sachet eaten by consumers, so that the incorporation of oxygen scavenging agents into plastic polymers or packaging films has better consumer acceptance than the use of sachets [34,35]. Several studies have discussed and carried out active packaging using biodegradable films based on BC [7]. Based on the background, the aim of this review is to provide an overview of the potential for the development of BC as a primary material for the production of biodegradable packaging films, as well as to expand its application as a packaging material through the incorporation of oxygen scavenging agents to increase the dual function of food packaging.

2. Biodegradable packaging

A polymer must be made of renewable or biocompatible materials and can be biodegradable in order to be categorized as a bioplastic or biopolymer [36]. Biodegradable packaging polymers are generally grouped based on the raw material extraction process or method of manufacture, as well as the type of raw material used (renewable or non-renewable) [12]. The manufacture of biodegradable packaging polymers developed from non-renewable material sources can be derived from synthetic polymers derived from fossil resources (petroleum) such as Polyvinyl alcohol (PVA), Poly Glycolic Acid (PGA), Polycaprolactone (PCL), Polybutylene Succinate (PBS), Polytrimethylene Terephthalate (PET), Polybutylenen Adipatecoterephthalate (PBAT) [7,12,36].

Biodegradable packaging polymers from renewable resources can be derived from the extraction of polymers sourced from natural materials (biomass) such as polysaccharides and proteins [7,12,36]. Sources of polysaccharides can be derived from cellulose, starch, chitin, chitosan, gum, alginate, pectin, pullulan and carrageenan [37]. Protein ingredients can be derived from plant origin protein in the form of peanut protein, gluten, wheat, corn-zein, cottonseed protein, and animal origin protein in casein and whey protein [12]. Biodegradable polymers from other renewable sources come from polymers derived from renewable resources such as the extraction of microbial polymers from microorganisms and polymers from natural monomers resulting from chemical synthesis [12]. Microbial polymers can be in the form of PolyHydroxy Alkanoates (PHAs), and Bacterial Cellulose (BC), the polymer which is chemically synthesized is Poly Lactic Acid (PLA) [12].

In the application of biodegradable packaging, it has various forms according to its designation, including bags, gels, boxes equipped with covers, and films [12]. The most common use of biodegradable packaging is in the form of films, including bioplastics, edible films, and edible coatings [38]. Priyadarshi and Rhim [39] reported the potential of biodegradable packaging in the form of flexible packaging films and edible coating based on chitosan through various manufacturing techniques, including pure chitosan films, chitosan films with a mixture of polysaccharides or proteins, and chitosan films with a mixture of synthetic polymers. Polyvinyl alcohol (PVA) is a synthetic polymer widely mixed in biodegradable packaging polymers because it shows high mechanical properties and barrier performance against oxygen and water, and is non-toxic and water-soluble [39]. Sajjan *et al.* [40] reported in their research that the interaction between gelatin (Ge), 5 mass% polyethylene glycol-400 (PEG-400) and polyvinyl alcohol (PVA) in the manufacture of biodegradable films, results showed that the film has good tensile strength equivalent to conventional plastics (79.66 MPa) and high moisture retention ability (95.64 to 96.2%), so it has the potential to be used as a bioplastics for food.

The use of plasticizers is commonly used in the production of biodegradable packaging to increase film flexibility, strength chain mobility [39–41], reduce deformation stress [37], and not break easily [42]. Plasticizers will weaken the intermolecular forces on adjacent polymer chains, thereby reducing cohesion in the film network [37,43]. Glycerol and Polyethylene Glycol-400 (PEG-400) plasticizers can reduce water vapour and oxygen permeability [12,39,40,44], Sorbitol plasticizers can increase film flexibility, but the addition of excessive sorbitol will reduce the elasticity properties of the film strength and water vapour barrier [12], Triethyl citrate is very promising as a plasticizer in biodegradable food packaging [12], Tydrophilic plasticizers such as polyols (glycerol, sorbitol, and polyethylene glycol) will increase the flexibility and extensibility of films [38,45], D-Galactitol and D-Glucitol plasticizers produced a more flexible kefiran-based film by reducing the micro-hardness and Modulus Young of the film by about 30% and 74%, respectively [46].

The fabrication and production of biodegradable films have been developed through several methods, including coating (spread coating, Spray coating, dip-coating), extrusion, direct casting and layer-by-layer assembly [39]. Orozco *et al.* [47] reported a method of fabricating biodegradable films based on poly (lactic acid) (PLA) nanoparticles through layer-by-layer assembly based on hydrogen bonding and electrostatic interactions to produce ultra-thin films, layer-by-layer assembly film assemblies on PLA / poly (vinylpyrrolidone) (PLA / PVPON) via hydrogen bonding could only be made under very acidic conditions, i.e. at pH <3 [47]. Oldoni *et al.* [48] have worked on a biodegradable film from palmer mango pulp with physiological abnormalities of Internal Breakdown (IB), pectin as the base material for the film made through the continuous solution casting method to produce a consumable film with the mango colour attribute. Films obtained from mango pulp with an IB level of more than 2/3 of the pulp showed low water permeability, largest opacity and elongation, and showed a short breakdown time of only ten days [48].

Currently, biodegradable packaging cannot completely replace conventional petroleum-based plastic packaging; some of the physical and chemical characteristics of biodegradable packaging still need to be developed. The tensile strength and elongation at break of the chitosan film showed the same good value as the cellophane, HDPE, and LDPE-based packaging. However, chitosan packaging film is sensitive to moisture and humidity, is difficult to stretch, and is not thermoplastic [39,49]. The films made from peanut starch and polylactic acid destined for cherry tomato packaging have poor mechanical properties compared to polylactic petroleum films [50,51]. Polysaccharide-based films have good properties against gas removal, such as O_2 and CO_2; however, their water vapour barrier properties are poor because they are highly hydrophilic [37,52].

3. Bacterial cellulose as biodegradable films

The most widely used biopolymers in the production of biodegradable films are those of the polysaccharide and protein groups [53]. Linear polysaccharides composed of β-D-glucopyranose monomers connected through β-1,4-glycosidic bonds known as Bacterial Cellulose (BC) are the most potential natural polymers the manufacture of biodegradable food packaging films [7,53]. Bacterial Cellulose (BC) is an exopolysaccharide synthesized purely by microorganisms outside its cells [53,54]. Bacterial Cellulose (BC) can be produced from several types of aerobic bacteria such as *Aerobacter, Agrobacterium, Rhizobium, Azotobacter, Gluconacetobacter (*this genus has been reclaimed into a new genus, namely *Komagataeibacter), Pseudomonas, Alcaligenes, Sarcina,* and *Rhodobacter* [7,14,53].

Bacterial Cellulose (BC) has the advantage of being a biodegradable film material because of its high level of purity compared to cellulose from plants, free from hemicellulose compounds, lignin and other non-cellulose compounds, high tensile strength, high mechanical properties and porosity, such as hydrogels, high strength good liquid absorption, polymerization power and a high degree of crystallinity of 84-90%, while the degree of crystallinity of plant cellulose is only around 40-60% [55], good biocompatibility, biodegradable, and is renewable [7,14,16,17,19,20,53]. The high purity means that BC does not require expensive extraction and refining processes and the use of hazardous chemicals [53,55].

The bacterium that is widely used in the production of BC is Acetobacter xylinum [16,18]. This strain is cheap and easy to handle; Acetobacter xylinum can grow in various types of fermentation media such as coconut water with optimal cellulose production [53,54]. In its growth, the fermentation medium of Acetobacter xylinum needs to be enriched with sources of nutrients C, H, N and minerals, which are carried out in a controlled process in the medium. As a source of C, sucrose, glucose, fructose, invert sugar, ethanol, glycerol, and flour can be added [16,54]. Dirpan et al. reported that coconut water fermentation media fed with nitrogen sources in the form of Ammonium Sulphate had better biomass than yeast extract (Figure 2). Then the BC membrane was used as a carrier for smart packaging indicator solution to identify the decline in beef quality, the results showed the bromothymol blue indicator changed from orange to green and the phenol red indicator changed from orange to red during 16 hours of storage from the fresh meat [16].

Figure 2. synthesis of bacterial cellulose with the addition of Ammonium Sulfate (left) and yeast extract (right) [16].

BC is reported as a packaging material that can be consumed. However, pure BC in the packaging manufacturing process has a weakness in its mechanical properties, so that other biodegradable polymers accompany its use to improve mechanical properties [20,56,57]. Bacterial cellulose that is formed on the surface of the media during fermentation must be purified before being made into a film to remove bacterial cells and substrates that are still attached to the cellulose layer using 70% alcohol, heated in distilled water at 100°C, and reheated in 1N 5% NaOH solution at 100°C, purified cellulose appears transparent [7,16,29,58]. In general, Bacterial Cellulose (BC) in the manufacture of film layers has undergone a process of reforming into microfibrils (BCMFs), nanofibrils (BCNFs), and nanocrystal (BCNCs) components in powder or suspension form. This BC form will be used as a reinforcing material for packaging film polymer composites incorporated into the polymer matrix [7,59].

Bandyopadhyay et al. [20] reported the production of packaging in the form of hydrogel film based on polyvinyl pyrrolidone - carboxymethyl cellulose (PVP-CMC) with a mixture of bacterial cellulose and guar gum (BC-GG) to improve its mechanical properties. This packaging film intended for blueberries has good hydrophobic and barrier properties. The incorporation of GG causes an increase in elasticity and bearing capacity of the PVP-CMC-BC film, and the film is 80% biodegradable within 28 days. The modified BC has a high surface area which increases the physical interaction and hydrogen bonding, the structural strength of the polymer, the barrier properties, mechanical properties, and is more resistant to thermal processes [7,59,60]. Ju *et al.* [61] have investigated films made from bacterial cellulose/poly (vinyl alcohol) with the incorporation of Bulk Chitosan (CS) and chitosan nanoparticles (CSNPs), the addition of CS to the bacterial/poly cellulose film (vinyl alcohol) improves the tensile strength and transparency of the film. CS and CSNP caused a decrease in water vapour permeability. In addition, the CSNP membrane showed antibacterial properties in *Escherichia coli* and S*taphylococcus aureus*.

Salari *et al.* [60] Have investigated bacterial cellulose, which is hydrolyzed using acid to produce bacterial cellulose nanocrystals (BCNC), along with silver nanoparticles (AgNPs) to be added in the manufacture of chitosan (Ch) based nanocomposite films. The incorporation of these polymers led to an increase in the nanocomposite film's physical, mechanical and antimicrobial properties. Another BC modification is Cellulose Acetate (CA), a cellulose derivative used in the manufacture of plastics, films, filters, membranes [62]. Cellulose Acetate is widely produced from homogeneous or heterogeneous acetylation processes of cellulose sources. In the research of Barud *et al.* [62] Cellulose acetate (CA) is produced by homogeneous acetylation of BC using acetic acid, acetic anhydride and sulfuric acid as catalysts. Khami *et al.* [63] reported BC from fermentation media of seafood canning wastewater that had been treated with semi-acetylation could produce nanofiber cellulose acetate (CANF). CANF shows good mechanical properties, including high tensile strength (90.71 MPa) and modulus young (439.36 MPa). Synthesized CANF has similar properties to petroleum plastic, except for the methyl group (CH_3), which is responsible for the strength of the plastic. Thus, CANF is not as strong as petrochemical plastics but can be used to produce bioplastics due to the presence of the -CH and -CH_2 functional groups.

4. Oxygen scavenging packaging system

The high oxygen content in food packaging will adversely affect the quality of most foodstuffs, resulting in a significant reduction in the shelf life of these foods. Oxygen will cause many deteriorative reactions such as oxidation of food components, reducing the nutritional value, discolouration, off-flavour, and facilitating aerobic microbial growth [24,31,32,34]. The presence of oxygen will become a substrate for enzymatic browning, which is undesirable for fresh-cut fruit and vegetables, and has a major impact on increasing the respiration rate and ethylene production of fruits and vegetables, especially climacteric fruits [34,64,65]. Therefore, it is very important to control the oxygen level in the package. The application of oxygen absorbers is usually carried out on wine, snack foods, nuts, bread, dried foods, tea, cakes, and milk powder [32].

Food packaging with vacuum technology and Modified Atmosphere Packaging (MAP) is widely practised. However, these technologies cannot completely remove oxygen from the headspace, there is still a possibility that oxygen is trapped in food and oxygen is dispersed during storage, or there may be subtle leaks in the packaging [32,34]. Therefore, oxygen scavenging systems are critical to use in conjunction with MAP or vacuum packaging, or oxygen scavenging is used independently [31,34,35].

The oxygen scavenging agent will interact with food to absorb excess oxygen in the packaging headspace or from the product through the transfer of active agent activity by releasing the vapour phase out into the packaging headspace or through direct contact between the product and the oxygen scavenging agent on the packaging film [34,66].

There are many methods of applying oxygen scavenging systems to packaging films, coating the surface of packaging materials with oxygen scavengers. This mechanism is expected to move the active agent from packaging to food to prevent oxidative reactions (Figure 3A) [34,67,68]. The method of incorporating into packaging is done by embedded or combining the active agent into the polymer packaging either by casting or extrusion. From this method, there will be inaction between the active agent migrating to the surface with food (Figure 3B) [24,34,66]. The active agent will be dispersed into the polymer film matrix of the packaging [24,34]. The disadvantage of incorporating active agents, especially the type of iron into the polymer film, is the risk of changing the taste (off-flavour). In addition, the extrusion method through thermal processes makes heat-sensitive active agents unusable [34]. Byun *et al.* [69] reported the manufacture of oxygen scavenging packs using ferric chloride (II) agent incorporated into warm-water fish gelatin film. The results showed the oxygen scavenging fish gelatin (OSFG) had increased water vapour and oxygen permeability, rough surface film, decreased tensile strength, and good oxygen cleaning capacity of 1969.08 cc O_2/m^2/mil.

The multilayer method is applied by placing the active agent between the film layers that flank it (Figure 3C).

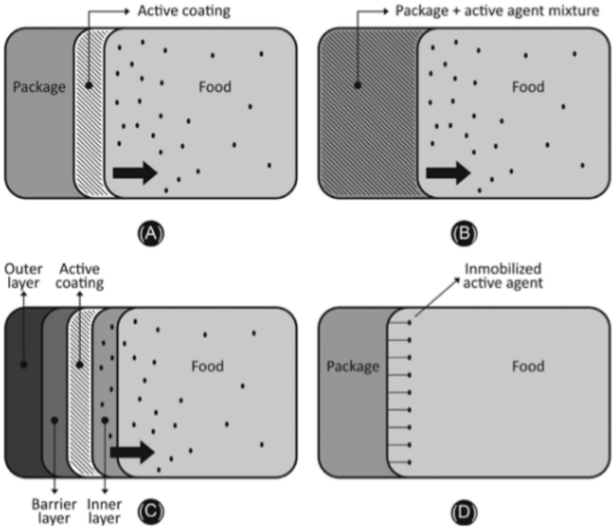

Figure 3. Various methods of application of oxygen scavenger to packaging films and ways of migrating oxygen scavenger agents : the packaging surface is coated with an active agent (A); incorporation of the active agent into the polymeric packing matrix (B); multilayer active film (C); and immobilized active agent (D) [34].

The multilayer method is expensive, so its use is not as extensive as the coating and sachet method [34]. Apicella *et al.* [70] reported research on an oxygen scavenger packaging system made with a 4-layer film set using the multilayer method with laboratory-scale co-extrusion cast-film equipment. The packaging film uses a PET layer incorporated with Amosorb DFC 4020 as an active oxygen scavenger agent (active layer) and Amosorb DFC 4020 inserted between 2 inert PET layers as a passive oxygen scavenger agent (inert layer). The active multilayer film has a longer exhaustion time than the monolayer. The exhaustion time increases as the thickness of the inert layers increases, while the remaining oxygen concentration decreases with the increase in the thickness of the active layer. Enzyme-based active packaging is usually carried out by immobilizing the enzyme in the packaging material component (Figure 3D) [34]. Winestrand *et al.* [71] reported a study of co-immobilization of oxalate oxidase and catalase as oxygen scavengers in packaging film systems made of latex polymers, oxalate oxidase immobilized in film polymers is very well used with two dual properties as oxygen scavengers and oxalic acid, while the addition of catalase in packaging films are effective in preventing the release of hydrogen peroxide compounds.

Many types of active agents can be used in the oxygen scavenging system (OS), such as iron and other metallic scavengers, ascorbic acid, gallic acid, types of antioxidants, quinones, catechols, hydroxylamines, and ketoximes and other natural OS agents, enzymatic scavengers such as oxalate oxidase, catalase, glucose oxidase, and Photosensitive dyes [34,69,71]. Oxalate oxidase can act as an oxygen scavenger and can also produce carbon dioxide protective packaging gas in the product [70]. The breakthrough nanoencapsulation of heat-sensitive active agents can blend with the packaging polymer matrix during the extrusion process [72].

5. Potential of biodegradable and oxygen scavengers packaging system based on bacterial cellulose

Biodegradable packaging breakthroughs are increasingly being developed to obtain mechanical properties as good as conventional plastic packaging. One way of developing Bacterial Cellulose (BC) is mixing with other biodegradable and renewable polymers to produce polymers with superior physical and chemical properties in packaging production. Biodegradable packaging materials are generally made in the form of packaging films. Likewise, oxygen scavengers are mostly made by being combined in a packaging film. Therefore, the combination of BC-based packaging films combined with oxygen-scavenging active agents has the potential to be applied in food packaging.

Bacterial cellulose (BC) biopolymers are suitable carriers for active packaging materials such as oxygen scavengers [20,73]. The application of BC as a film material incorporated with oxygen scavenging agents will open new alternatives to active packaging. Active packaging allows the shelf life of food in packaging to be longer, and the quality is maintained through the interaction of food with active ingredients on the package [53,74] so that the resulting packaging film has a dual function as an oxygen scavenger and is biodegradable.

6. Conclusion

Bacterial Cellulose has excellent potential as raw material for making biodegradable packaging films or bioplastic. The superior basic properties supported by polymer modification treatment will produce a film with a good polymer matrix. The development of the dual function of bacterial cellulose packaging film can be done by adding an oxygen scavenger function with the addition of an oxygen scavenging agent according to the properties of the bacterial cellulose polymer.

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