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Review

Antimicrobial packaging based on starch, poly(3-hydroxybutyrate) and poly (lactic-co-glycolide) materials and application challenges

Nichrous Mlalila[a,c,*], Askwar Hilonga[b], Hulda Swai[a], Frank Devlieghere[c], Peter Ragaert[c]

[a] School of Life Sciences and Bioengineering, Nelson Mandela African Institution of Science and Technology, P.O. Box 447, Arusha, Tanzania
[b] Department of Materials Science and Engineering, Nelson Mandela African Institution of Science and Technology, P.O. Box 447, Arusha, Tanzania
[c] Laboratory of Food Microbiology and Food Preservation, Department of Food Safety and Food Quality, Faculty of Bioscience Engineering, Ghent University, Coupure links 653, B-9000 Ghent, Belgium

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ABSTRACT

Background: In recent years, food packaging has focused on two scientific pillars; adopting the biodegradable packaging materials and development of antimicrobial packaging for extended shelf life, quality and safety of food products. The bioplastic materials provide a promising application in the packaging industry to substitute environmentally deleterious petrochemical-based plastics.

Scope and approach: This paper gives insights to very recent progress on the antimicrobial application of starch, polyhydroxybutyrate (PHB) and poly (lactic-co-glycolide) (PLGA) as well as their blends and nanocomposites in food packaging research. It also presents an overview of the antimicrobial application of these materials particularly in food and biomedical industry.

Key findings and conclusions: PHB, starch and PLGA materials have unique properties towards novel application in foods, cosmetics, medicines as well as various composites. The materials necessitate critical studies to improve their industrial performance both for processing engineering and antimicrobial packaging due to functional and technical limitations.

1. Introduction

Antimicrobial packaging refers to the integration of an antimicrobial agent into packaging systems for the purpose of preventing microbial growth on food products and extending its shelf life (Sung et al., 2013; Wang et al., 2015). Antimicrobial packaging aims to ensure safety, quality and extension of shelf life of food products by preventing or retarding the growth of the spoilage and/or pathogenic organisms (Lavoine et al., 2014). The package materials may acquire antimicrobial activity (1) by incorporating antimicrobial components in a polymer matrix, (2) surface irradiation of polymer matrix which produces reactive oxidizing species, (3) by gas emission/flush through modified atmosphere packaging (Fortunati et al., 2013) or (4) by using inherently antimicrobial polymer resins. The use of antimicrobials in food packaging systems has been motivated with the increasingly global food-borne outbreaks with related to health and safety concerns (Ehivet, Min, Park, & Oh, 2011; Kim & Rhee, 2016). In addition, the stability challenges of traditional preservatives in food packaging systems due to their reactions with food matrix have augmented the development of novel packaging development (Hill, Taylor, & Gomes, 2013). Furthermore, the consumer preferences for fresh, minimal processed foods, minimum allowed or chemical-free food products, have also contributed to the interest of antimicrobial packaging (Kuorwel, Cran, Sonneveld, Miltz, & Bigger, 2011a; Moon & Rhee, 2016; Narayanan, Neera, Mallesha, & Ramana, 2013; Ollé Resa, Gerschenson, & Jagus, 2014).

Antimicrobial packaging has two main categories, namely migratory and non-migratory packaging systems. Migratory packaging system allows the reversible release of non-volatile or volatile active components from polymer matrix to food constituents or packages headspace by diffusion and/or partition at interfaces. In the non-migratory system, the active component is irreversibly tethered to the package's surface and no diffusivity of the antimicrobial agents occur (Azlin-Hasim, Cruz-Romero, Morris, Cummins, & Kerry, 2015b; Kugel, Stafslien, & Chisholm, 2011). The migratory system favors lower concentrations of preservatives in food matrix as only small doses are released over an extended period from the package. In the non-migratory system, the preservative acts at the food surface only where microbial growth occurs without migrations. Enzymes, proteins peptides, chitosan, UV irradiated nylon, poly (ethylene glycol) (PEG), polylysine and polylactic acid (PLA) with polyanion backbones are antimicrobial agents which can be used in the model packaging system (Kugel et al.,

* Corresponding author. School of Life Sciences and Bioengineering, Nelson Mandela African Institution of Science and Technology, P.O. Box 447, Arusha, Tanzania.
E-mail address: nichogm_2006@yahoo.com (N. Mlalila).

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2011). Common antimicrobial packaging systems include inserts of sachets/pads into the headspace, polymer formulated homogeneously with antimicrobial agents, coating and immobilization of antimicrobial agent to the surface of the package by grafting. Antimicrobial packaging offers more advantages over traditional systems of directly mixing of preservatives to the processed food product. These additives can be deactivated in the food matrix or migrate out of the food surface towards locations in the food where initial microbial attacks are not taking place (Jalvandi et al., 2015; Sung et al., 2013; Takala, Vu, Salmieri, Khan, & Lacroix, 2013).

The antimicrobial agents used for preservation of the processed food products are either chemically synthesized or downstream extracted from biomass of plants, animals and microorganisms. The conventional chemical preservatives including organic acids and their salts such as potassium sorbate (KS), propionic acid, benzoic acid, ascorbic acid, citric acid, acetic acids and alcohols are predominant food preservatives due to their low prices and affordability. However, the research has surged on to replace them with natural antimicrobial agents (Hill et al., 2013; Ollé Resa et al., 2014) such as enzymes (Ghalanbor, Körber, & Bodmeier, 2010), chitosan, bacteriocins and natural extracts and essential oils (EOs) from plants (Gabriela et al., 2010; Pereira et al., 2015; Tomé et al., 2012). The natural antimicrobial agents have broad-spectrum actions against most of pathogenic and food spoilage microorganisms (Efrati et al., 2014; Kim & Rhee, 2016). Additionally, nanomaterials from titanium (El-Wakil, Hassan, Abou-Zeid, & Dufresne, 2015), magnesium, copper, silver (Fortunati et al., 2013; Mlalila, Swai, Hilonga, & Kadam, 2017b), platinum, gold (Mlalila, Kadam, Swai, & Hilonga, 2016; Stevanović, Uskoković, Filipović; Škapin, & Uskoković, 2013) and zinc (Díez-Pascual & Díez-Vicente, 2014) have also received attention in antimicrobial food packaging due to their large surface-volume ratio (Benhacine, Hadj-Hamou, & Habi, 2015).

Another vibrant research topic within food packaging is performance improvement of biopolymers for packaging through diverse modification techniques. Together with biobased antimicrobial packaging, this has increased the number of publications focusing on the antimicrobial packaging. Most studied bioplastics recommended for applications in antimicrobial packaging include poly (butylene succinate) (PBS), starch, cellulose derivatives, PLA and PHB because of their compatibility with a variety of antimicrobial compounds (Narayanan, et al., 2013; Nicosia et al., 2015; Sung et al., 2013; Tawakkal, Cran, & Bigger, 2016). Starch is one of the richest natural food sources available in most of the plants, commercially available, relatively inexpensive and is highly biodegradable (El-Wakil et al., 2015; Kuorwel et al., 2011a). Starch is one of the most promising biopolymers as biobased packaging material. It is readily available as carbohydrate resource from renewable stock feeds in cereals and tubers, edible, nontoxic biopolymers, nonpolluting, biocompatible with other biopolymers and has excellent film-forming behavior through plasticity. The modification of natural starch by thermal conversion into thermoplastic starch (TPS) in presence of plasticizers and other additives improves its processability in conventional equipment. Further addition of starch nanocomposites and composites improves the mechanical performance, water vapor permeation (WVP), optical, structural and morphological properties of the starch matrix (Abreu et al., 2015; Díez-Pascual & Díez-Vicente, 2014; Rhim, Park, & Ha, 2013).

PHB is an active member of polyhydroxyalkanoates (PHAs) family known for its good biodegradability, biocompatibility and being bioderived from renewable resources by bacterial synthesis (Solaiman, Ashby, Zerkowski, Krishnama, & Vasanthan, 2015). The 3-hydroxybutyrate, a monomer of PHB, is an ordinary metabolic product in living organisms. PHB has relative promising properties including melting point, glass transition temperature, moisture barrier, mechanical performance and crystallinity which are analogous to some widely used polyolefins like polypropylene (PP), polyethylene (PE) and (polyethylene terephthalate (PET) (Xavier, Babusha, George, & Ramana, 2015). Poly (lactic-co-glycolide) (PLGA) is an aliphatic linear copolymer of polylactic acid (PLA) and polyglycolic acid (PGA) (Makadia & Siegel, 2011). The copolymer is biodegradable, has the flexible morphology of crystallinity and amorphous states, high barrier properties, high compatibility and has been approved by US Food and Drug Administration (FDA) as food contact material (McConville, Tawari, & Wang, 2015). In addition, PLGA has temperature-responsive and self-healing properties and its common copolymers of PEG such as diblock (PLGA-PEG) or triblock of ABA (PLGA-PEG-PLGA) or BAB (PEG-PLGA-PEG) can be easily tailored for active and intelligent packaging (Huang, Mazzara, Schwendeman, & Thouless, 2015; Lee, Towslee, Maia, & Pokorski, 2015).

These peculiar characteristics of starch, PHB and PLGA have inspired their advanced studies and characterizations for applications in food packaging industry. Currently, starch and PHB are among the most used bioplastics, being both compostable and biobased, in food packaging industry that needs further research. In the market, starch and PHB materials are available as pure materials or blended with petrochemical-based plastics for packaging purposes (Avérous & Pollet, 2012; Plackett & Siro, 2011, pp. 498–526). Additionally, PLGA is a most researched polymer in the biomedical industry used in tissue engineering, development of drug delivery systems and responsive materials. In drug delivery systems, PLGA have shown attractive sustained delivery of active compounds and produces responsive materials, which need further investigations of their applicability in the food industry (Gabriela et al., 2010; Hill et al., 2013; Huang et al., 2015; McConville et al., 2015).

This review is restricted to these three bioplastic polymers being starch, PHB and PLGA as potential packaging materials with respect to antimicrobial food packaging applications. The first part gives the details of individual materials with respect to antimicrobial packaging based on recent publications. Information on antimicrobial packaging engineering, antimicrobial performance maximization and the limitations of antimicrobial packaging has been synthesized further in this review. The features described in the paper give new insights on successful design and applications of antimicrobial packaging systems in the food industry.

2. Antimicrobials packaging applications

Starch and starch derivatives, PHB and PLGA have many features compatible with many antimicrobials agents for packaging. They have been scrutinized for both pathogenic and spoilage microorganisms in biomedical and food applications at different testing conditions (Table 1). In laboratory scale, the preparation of active films is commonly carried-out by solvent casting method (Abreu et al., 2015; Bhatia & Bharti, 2015). Most of the films tested for release kinetics of antimicrobial agents have shown initial burst release of antimicrobial components from polymer matrix followed by extended slow release profile, as discussed in section 2.3. The films have demonstrated release of antimicrobial agents and worked effectively against target microorganisms. However, PLGA studies have been focused mainly on biomedical applications and have attractive results to be applied in the food industry (Gomes, Moreira, & Castell-Perez, 2011; Prabhakaran, Zamani, Felice, & Ramakrishna, 2015; Yüksel & Karakeçili, 2014).

2.1. Starch-based materials

Starch is compatible with many antimicrobial agents and its active films have shown efficacy against many species of microorganisms. Sweet potato starch films was successfully blended with chitosan and KS (Shen, Wu, Chen, & Zhao, 2010) and the active films were observed to suppress effectively the growth of *E. coli* and *S. aureus* and improve the barrier properties of films (Arrieta et al., 2014; Barzegar, Azizi, Barzegar, & Hamidi-Esfahani, 2014; Basch, Jagus, & Flores, 2013; Tomé et al., 2012). When various ratios of chitosan and lauric acid were blended in starch, the active films improved in efficacy against *B.*