Plastics in Food Packaging

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Key terms

The key term for active packaging is to extend the food product's shelf life via the addition of active agents whereas intelligent packaging integrating technology such as sensor to indicate the food quality. Biodegradable polymer derived from biomass is a promising candidate to replace fossil-based plastics for greener food packaging.

Introduction

Among paper, metal, glass and plastic, plastic is the most widely used material for food packaging (Markets and markets, 2014). The continued growth of plastics to be the top choice material in food packaging are contributed to its relative low material cost, light weight and versatile characteristics such as can be flexible or rigid, transparent or opaque, heat sealable, and easy to process into different sizes and shapes (Berk, 2013; Marsh and Bugusu, 2007).

The total production of the plastics in the world has reached 360 million tonnes in 2018, as compared to 348 million tonnes in 2017 (PlasticsEurope, 2019). Among the plastics produced are thermoplastics and thermosets. According to the plastics demand by segment in Europe, the packaging represented the biggest end user-market of nearly 40%, followed by building and construction of 19.8% (PlasticsEurope, 2019). There are more than 30 types of plastics available in the market for packaging, the leading plastics are from the polyolefins family, such as polypropylene (PP) and polyethylene (PE) (Raheem, 2013). PP and PE are commonly used as food wrappers, films, beverage bottles, container etc, as shown in **Fig. 1**. The other plastics for food packaging including polyvinyls and polyesters family, such as polyvinylchloride (PVC), polyethylene terephthalate (PET) and polystyrene (PS), which are used for food tray, bottles, jars, hot beverage cups etc (Marsh and Bugusu, 2007). **Fig. 2** shows the water bottle made from PET.

The conventional roles for plastics as food packaging including to contain the food, protect food from contamination, display the food composition information to consumer and convenient for handling or transportation (Kuswandi and Jumina, 2020; Müller and Schmid, 2019). As the technology advances, the plastic food packaging provides more functions and features than before, such as the evolution of the smart packaging, which is referring to the active and intelligent packaging system (Kuswandi and Jumina, 2020). The plastic food packaging can extend the food product's shelf life via the addition of active agents such as oxygen scavengers and antimicrobial agent. It can also indicate the food quality after integrate technology such as sensors into the

Fig. 1 Plastic food containers made from polypropylene.

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Fig. 2 PET water bottle.

intelligent packaging. The global smart packaging market was valued at around USD 37921.56 million in 2018 to USD 58791.56 million by the end of 2025 at a Compound Annual Growth Rate (CAGR) of 6.46% (360iResearch, 2020). Besides the smart packaging, the recent development of biodegradable polymers as food packaging will be discussed as well.

Active Packaging

Active packaging is an approach to incorporate active agents such as antioxidants or antimicrobials that can maintain or pro-long the food product shelf life. It can be divided into active scavenging system and active releasing system, which is referring to the absorb or release of substances into the food or its environment (Yildirim *et al.*, 2018). The food spoilage happens when microbiological, chemical, or physical changes occur, making the food product becomes unsuitable for human consumption. The growth of microorganisms such as molds, yeasts and bacteria produce enzymes that lead to objectionable by-products in the food. Chemical food spoilage generally refers to the oxidation process and enzymatic browning, while the physical food spoilage refers to the gain or loss of moisture such as moist food becomes excessive dehydrated or dried food absorbed moisture (Benner, 2014). The absorbers remove unwanted components that can cause deterioration such as oxygen, moisture, ethylene from the food or its environment whereas emitters are releasing compounds such as antimicrobial, carbon dioxide to the packaged food (Kumar *et al.*, 2018). Traditionally, the active compounds are added to the bulk of the food, and the effectiveness of the active compounds maybe reduced or inhibited because of interaction between the active compounds and the food components. The incorporation of active compounds into the sachets, pads or adhesive labels within the food packaging is also posing risks such as accidental leakage of active compounds, or ingestion of the active compounds after the sachet rupture (Han *et al.*, 2018). The addition of active compounds into the plastics to create active packaging not only can reduce the amount of substance used, but also ensure food safety besides enhancing the food product shelf life (Yildirim *et al.*, 2018).

Oxygen Scavengers

Plastics generally provide a relatively poor barrier to the diffusion of oxygen in air. As a result, in addition to the oxygen that is initially present in the packaging, more oxygen may permeate into the packaging over time. The present of oxygen in the food packaging can promote the oxidative deterioration in food, growth of aerobic microorganisms, undesired color change, off-odors or flavors (rancid food due to lipid oxidation) or even lead to nutritional losses (Biji *et al.*, 2015; Mohan and Ravishankar, 2019). Oxygen scavengers is also known as oxygen absorbers react with the free oxygen trapped in the packaging to form stable compounds, and the oxygen scavengers can be found in the form of metallic, organic or inorganic (Solovyov, 2014). The most widely known oxygen scavengers are iron, ascorbic acid and its derivative, enzymes such as glucose oxidase, salts and fatty acids, such as oleic acid. These scavengers can be used individually or in combination, either in sachets form insert into plastic packaging or directly incorporate into the plastic packaging structure (Kuswandi and Jumina, 2020). Investigation has been carried out on oxygen scavenger plastic film, PE filled with iron-based oxygen scavenger masterbatch, showed certain protection on the sausage via the absorption of oxygen in headspace against food deterioration (Gibis and Rieblinger, 2011). Plant based extracts, *Satureja thymbra* was coated on a laminated film used as active packaging, showed the decrease of oxygen in the packaging and successfully pro-long the shelf life of the fried potato chips. The addition of natural extracts into the packaging material is more favorable in delaying the fried chips deterioration than adding the extracts to the frying oil or the chips itself (Choulitoudi *et al.*, 2020).

Moisture Absorbers

Moisture control in food packaging is crucial, as the accumulation of excess moisture inside the package can promote the growth of microorganisms and lead to food spoilage. The water can be accumulated in the food packages due to the temperature fluctuations along the supply chain, drip loss of tissue fluid from fresh foods and transpiration of horticultural product (Kuswandi and Jumina, 2020). The moisture absorbers that are widely used in food packaging including inorganic absorber such as silica gel and natural clays, whereas organic forms are sorbitol and fructose. The polymer-based moisture absorbers including starch copolymers, polyvinyl alcohol, polyacrylate salts and carboxyl methyl cellulose (Gaikwad *et al.*, 2019; Kumar *et al.*, 2018). There are two types of water absorption mechanism, either by physical adsorption or chemical reaction. Silica gel with porous structure is an example of physical adsorption, whereas the chemical absorber, calcium oxide reacts with water via chemical reaction (Chen, 2017). The moisture absorbers are commonly used in the form of sachets, pads, or incorporated into the plastic material to form tray or film for food packaging. **Figs. 3** and **4** show the usage of silica gel sachet for moisture control in dried nuts. Chen *et al.* (2018) developed an active multifunctional film which incorporated the green tea extract into polyvinyl alcohol to prevent the dried eel from absorbing moisture and oxidizing of lipid (Chen *et al.*, 2018). In a study of desiccant film made of low-density polyethylene (LDPE) filled with silica gel powder, the result showed the desiccant film can absorb up to 0.08 g water vapor per 1 g of film (Sängerlaub *et al.*, 2019). The addition of moisture absorber or other active compound into the plastic material structure is of high interest as it can reduce the risk of sachet rupture, or accidentally consume the sachet as part of the food, and free up the space inside the pack.

Antimicrobial Agents

The use of antimicrobial agents in active packaging has created a lot of research interest, as it can control the growth of pathogenic microorganisms on the surface of the food by extending the lag phase and reducing the growth rate or decreases live counts of microbials (Han, 2000). Besides the addition of antimicrobial agents (e.g., benzoic acid, sorbic acid, nisin, lysozyme, essential oil and silver) into the packaging structure or in the form of sachet, the antimicrobial function can be achieved via the use of antimicrobial polymers such as chitosan film and ε-poly-L-lysine (Han, 2003; Otoni *et al.*, 2016; Sayed and Jardine, 2015). The polymer matrix that has been used to blend or coat with the antimicrobial agents including PE/LDPE, PVC, PS, ethylene vinyl alcohol (EVOH) etc., which are mainly derived from fossil resource (Carbone *et al.*, 2016; Catalá *et al.*, 2016; Huang *et al.*, 2019) The antimicrobial effects can happen in one of three methods: release, absorption or immobilization. The release mode involves the migration of antimicrobial substrate into the foods or headspace inside packaging, whereas the absorption mode removes the essential factors such as oxygen and moisture that allow the microbial growth, and the immobilization system requires direct contact between the packaging material with food product, as it does not release antimicrobial substrate but suppress the growth

Fig. 3 Silica gel sachet in the dried nuts.

Fig. 4 Silica gel changed color after absorbed moisture.

of microorganisms at the contact surface (Han, 2003). Active films made from the renewable polymer, poly(lactic acid) impregnated with herbal essential oils showed effective antibacterial activity and potential to use for modern active food packaging (Talebi *et al.*, 2018). The other biopolymers such as polysaccharide, poly(hydroxyalcanoates) and polyurethanes are also promising candidates to be modified or incorporated with antimicrobial agent to produce sustainable food packaging with antimicrobial activity (Muñoz-Bonilla *et al.*, 2019). However, many factors need to be considered in the design of the antimicrobial packaging system, such as the resistance of microorganism, chemical nature of foods and antimicrobials, physical and mechanical properties of the packaging material, processing condition of the packaging, toxicity of the antimicrobial agents and the corresponding regulations (Han, 2003).

Antioxidant Releasers

Oxidation is a chemical reaction where the food reacts with the oxygen and this leads to food spoilage such as rancidity of fats and discoloration of food (Gibis and Rieblinger, 2011). The antioxidant releasers which can be derived from synthetic or natural source are added into food packaging, and later release to the packaging headspace to inhibit the oxidation process. The antioxidant active packaging is developed via the addition of antioxidant releasers such as butylated hydroxy-toluene (BHT) and butylated hydroxyanisole (BHA) into the polymer matrix or as a coating on polymer surface to prevent lipid oxidation (Yildirim *et al.*, 2018). Various types of polymers have been used for the development of antioxidant packaging including polyolefins (e.g., LDPE, PP) and biopolymers (e.g., PLA, cellulose acetate) to extend the shelf life of oxidation-sensitive foods (El Fawal *et al.*, 2019; Jamshidian *et al.*, 2012; Lee and Yam, 2013; Yildirim *et al.*, 2018) The synthetic antioxidants are released into the packaged food by control mechanism of diffusion, it terminates the free radical chain reactions by donating hydrogen to produce a more stable compound (Nwakaudu *et al.*, 2015; Soto-Cantú *et al.*, 2008). However, the trend for using the natural antioxidants (e.g., tocopherol and plant extracts) is growing due to the health concern raised over the use of synthetic antioxidants (Dalla Rosa, 2019; Lee and Yam, 2013; Nieva-Echevarría *et al.*, 2015).

Intelligent Packaging

Active packaging discussed in Section "Active Packaging" refers to the incorporation of active compounds into the packaging system to extend the product shelf-life. Intelligent packaging, on the other hand, refers to the ability of the food packaging to monitor the freshness, quality, and safety of the food inside the package and to communicate this information to the consumers. In addition, intelligent packaging allows the tracking of the quality of the food from the source to the customers. The monitoring is usually carried out using sensors or indicators. A sensor is a tool or device to determine, detect or quantify matter or energy and give a response or signal for the determination or measurement of a chemical or physical characteristic (Kerry *et al.*, 2006; Kress-Rogers, 1998; Kuswandi and Jumina, 2020) whereas an indicator is a tool or device that indicate the absence, presence, or concentration of an analyte, or the reaction degree between two or more analytes by a color change (Hogan and Kerry, 2008;

Kuswandi, 2017; Kuswandi and Jumina, 2020). To keep the discussion within the topic of this encyclopedia, plastics and polymers, the following section will focus on sensors and indicators that incorporate the use of polymeric materials, either as the main substrate or the sensing element.

Sensors

Sensors can be categorized into chemical sensors, gas sensors, and biosensors. Chemical and gas sensors use chemical reagents as receptors that can detect changes in pH, volatile organic compounds such as amines, and gases such as oxygen, carbon dioxide, hydrogen, and ammonia whereas biosensors are analytical devices that detect, record, and transmit information pertaining to biochemical reactions (Kuswandi and Jumina, 2020; Yam *et al.*, 2005). Sira Technologies Inc. and Toxin Alert Inc. developed and commercialized biosensors that can be incorporated into plastic packaging film to detect pathogens (Yam *et al.*, 2005). A visual alert is displayed when pathogens are detected by the specific antibody immobilised in the substrate. Currently, commercial biosensors are available for clinical diagnostics with that for smart packaging mostly still in the prototype or research phase.

Research has shown that conducting polymer composites can be used to detect an array of gases and odours released during microbe metabolism (Kuswandi *et al.*, 2011). Arshak *et al.* (2007) utilized polyaniline (PAN) grafted to lignin and polyethylene adipate (PEA) and Hypermer™ PS3 polymeric surfactant with PAN loaded carbon black as sensors. The sensors could detect the gases released by food borne pathogens such as Salmonella spp., Bacillus cereus, and Vibrio parahaemolyticus. In addition, enzyme can be immobilised in polymers to create biosensors. Polyphenol oxidase immobilised in polypyrrole (PPy) have been shown to be able to detect the herbicide atrazine (Kaoutit *et al.*, 2004). Amperometric xanthine biosensor made from chitosan–polypyrrole–gold nanoparticles with immobilized xanthine oxidase were used to detect xanthine, which is an indicator for fish and meat spoilage and freshness determination (Dervisevic *et al.*, 2017). Another xanthine biosensor based on electrochemically polymerized 10-[4H-dithieno(3,2-b:2,3-d)pyrrole-4-yl]decane-1-amine film was also used successfully to detect xanthine (Dervisevic *et al.*, 2016).

Polymers are also used as sensitive layers and coatings in surface acoustic wave (SAW) sensors which can be used to detect gases and vapors (Garcia *et al.*, 2006; Penza *et al.*, 2001), and bacterial contamination such as E-Coli (Lamanna *et al.*, 2020). This type of sensors is seen as more promising for use with radio frequency identification (RFID) to transmit food quality data wirelessly (López-Gómez *et al.*, 2014). Polymer-based RFID will be discussed in Section "Polymer-Based RFID".

Indicators

Polymer and polymer composites can be used as indicators such as time temperature indicators (TTI), gas indicators and freshness indicators. Compared to sensors, indicators for food packaging are more readily available commercially. Fresh-Check by Temptime Corporation (2018) is based on polymerization reaction of diacetylene (Kuswandi *et al.*, 2011; Kuswandi and Jumina, 2020). When exposed to high temperature, the polymer changes color from light to dark and can be used to show the freshness of the food. 3M's MonitorMark (2020) which is based on diffusion of dye, have included the use of rubbery polymers such as polybutadiene (He *et al.*, 2015).

Besides commercial products, other polymeric materials have been used in research. Fish spoilage could be monitored by entrapping cellulose acetate with pH sensitive dye such as sodium salt (Pacquit *et al.*, 2006). Copolymer of ethylene and norbornene mixed with small percentage of photoluminescent was used as TTI for higher temperature ($>140^\circ$C) requirement (Lee and Shin, 2012). Active chitosan/polyvinyl alcohol (PVA) films with anthocyanins from red cabbage (Pereira *et al.*, 2015) could potentially be used to indicate milk freshness. Nafion film (sulfonated tetrafluoroethylene-based fluoropolymer-copolymer) containing crystal violet was successfully used to monitor the fermentation process of tape ketan (Indonesian fermented steamed glutinous rice product) (Kuswandi and Jember, 2020). Wheat gluten/chlorophyll film was used to increase the shelf life of sesame oil and in the same time indicate the oil expiration due to oxidation by color change (Chavoshizadeh *et al.*, 2020). Rahimah *et al.* (2020) used dragon fruit peel dye mixed with PVA to measure the pH of goat milk, which can indicate the freshness of the milk. Pirsa *et al.* (2020) used biopolymer chitosan with pomegranate peel extract and Melissa officinalis (lemon balm) essences to detect changes in the pH of cream cheese.

Polymer-Based RFID

The sensors and indicators mentioned before this require direct visual readout, for example, changes in color, to determine food quality or freshness. It would be an added advantage to adopt electronic readout and transmission of data wirelessly (Ahmed *et al.*, 2018; Zhou *et al.*, 2020). In addition, radio frequency identification (RFID) can provide product tracking from source to the consumer and in the same time store various data such as origin, temperature history and real-time location. Huang *et al.* (2012) developed a flexible pH sensor embedded in a batteryless radio-frequency (RF) transponder. The pH sensor was made of iridium oxide and silver chloride electrodes integrated onto polyimide substrate. Zhou *et al.* (2020) integrated silicon RFID with ammonia and anti-open surface acoustic wave (SAW) sensors using conducting polymer. Dimethyl sulfoxide (DMSO) doped poly(3,4-ethylene dioxythiophene): poly(styrenesulfonate) (PEDOT: PSS) composite films with silver nanowires (AgNWs) is used as ammonia sensitive layer. Using conventional passive RFID tags modified with chemically sensitive conductive composites, Fiddes *et al.* (2014) successfully detected different types of biogenic amines associated with food spoilage. The conductive composite consists of carbon black, maleic anhydride, and poly(ethylene-*co*-vinyl acetate) (PEVA) as the sensing material.

In order to find wider applications in food packaging, the sensor have to be more robust and cost-effective.

Biodegradable Polymers for Food Packaging

One of the trends in recent decades related with plastics and polymers for food packaging application is the utilization of biodegradable polymers (Din *et al.*, 2020; Siracusa *et al.*, 2008). The waste of food packaging materials are often contaminated with food products, limiting the recycles activities for these materials, and therefore resulting in large amounts of plastic packaging waste (Muthuraj *et al.*, 2018; Siracusa *et al.*, 2008). Therefore, utilizing biodegradable polymers for food packaging, will help in lessening environmental burden. In this section, recent progress on application of biodegradable polymers for food packaging will be highlighted.

Biodegradable polymers are polymers that can be degraded to carbon dioxide, water, and biomass, by the action of microorganisms, once they end up in the environment (Haider *et al.*, 2019; Iwata, 2015). As shown in **Table 1**, they can be broadly classified into three categories, i.e., (1) polymers derive from biomass, such as starch, chitosan, alginate, etc., (2) synthetic biodegradable polymers, that may originate from renewable resources such as poly(lactic acid) (PLA) or petroleum based materials such as polycaprolactone (PCL), and (3) polymers produced by microorganisms, such as polyhydroxybutyrate (PHB) (Din *et al.*, 2020; Doppalapudi *et al.*, 2014; Mangaraj *et al.*, 2019; Rhim *et al.*, 2013; Siracusa *et al.*, 2008).

Polymers Derive From Biomass

One of the polymers derive from biomass that has gained a lot of attention in recent is starch. Starches are abundantly available and one of the low-cost groups of biodegradable polymers. It is consisted of amylose and amylopectin with a range of around 10%–30% and 70%–90%, respectively, depending on the source. Various kind of starches like potato, corn, rice, and cassava are used for the preparation of biodegradable polymers (Balakrishnan *et al.*, 2017; Khan *et al.*, 2017; Mangaraj *et al.*, 2019; Medina-Jaramillo *et al.*, 2017). However, native starches are brittle, and they need to be plasticized to have deformable materials called thermoplastic starches (TPS). In order to enable them to be used as food packaging, the TPS are generally blended with other polymers, as well as incorporated with nanoparticles. For example, TPS may be blended with PCL and PLA to improve their mechanical properties (Guarás *et al.*, 2015; Muller *et al.*, 2017), and nanoclays can be dispersed in TPS based materials to reduce the water vapor transmission rate, improving their barrier properties (Guarás *et al.*, 2016; Park *et al.*, 2003; Rhim *et al.*, 2013).

Synthetic Biodegradable Polymers

Among various types of biodegradable polymers, particular interest has been paid to PLA, especially when it is used as packaging materials, including food packaging. Because in addition of being thermoplastic, biodegradable, compostable, and produced from renewable resources, PLA shows comparable performance in terms of mechanical, thermal, and barrier properties to conventional synthetic polymers, such as polystyrene and poly(ethylene terephthalate) (Siracusa *et al.*, 2017, 2012). One limitation of this polymer is its brittleness in unmodified form, therefore in many instances exploring the application of PLA as food packaging

Table 1 Classification of biodegradable polymers

Category	*Origin*	*Examples*
Polymers derive from biomass	Plants	Starch Cellulose Gluten
	Animals	Chitosan Collagen
	Algae	Alginate Carrageenan
Synthetic biodegradable polymers	Renewable resources (plants)	Polylactic acid (PLA)
	Petroleum based	Polycaprolactone (PCL) Polyvinyl alcohol (PVA)
Polymers produced by microorganisms	Microorganisms (bacteria)	Polyhydroxybutyrate (PHB) Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) Pullulan

materials, blends of PLA, or introduction of foreign substances such as plasticizers into PLA are commonly reported (Arrieta *et al.*, 2014a,b; Muthuraj *et al.*, 2018).

Besides PLA, PCL has also been used for food packaging applications due to its biodegradability and high flexibility. However, its high cost still limits this material to be widely used in broader packaging applications. Various attempts to reduce costs while preserving or improving the biodegradability and mechanical properties of the resulting materials include blending it with other polymers such as starch (Guarás *et al.*, 2015, 2016) and chitosan (Cesur *et al.*, 2018; Joseph *et al.*, 2011), as well as incorporating natural fiber into it (Ludueña *et al.*, 2012).

Polymers Produced From Microorganisms

Polyhydroxyalkanoates (PHAs) family, such as PHB and PHBV are the biodegradable polymers fall under this category. These polymers can be produced in nature by bacterial fermentation of sugar and lipids (Bugnicourt *et al.*, 2014; Din *et al.*, 2020; Mangaraj *et al.*, 2019). Due to their high biodegradability in different environments, as well as better barrier properties and mechanical strength compared to other more widespread biodegradable polymers such as PLA, this class of polymers show promising properties to be applied in wide range of application, including food packaging. Similar to PLA, their inherent brittleness, it is common to blend or copolymerize them with other polymers or additives. Nevertheless, the high cost of commercial PHAs still limits these materials to penetrate a broader market (Bugnicourt *et al.*, 2014).

Other type of polymer in this category which having potential applications in the field of food packaging materials is pullulan. Pullulan is a microbial extracellular polysaccharide, obtained from different strains of *Aureobasidium spp*. Beside biodegradable, biocompatible, non-toxic, non-carcinogenic non-mutagenic, and edible, it is also transparent with good film-forming abilities (Kraśniewska *et al.*, 2019). However, the use of a pure pullulan film has been restricted by its hydrophilic nature, brittleness and breakability (Silva *et al.*, 2018), as well as its high cost (Tabasum *et al.*, 2018). Modification are thus needed to overcome the disadvantages of pure pullulan, such as incorporating glycerol as a plasticizer to improve the flexibility and elongation of break of the produced materials (Trovatti *et al.*, 2012), adding hydrophobic substances such as rice wax to improve the water barrier properties (Shih *et al.*, 2011), and other techniques.

Conclusion

Plastics food packaging play significant role in the preservation of the world's resources. The use of plastic packaging protects the food product against contamination from any physical, chemical, or biological source, reducing the food waste and ensuring the food quality and safety along the food chain. Plastics can be used as main substrate for packaging, ingredient in active compounds, or sensing elements in preserving the food product. The use of biodegradable polymers, particularly derived from renewable resources have been of great interest, it is viewed as environmentally friendly material with lower carbon dioxide emission and alternative for the current fossil-based polymers in food packaging. The food packaging industry is undergoing rapid changes with more extended functions to prolong the shelf life of the food. The future development will continue to consider the environmental impact such as sustainable material including the recycle of packaging material, and the integration of advanced technologies into the packaging to develop communication link between the food producer and consumer.

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