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Development of novel active polypropylene based packaging films containing different concentrations of sorbic acid

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ABSTRACT

Active polypropylene (PP) based packaging films containing sorbic acid (SA) at concentrations of 2, 4 and 6% w/w were prepared using extrusion molding method. Mechanical, physical, spectroscopy and microbial properties of the films were evaluated. The results showed a significant ($P < 0.05$) improvement in tensile strength and depressing in elongation at break. Incorporating higher contents of SA diminished water vapor permeability and the transparency of the films. Nevertheless, PP-SA films showed UV absorption which increased significantly ($P < 0.05$) by increasing SA concentration to 4 and 6% (w/w). Color attributes (L*, a* and ΔE) of PP-SA films were not significantly ($P > 0.05$) different from those of control film. Increasing SA concentration to more than 2%, led to an increase in b* and yellowness index, while decreased whiteness index. PP-SA films showed antibacterial properties against *E. coli* and *S. aureus* as well as antifungal effects on *A. niger*. Antifungal effects of active films were more noticeable than their antibacterial effects.

1. Introduction

The plastic industry is developed in parallel with the extension of packaged food products. Flexibility, mechanical strength, recyclability, and most prominently, barrier properties of plastic packaging material have increased the demand for their application over typical packaging materials such as paper, jute, and glass (Lord, 2005; Motey & Lele, 2003). Different types of synthetic polymeric packaging materials such as polypropylene (PP), low density polyethylene (LDPE), polyvinyl chloride (PVC), ethylene vinyl alcohol (EVOH), ethylene acrylic acid (EAA) and ethylene vinyl acetate (EVA) are used in the preparation of packaging films for food applications (Beigmohammadi et al., 2016; Dehghani, Hosseini, & Regenstein, 2018; Fasihnia, Peighambardoust, & Peighambardoust, 2017; Han, 2000). Polypropylene (PP) is one of the most multipurpose packaging materials with extensive utilization due to its low density, high melting temperature, chemical resistance, recyclability, and enhanced mechanical properties (Gotsis, Zeevenhoven, & Tsenoglou, 2004; Manikantan & Varadharaju, 2011; Meira et al., 2014; Wan et al., 2013).

The principal objective of packaging is the preservation of food from microbial contamination, oxygen, moisture and light effects (Motey & Lele, 2003). Microbial contamination of food surface accounts for the most of food spoilage problems. The application of chemical preservatives in food has been a matter of grave concern for food safety; hence, active packaging, especially mostly-used polymer-based ones, is playing an imperative role in shelf-life extension of foods by eliminating the direct use of additives. Antimicrobial preservatives which are added to packaging materials can fulfill the purpose of overcoming such surface contamination (Weng & Chen, 1997). Antimicrobial food packaging as an active packaging serves as a novel methodology to prolong the microbial lag phase by retarding microorganisms' growth with the aim of extending packaged food shelf life (Barzegar, Azizi, Barzegar, & Hamidi-Esfahani, 2014; Buonocore, Conte, Corbo, Sinigaglia, & Del Nobile, 2005; Haghighi-Manesh & Azizi, 2017; Han, 2000; Persico et al., 2009; Suppakul, Miltz, Sonneveld, & Bigger, 2003).

Sorbic acid (SA) ($CH_3(CH)_4CO_2H$), a natural compound that was first isolated from berries in the 1800s, is a colorless solid with slight solubility in water (Allen & VanAllan, 1944). The conserving (antimicrobial) properties of SA were documented in the 1940′s and it became commercially available. Since then, SA (E200) and its salts (sodium, potassium, and calcium sorbate) have been extensively tested and used in food and drinks as preservative and antimicrobial agents

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either to inhibit the growth of microorganisms such as mold, yeast, and fungi, or to maintain freshness and wholesomeness (Liewen & Marth, 1985; Lück, 1990). Despite lower desirability of SA in comparison to the salts in food matrixes which is due to their higher solubility, the active form is the acid. The LD_{50}[1] value of SA (7.4 and 10 g/kg) demonstrates its low toxicity; hence, it is listed as a GRAS[2] additive in food by the FDA (Flores, Haedo, Campos, & Gerschenson, 2007; Winter, 2009). SA is relatively unstable and is swiftly degraded in soil; therefore, it is often considered environmentally friendly and used as a model additive in food packaging in previous (Baron & Sumner, 1993; Barzegar et al., 2014; Fleischberger, Buchholz, Archodoulaki, & Steiner, 2017; Guillard, Issoupov, Redl, & Gontard, 2009; Jipa, Stoica-Guzun, & Stroescu, 2012; Lück, Jager, & Raczek, 2000; Silveira et al., 2007; Weng & Chen, 1997). The direct incorporation of chemical and antimicrobial additives in industrially processed foods has been a focus of food safety-related investigations (Davidson, Taylor, & Schmidt, 2013; Degirmencioglu et al., 2011; Guynot, Marín, Sanchis, & Ramos, 2004; Uz & Altınkaya, 2011). Nowadays the growing demand for minimally-processed and preservative-free products necessitates the incorporation of the preservative agents to the packaging systems which can serve as a suitable methodology to achieve a low level of additives contact with the food (Dong & Manjeet, 2004; Baldevraj & Jagadish, 2011). The direct application of chemical preservatives in preparation of packaging films is an appropriate strategy to achieve antimicrobial activity in the films. Some of these additives are more effective when they are indirectly added as food preservatives (Baldevraj & Jagadish, 2011). There are reports in literature concerning active polymer-based films and coatings incorporating natural compounds such as nisin-protein (Lee, Son, & Hong, 2008), carvacrol and thymol (Ramos, Jiménez, Peltzer, & Garrigós, 2012), chitosan–propolis (Torlak & Sert, 2013), montmorillonite and nisin (Meira et al., 2014), *rhubarb* ethanolic extracts and *cinnamon* essential oil (Han, Wang, Li, Lu, & Cui, 2014), oregano essential oil and *Allium* extract (Llana-Ruiz-Cabello et al., 2017), and carvacrol (Krepker et al., 2018).

It is noteworthy that the selection of the antimicrobial agents depends on their compatibility with the packaging material or their heat stability during the extrusion process (Han & Floros, 1997; Baldevraj & Jagadish, 2011). Various natural and chemical antimicrobial agents have been used in different polymers and biopolymers employed in food packaging (above-mentioned). But due to the fact that each of these materials has a specific chemical structure and performance, they may not be compatible with each other; hence, a suitable choice of specific polymer and compatible additives will be very important. Therefore, polypropylene as one of the most applicable packaging material utilized in food industry, especially for those which are likely to be used or packaged in high temperatures could be selected. Considering the chemical properties of PP as well as its preparation method and temperature, sorbic acid as a highly effective anti-mold preservative comparing to other organic acids and even their salts could be chosen due to better compatibility with PP polymer matrix and high temperature tolerance antimicrobial additive. Furthermore, antimicrobial packaging films are often prepared by solvent or coating onto polymer surfaces. Solvent based methods could have environmental negative impacts and possible toxic substances in the film production; moreover industrializing their application requires industrial scale tests, which will be time-consuming and costly. Nonetheless, extrusion process with industrial equipment is used for the production of commercial food packaging films.

This paper aimed at developing new antimicrobial packaging films based on multipurpose packaging material incorporated with a GRAS and natural additive for further applications to extend the shelf-life of foods without the direct use of additives in the light consumer health concerns. Accordingly, the prominent purpose of this study was to develop PP-based antimicrobial packaging films as a multiuse packaging polymer containing different concentrations of sorbic acid used as a safe and effective organic acid served as the first novelty of the plan by the use of commercial and safe extrusion method for all types of packaging films in contact with food and eco-friendly (solvent-free green method) as the subsequent interest of the work. To reach this purpose the effect of the additive concentration on the mechanical, physical, and barrier characteristics, as well as antimicrobial properties of the prepared active films for potential further food applications were investigated.

[1] Lethal dose.

[2] generally recognized as safe.

2. Materials and methods

2.1. Active films preparation method

PP granules (Moplen Z30S: MFI = 25 Dg/min, a density of 0.9 g/cm^3, suitable for food contact, Marun Petrochemical co., Bandar-e Emam Khomeyni, Iran) and SA powder (AppliChem, USA) in an amounts of 2, 4 and 6% (w/w) were mixed well in a stainless steel container. The mixture was introduced to a co-rotating twin screw extruder (SM PLATEK, South Korea) with a screw diameter of 20 mm and L/D = 32. The heating zones of the twin-screw extruder at heating profile were 145–190 °C. Feeder rotational speed was set to 22.4 rpm and extruder rotation was 145 rpm. The extruder pressure was set to 60 bars. The mixture of the materials melted and mixed together inside the extruder as a result of the shear and pressure forces. The molten materials were left out of the extrusion in string form. They passed through cold water basin and were then cut into the granules. The granules were collected and added into another twin-screw extruder (Castiny Ghioldys, Italy) with a temperature profile of 190–230 °C to produce packaging films with a width and thickness of 30 cm and 50 ± 5 μm, respectively (Fasihnia et al., 2017).

2.2. Determination of film properties

2.2.1. Thickness measurement

The thickness of films was measured by a micrometer (Mitutoyo, Japan) with an accuracy of 0.01 mm. It was measured at three different points and averages were used.

2.2.2. Mechanical properties

To evaluate the mechanical properties of the films, the tensile strength at break (T) and elongation at break point (E) criteria were used. Film T and E were determined by ASTM D882-12 (2012) using a Tensile Machine (Adamel Lhomargy, Italy) with a load cell capacity of 100 N. The grips distance was 50 mm and the test speed was 500 mm/min. The T and E measurements for each sample were taken in triplicates (Durmus, Woo, Kasgoz, Macosko, & Tsapatsis, 2007; Fasihnia et al., 2017; Han & Floros, 1997).

2.2.3. Fourier-transform infrared (FT-IR) spectra analysis

The possible interactions of functional groups between antimicrobial additives with PP film were studied by Fourier-transform infrared (FT-IR) spectrometry. Films adhered to a sample holder in the shape of 3 × 3 cm squares. The FT-IR spectra were obtained with an FT-IR spectrophotometer by scanning from 4000 to 400 cm^{-1} (Bruker, Tensor 27, Germany) at room temperature (25 °C) (Jongjareonrak, Benjakul, Visessanguan, & Tanaka, 2008; Weng & Chen, 1997).

2.2.4. Water vapor permeability (WVP)

For WVP tests cups with an average interior diameter of 2 cm, an outlet diameter of 1.5 cm and a depth of 4.5 cm were used according to ASTM methodoligy E96-95 (1995) (Vartiainen, Skytta, Enqvist, & Ahvenainen, 2003; Almasi, Ghanbarzadeh, & Entezami, 2010; Jipa et al., 2012). Samples were cut into discs with a diameter of 1.6 cm. All