Food Control 73 (2017) 663–671

Contents lists available at ScienceDirect

Food Control

journal homepage: www.elsevier.com/locate/foodcont

Antimicrobial efficacy of clove essential oil infused into chemically modified LLDPE film for chicken meat packaging

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ARTICLE INFO

Article history:
Received 27 June 2016
Received in revised form
8 September 2016
Accepted 9 September 2016
Available online 10 September 2016

Keywords:
Antimicrobial packaging
Clove essential oil
Melting temperature
Surface modification
L. monocytogenes
Salmonella Typhimurium

ABSTRACT

Linear low-density polyethylene (LLDPE) is one of the most commercially successful packaging materials; however, to be used for active packaging purpose functionality must be crafted. In this work, LLDPE surface was chemically modified by chromic acid (CA) treatment and coated with clove essential oil (CLO); afterwards, the films were tested for antimicrobial properties. The optical, mechanical, barrier, thermal, and morphological properties of the films were characterized. FTIR spectroscopy confirmed the infusion of CLO in the film, and coating of CLO influenced the yellow color value of the films. The tensile stress and elongation at break of acid-treated and CLO coated films differed significantly from the control sample. CLO coating improved the UV-barrier property of LLDPE film. The melting point, T_m of the composite films dropped significantly from 122 to 117 °C when CLO was coated on the etched film surfaces. The morphology of the LLDPE/CA films exhibited pitting and roughness of the surface. LLDPE/CA/CLO films exhibited strong antimicrobial activity against *Salmonella* Typhimurium and *Listeria monocytogenes* in a packed chicken sample for 21 days of refrigerated storage. Thus, the developed film could be used as active packaging for fresh chicken.

1. Introduction

Consumers of today are demanding for minimally processed food products without any chemical preservative. Thus, the food industry has focused on the development of active or intelligent packaging where active compound/s directly or indirectly interact with the packaged food products by avoiding the production of undesired compounds and restricting the growth of pathogens (Jideani & Vogt, 2016; Pereira de Abreu, Cruz, & Paseiro Losada, 2012). Many approaches of active food packaging have been employed including moisture absorbents, oxygen scavengers, UV-barriers, nanoparticles, nanosensors, and incorporation of synthetic and natural antimicrobial agents, among others (Arfat, Benjakul, Vongkamjan, Sumpavapol, & Yarnpakdee, 2015; Pereira de Abreu et al., 2012; Ramos, Valdés, Mellinas, & Garrigós, 2015). Antimicrobial active packaging can be produced by direct incorporation of an active compound into a polymer matrix, coating the compound onto the packaging surface, or immobilizing it in sachets (Otoni, Espitia, Avena-Bustillose & McHugh, 2016). Additionally, naturally derived antimicrobial agents (*e.g.*, essential oil and herb extract) have gained increasing acceptances among consumers as healthier preservatives, and their use in packaging is becoming an increasing alternative to chemical synthetic additives (Goñi, Gañán, Strumla & Martini, 2016).

Linear low-density polyethylene (LLDPE) is a commonly used thermoplastic with a wide range of packaging applications because of its abundant supply, low cost, superior processability, low energy requirement for processing, and high specific modulus and strength. However, LLDPE is an inert hydrophobic polymer, and it needs to be modified to render antimicrobial packaging functionality. Various techniques have been employed for the suitability of LLDPE for antimicrobial packaging purpose. Incorporation of potassium sorbate into low-density polyethylene (LDPE) or in a blended film like ethylene vinyl alcohol (EVA)/LLDPE through extrusion/heat press has been used by many researchers for lowering the growth rate of yeasts and molds (Devlieghere, Vermeiren, Jacobs, & Debevere, 2000; Han & Floros, 1997; Kuplennik et al., 2015). Hauser and Wunderlich (2011) showed

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http://dx.doi.org/10.1016/j.foodcont.2016.09.018
0956-7135/

that films coated with a lacquer containing sorbic acid (2, 4-hexadienoic acid) inhibited *Escherichia coli*, *Listeria monocytogenes*, and *Saccharomyces cerevisiae*. Shemesh et al. (2015) used organo-modified montmorillonite (OMMT) clays, as active carriers for carvacrol in LLDPE composite films, and found that the blended film exhibited excellent and extended antimicrobial activity against *E. coli*. However, LDPE/OMMT/carvacrol films lost their antimicrobial activity within several days, maybe due to the fast release of carvacrol from the LDPE matrix. A number of researchers (Rojas et al., 2015; Torres, Romero, Macan, Guarda, & Galotto, 2014) have impregnated 2-nonanone and thymol into LLDPE using supercritical CO_2. These essential oils showed antimicrobial activity; however, their major limitation was their volatility and thermal degradation when incorporated during the mechanical processing of the polymeric films. A limited number of publications are available on the essential oil coating of LLDPE to exhibit antimicrobial property.

Since LLDPE is a non-polar polymer with low surface energy (Kuhn et al., 2000), it has poor bonding and coating properties. To improve these properties, different surface-treatment methods, like plasma, corona and flame discharge, laser, UV, laser, electron beam irradiation, and chemical treatment with strong acids have been reported (Bag, Kumar, & Maiti, 1999; Lee, Goddard, & Hotchkiss, 2009; Novak et al., 2013; Sanchis, Blanes, Blanes, Garcia, & Balart, 2006; Simonović et al., 2014). However, each technique has advantages and limitations with specific applications. In general, the objective of these techniques are to create a polar surface to yield negative charges that basically will improve the hydrophilicity and wettability of LLDPE, so that a functional compound can be adhered and/or attached to the surface.

Clove oil (*Syzygium aromaticum*, Lin) is a natural essential oil and flavouring substance with antimicrobial and antioxidant activities. It contains a variety of bioactive compounds such as sesquiterpenes and triterpenoids (Ramadan, Asker, & Tadros, 2013). Eugenol (4-allyl-2-methoxyphenol) is the main bioactive ingredient of CLO, which has strong insecticidal, antioxidant, and antifungal activity (Chami, Chami, Bennis, Bouchikhi, & Remmal, 2005; Gülçin, Şat, Beydemir, Elmastaş, & Küfrevioğlu, 2004; Park, Lee, Lee, Park, & Ahn, 2000). As a food additive, eugenol was classified by the U.S. Food and Drug Administration (FDA) to be a substance that is generally regarded as safe (GRAS) (Dai, McLandsborough, Weiss, & Peleg, 2010). CLO has been used as an antimicrobial agent in different types of films and coatings; however, to the best of the authors' knowledge no studies have been carried out on CLO coating on surface modified LLDPE film and its antimicrobial activity.

Therefore, the present paper aims to report the effect of CLO coating on the surface of LLDPE films created by acid treatment, followed by the evaluation of the coated film for its antimicrobial activity against *Listeria monocytogenes* and *Salmonella* Typhimurium. Additionally, the thermo-mechanical and structural properties of the developed films were studied leading to industrial applications.

2. Materials and methods

2.1. Materials

A commercial grade LLDPE (EFDC-7087; Melt Index 1; density 0.92; Melting Point 124 °C) was procured from Equate Petrochemical Co. (Kuwait). Potassium dichromate, sulfuric acid, and clove essential oil (CLO) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Chromic acid (CA) was freshly prepared by dissolving potassium dichromate in hot concentrated sulfuric acid (18.4 M) prior to use.

2.1.1. Bacterial media and strains

To evaluate the antimicrobial properties of developed films, two strains, namely, Culti-loops® of Gram-negative *Salmonella enterica sv* Typhimurium (ATCC 14028) was obtained from Remel Europe Limited (Dartford, Kent, UK) and Lyfo Disk pellets® of Gram-positive *Listeria monocytogenes* (ATCC 19114) strain was purchased from MediMark Europe (Grenoble, CEDEX, France). Tryptic soy broth (TSB) and Muller Hinton agar (MHA) were procured from TM Media (Bhiwadi, India). Brain heart infusion broth (BHIB) was obtained from Conda Laboratories (Torrejón de Ardoz, MD, Spain). Polymyxin-Acriflavin-Lithiumchloride-Ceftazidime-Aesculin-Mannitol (PALCAM) agar base, Xylose lysine deoxycholate agar (XLD) and PALCAM selective supplements were obtained from Oxoid (Basingstoke, HM, UK).

2.2. Extrusion of LLDPE films

LLDPE films were produced using a single screw extruder (RCP-0625 Microtruder; Randcastle Extrusion Systems, Inc., NJ, USA) with a screw diameter of 160 mm and a screw L/D ratio of 27/1 with 34 cc volume. The temperature profile used was 170–185 °C with a screw speed of 20 rpm and a chill temperature equal to 14 °C.

2.3. Surface modification

Surface modification of LLDPE film was carried out following the method described by Bag et al. (1999). Briefly, LLDPE film was cut into sizes of 9 × 5 cm, and the film samples were washed with acetone and distilled water, and dried. The dried films were etched by CA in a water bath maintained at 55–60 °C for a period of 30 min. After acid treatment, the films (LLDPE/CA) were immediately washed with deionized water and vacuum dried at 40 °C for 20 min. CLO (0.5 g) was spread on the surface of the dried films using a sterile spreader. Films (LLDPE/CA/CLO) were suspended for drying for a period of 4 h. For antimicrobial studies, the film was coated and identified only on one side. All measurements were carried out two days after acid treatment.

2.4. Weight and thickness measurement

The weight and thickness of the films were measured before and after the acid treatment and also after the coating with CLO by an analytical balance and a micrometer (Mitutoyo, Model MCD-1″PXF, Mituyoto Corp., Kawasaki-shi, Japan), respectively. The thickness of each film was measured in at least ten different locations at three central and seven edge locations with an accuracy of 0.001 mm. Precision of thickness measurement was ±5%.

2.5. Measurement of color, UV barrier, and transparency

The instrumental color parameters, namely, *L* (degree of lightness), *a* (redness/greenness), and *b* (yellowness/blueness) were measured using a Mini EZ Scan portable colorimeter (model 4500L HunterLab, Reston, VA, USA) consisting of a D65 illuminant and a 10° observatory angle. Film samples were positioned on a white standard plate ($L = 93.49$, $a = -0.95$, $b = 2.76$) and color values were recorded. Total color difference (ΔE) was calculated based on the LLDPE film following Eq. (1):

$$\Delta E = \left[\left(L_{test\ film} - L_{LLDPE} \right)^2 - \left(a_{test\ film} - a_{LLDPE} \right)^2 - \left(b_{test\ film} - b_{LLDPE} \right)^2 \right]^{1/2} \quad (1)$$