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Development of anti-insect food packaging film containing a polyvinyl alcohol and cinnamon oil emulsion at a pilot plant scale

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ABSTRACT

Anti-insect packaging films containing cinnamon oil (CO) encapsulated by polyvinyl alcohol (PVA) for repelling *Plodia interpunctella* (Hübner) larvae were manufactured using pilot plant scale instruments. The microcapsule emulsion of CO and PVA was printed onto polypropylene (PP) film as an ink mixture using the gravure printing method. The printed PP film was then laminated with a low-density polyethylene (LDPE) film to protect the printed side. Four types of multilayer films were produced: control film, 2% CO film, encapsulated 1% CO film, and encapsulated 2% CO film. When mechanical properties were assessed, tensile strength and elongation-at-break were not significantly different among films, whereas the Young's modulus values were different between the control film and the three types of CO-containing films. In a repellent test, 2% CO film repelled *P. interpunctella* larvae most effectively. For sensory evaluation, which was performed using milk chocolate, caramel soft candy, and cookies packaged with the produced films, the produced films did not affect the sensory characteristics. Therefore, the films printed with emulsions of CO and PVA could be applied in the food industry to help protect foods from infestation by *P. interpunctella*.

1. Introduction

In recent years, the use of plastic films instead of traditional packaging materials such as paper and cardboard has increased (Riudavets et al., 2007). Plastic film is advantageous, since it could protect food products against external damage caused by handling, contamination, and attacks by different insects (Paine and Paine, 1993). The most frequently used plastic films are polyethylene (PE) and polypropylene (PP).

In spite of modern food distribution and storage systems, most packaged food products, except for canned and frozen goods, are vulnerable to attacks by insects (Xingwei et al., 2004; Lee et al., 2011). The most devastating insect pests of stored products are (in order): Coleoptera (beetles) and Lepidoptera (moths), which are found in a range of climates (Robertson, 2006). *Plodia interpunctella* (Hübner) (Lepidoptera: Pyralidae), also known as Indian meal moth, is a major stored product pest that develops on grains, nuts, and processed foods (Johnson et al., 1992). *P. interpunctella* are called “penetrators” because of their well-developed, sharp mandibles that can make small holes and tear plastic films to penetrate into foods (Highland, 1984). The penetration into packaged foods can occur during the distribution phase, such as transportation and storage in warehouses, or in retail stores (Licciardello et al., 2013).

Many synthetic pesticides, such as methyl bromide and phosphine, have been used to control *P. interpunctella*. However, there are environmental concerns associated with the use of these synthetic materials. Essential oils (EO) derived from plants as secondary metabolites have been used for many purposes including antibacterial, antifungal, and anti-insect effects. Cinnamon

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(*Cinnamomum zeylanicum* Blume) leaf oil (CO) potently kills insect pests in stored products. The major compounds in CO include *trans*-cinnamaldehyde (58.1%), benzaldehyde (12.2%), and eugenol (5.1%) (Yang et al., 2005). Although EOs are environmental friendly materials with effective anti-insect activity, the high volatility of EOs make it difficult for them to be applied in the food packaging industry. To control the volatility and achieve an intended release of EOs, a range of microencapsulation technologies that surround tiny particles or droplets with outer coatings have been investigated. The core contains the active compound, while the wall protects the core permanently or temporarily from the external atmosphere (Alikhani and Garmakhany, 2012). In addition, controlled release of active compounds could be accomplished by microencapsulation, which has many potential applications for which the active agents are desired to be left in a certain container at a specific concentration (Dowding et al., 2005). Maji et al. (2007) produced microcapsules that repelled mosquitos using *Zanthoxylum limonella* oil as an insect repellent and gelatin as an encapsulant (wall material). Jo et al. (2013) developed an anti-insect PVA polymer strips embedding CO droplets that efficiently repelled *P. interpunctella* larvae. The application of microencapsulation technologies on EOs affords several advantages including protecting unstable core materials and preventing the core materials from poor environments. Gum arabic, whey protein, maltodextrin, gelatin, and polyvinyl alcohol (PVA) are commonly used as the wall materials (Kim et al., 2013). Among these, PVA is a hydrophilic semicrystalline synthetic polymer produced commercially by the hydrolysis of poly(vinyl acetate) (Jansson et al., 2006). Despite being a synthetic polymer, PVA is biodegradable and water soluble (Lenz, 1993; Ishigaki et al., 1999), and has excellent chemical resistance and physical properties. Because of these properties, PVA has been frequently used as an encapsulant (wall material) in microencapsulation technology.

In this study, multilayer anti-insect films were developed based on PP and low-density polyethylene (LDPE) films laminated with a mixture of ink and CO or PVA–CO emulsions using pilot plant scale machines. The mechanical properties, repellent effects against *P. interpunctella*, and sensory effects of the produced films were evaluated to demonstrate the potential of the films as practical food packaging for industrial applications.

2. Materials and methods

2.1. Manufacturing of pilot plant scale film

2.1.1. Preparation of PVA-CO microcapsule emulsion

The microcapsule emulsion of PVA–CO was prepared according to the method described by Kim et al. (2013). PVA was purchased from Sigma–Aldrich Co., Ltd. (St. Louis, MO, USA), and CO was obtained from Scentpia Co., Ltd. (Bucheon, Korea). Two grams of PVA and 98 g of distilled water were mixed for 2 h. Next, 5.5 g of CO was added to the PVA solution, which was then homogenized for 5 min using a homogenizer (IKA T25-Digital Ultra Turrax; Staufen, Germany). Finally, 0.07 g Tween 80 (Ilshinwells Co., Ltd.; Seoul, Korea) was added to the mixture as an emulsifier to form the stable microcapsule emulsion of PVA–CO.

2.1.2. Manufacturing of pilot plant scale anti-insect film

The pilot plant scale anti-insect film was produced by printing a mixture of PVA–CO microcapsule emulsion and white ink on PP film (0.06 mm thickness, Au Co., Ltd.; Bucheon, Korea), and then laminating the printed PP film with LDPE film (0.01 mm thickness, Au Co., Ltd.; Bucheon, Korea). The ink (Daihan Ink Co., Ltd.; Anyang, Korea) consisted of methyl ethyl ketone (30–50%), toluene (11–21%), ethyl acetate (10–15%), urethane resin (aliphatic) (8–12%), white organic pigment (8–12%), propylene glycol methyl ether acetate (6–11%), isopropyl alcohol (3–5%), vinyl resin (3–5%), and polyethylene wax (<1%). The formulation of the emulsion-ink mixture and the types of films used is shown in Table 1. During the printing process, the mixture of ink and PVA–CO emulsion was printed onto PP film using a gravure printing press (Roto Gravure Printing Press; Ilsung Machinery Co., Ltd.; Gumi, Korea) at a speed of 20–200 m/min at room temperature. The printed PP film was laminated with LDPE film using a laminator (Dry laminating & Extrusion laminating machine, INT CO., Ltd; Ansan, Korea) at a speed of 120–160 m/min. The finished films were aged at 62–63 °C for 72 h before use. The film thickness was measured using a digital micrometer (ID-C112X. Mitutoyo Co., Kawasaki, Japan) accurate to within 0.1 μm, with the film thickness used in calculating the mechanical properties of the films.

2.2. Mechanical properties of the pilot plant scale anti-insect films

Tensile strength, percentage elongation-at-break, and Young's modulus of the films were measured according to the American Society of Testing and Materials (ASTM) standard method D 882-01. A texture analyzer (TAXTplus 50; Stable Micro Systems Ltd.; Vienna, UK) was used for the tests. Before measuring, the films were cut into 50 × 8-mm strips and incubated at 25 °C with 50% RH for 48 h. A prepared strip of film was placed between two grips at an initial distance of 50 mm apart, and cross-head speed was 30 mm/min. After the strip broke, a force–distance curve was obtained, and the TS and %E were calculated as follows:

$$\text{Tensile strength (MPa)} = \frac{F}{A}$$

where F is the peak force at failure (N), and A is the cross-sectional area of the film (mm^2).

$$\text{elongation} - \text{at} - \text{break (\%)} = \frac{l_b - l_0}{l_0} \times 100$$

where l_b is the measured elongation-at-break (mm), and l_0 is the original length of the film placed between the grip heads that were 50 mm apart.

$$\text{Young's modulus (MPa)} = \frac{\text{Stress}}{\text{Strain}} = \frac{F/A}{(l_b - l_0)/l_0} = \frac{F \cdot l_0}{A \cdot (l_b - l_0)}$$

At least 12 replicates were analyzed for each film type.

2.3. Repellency test

P. interpunctella was obtained from the Laboratory of Population Ecology at Korea University (Seoul, Korea), which maintained the *P. interpunctella* for 5 years on dried vegetable commodities at 28 °C with 70–80% RH. The ability of the developed anti-insect films to repel *P. interpunctella* was assessed according to the method

Table 1
Formulations of emulsion and ink mixtures used for coating layer of anti-insect food packaging films containing cinnamon oil.

Type of film	Formulation
Control (0% CO)	10 kg ink
CO 2%	9.8 kg ink + 0.2 kg CO
Encapsulated CO 1%	7.9 kg ink + 2.1 kg emulsion of encapsulated CO with PVA
Encapsulated CO 2%	5.8 kg ink + 4.2 kg emulsion of encapsulated CO with PVA

Control, film consisting of PP, ink, and LDPE; 2% CO, film consisting of PP/ink with 2% CO/LDPE; Encapsulated 1% CO, film consisting of PP/ink with encapsulated CO with 1% PVA/LDPE; Encapsulated 2% CO film consisting of PP/ink with encapsulated CO with 2% PVA/LDPE.