Insect-Resistant Food Packaging Film Development Using Cinnamon Oil and Microencapsulation Technologies

In-Hah Kim*, Jaejoon Han*, Ja Hyun Na, Pahn-Sik Chang, Myung Sub Chung, Ki Hwan Park, and Sea C. Min

Abstract: Insect-resistant films containing a microencapsulated insect-repelling agent were developed to protect food products from the Indian meal moth (*Plodia interpunctella*). Cinnamon oil (CO), an insect repelling agent, was encapsulated with gum arabic, whey protein isolate (WPI)/maltodextrin (MD), or poly(vinyl alcohol) (PVA). A low-density polyethylene (LDPE) film was coated with an ink or a polypropylene (PP) solution that incorporated the microcapsules. The encapsulation efficiency values obtained with gum arabic, WPI/MD, and PVA were 90.4%, 94.6%, and 80.7%, respectively. The films containing a microcapsule emulsion of PVA and CO or incorporating a microcapsule powder of WPI/MD and CO were the most effective ($P < 0.05$) at repelling moth larvae. The release rate of cinnamaldehyde, an active repellent of cinnamaldehyde, in the PP was 23 times lower when cinnamaldehyde was microencapsulated. Coating with the microcapsules did not alter the tensile properties of the films. The invasion of larvae into cookies was prevented by the insect-repellent films, demonstrating potential for the films in insect-resistant packaging for food products.

Keywords: cinnamaldehyde, cinnamon oil, Indian meal moth, insect-resistant packaging, microencapsulation

Practical Application: The insect-repelling effect of cinnamon oil incorporated into LDPE films was more effective with microencapsulation. The system developed in this research with LDPE film may also be extended to other food-packaging films where the same coating platform can be used. This platform is interchangeable and easy to use for the delivery of insect-repelling agents. The films can protect a wide variety of food products from invasion by the Indian meal moth.

Introduction

Insects can enter packaged products during transportation and storage in warehouses or in retail stores. The U.S. Dept. of Agriculture reported that the 3rd most common complaint from food consumers related to the presence of foreign materials in food products was having insects/insect parts (12%) in the products, exceeded only by complaints about bones (21%) and machine parts (19%) (USDA-FNS 2005).

The Indian meal moth, *Plodia interpunctella* (Hübner), is a serious pest to grain, seeds, meals, beans, flour, dried fruits, nuts, and chocolate and can infest food warehouses, food processing facilities, and retail stores (Jenson and others 2010). The larvae of the Indian meal moth must be repelled because they can penetrate through packaging (Browditch 1997).

There has been growing interest in research on the use of plant extracts as alternatives to synthetic insecticides. For example, the insect-repelling properties of cinnamon have been reported. The essential oil of cinnamon exhibited contact toxicity to *Tribolium castaneum*, *Sitophilus zeamais*, and pulse beetle (*Callasobruchus maculatus* L.) (Huang and Ho 1998; Ratnasekera and Rajapakse 2009).

Food packaging is the last line of defense against insect infestation of the finished product. Insect-resistant or -repellent packaging has been investigated as a plausible measure for controlling insect invasion (Mohan and others 2007). Coating and microencapsulation technologies have been employed in developing insect-repellent food packaging materials. Mohan and others (2007) developed an insect-resistant sheet that repelled *Sitophilus oryzae* (L.) and *Rhyzopertha dominica* (F.) by coating a protein-enriched pea flour solution on polyethylene. Maji and others (2007) produced microcapsules that repelled mosquitos using *Zanthoxylum limonella* oil as an insect repellent and gelatin as an encapsulant (wall material).

Gum arabic, whey protein isolate (WPI)/maltodextrin (MD), and poly(vinyl alcohol) (PVA) are common encapsulants for microencapsulation and have been successfully applied to protect and control the release of core materials (Krishnan and others 2005; Toure and others 2007; Sullad and others 2010). Gum arabic is particularly versatile for many encapsulation methods because it is a good emulsifier and has adequate solubility and sufficient viscosity to retain volatile compounds, including essential oils (Kaasgaard and Keller 2010). MD is another commonly used encapsulant, generally added in combination with proteins, gums, or starches

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Table 1–Procedures for preparation of microcapsule emulsions.

Wall material	Procedure	Reference
Gum arabic	- Mix 30 g of gum arabic with distilled water for 12 h at 12 °C, making 100 mL in total - Add 5.5 g of cinnamon oil (CO) to the gum arabic solution. - Homogenize at 3000 rpm for 5 min - Add 0.07 g of Tween® 80 to the homogenized solution	Krishnan and others (2005)
Whey protein isolate (WPI)/maltodextrin (MD)	- Mix 10 g of WPI and 20 g of MD with 100 g of distilled water for 30 min - Add 7.5 g of CO to the solution of WPI and MD - Heat the solution at 40 °C for 15 min - Homogenize at 13000 rpm for 10 min	Toure and others (2007)
Poly(vinyl alcohol) (PVA)	- Mix 2 g of PVA with 98 g of distilled water for 2 h - Add 5.5 g of CO in the PVA solution - Homogenize at 3000 rpm for 5 min - Add 0.07 g of Tween® 80 to the homogenized solution	Sullad and others (2010)

because of its poor emulsifying properties (Kaasgaard and Keller 2010). PVA is a water-soluble encapsulant that has been used extensively in controlled-release applications. Microcapsules have been made of PVA containing santosol and linseed oils (Bachtsi and Kiparissides 1996; Suryanarayana and others 2008). However, little information is available on using microencapsulation technologies to develop insect-repellent food-packaging films incorporating plant essential oils. Thus, the objectives of this study were to (1) develop food-packaging films incorporating encapsulated cinnamon oil; (2) study the effectiveness of the films at repelling moth larvae; (3) determine the release rates of an active repellent (cinnamaldehyde) from the developed films; (4) determine tensile properties of the developed films; and (5) examine the moth larva-resistant ability of the films to simulate what would happen if the films were used to package a cookie, an insect attractant.

Materials and Methods

Materials

Cinnamon oil (CO) (100%) was purchased from Scentpia Co., Ltd. (Bucheon, Korea). The encapsulant of gum arabic was purchased from Samchun Pure Chemicals Co., Ltd (Pyeongtaek, Korea) and those of WPI and MD were supplied by Samyang Genex Corp. (Seoul, Korea). The encapsulant of PVA (degree of hydrolysis: 88%) was purchased from Sigma-Aldrich Co. Ltd. (St. Louis, Mo., U.S.A.). Tween 80, used as an emulsifier, was purchased from Ilshinwells Co., Ltd (Seoul, Korea). Low-density polyethylene film (LDPE) (40 μm) was supplied by Aumart (Bucheon, Korea). Ink and a polypropylene (PP) solution were purchased from Daihanink Co., Ltd (Anyang, Korea). Ink contained methyl ethyl ketone (30% to 50%), toluene (11% to 21%), ethyl acetate (10% to 15%), urethane resin (aliphatic) (8% to 12%), yellow organic pigment (8% to 12%), propylene glycol methyl ether acetate (6% to 11%), isopropyl alcohol (3% to 5%), vinyl resin (3% to 5%), and polyethylene wax (<1%). The PP solution contained methyl ethyl ketone (55% to 60%), acrylic resin (25% to 30%), toluene (10% to 15%), silica (5% to 10%), dipropylene glycol monomethyl ether (4% to 10%), and polyethylene wax (1% to 3%). The stock culture of Indian meal moth, originated from the moth infested vegetable commodities in a food warehouse (Daegu, Korea), has been maintained for 5 y on dried vegetable commodities at 28 °C and 70% to 80% relative humidity (RH) in the Laboratory of Population Ecology at Korea Univ. (Seoul, Korea). The feed for larvae was composed of bran (53% (w/w)), dried yeast (13%), glycerol (33%), methyl *p*-hydroxybenzoate (0.13%), and sorbic acid (0.13%).

Determination of emulsion formation and encapsulation efficiency

The microencapsulated oil emulsions were prepared using gum arabic, WPI/MD, and PVA as encapsulants. The methods used to prepare each emulsion are summarized in Table 1. Encapsulation efficiency was determined following the method of Choi and others (2010b). Gum arabic (30 g), WPI (10 g) / MD (20 g), or PVA (2 g) was hydrated with distilled water (100 mL). An oil mixture prepared with CO, fully hydrogenated oil (Lottesamkang Co., Ltd., Seoul, Korea), and Oil Red O (1-([4-(xylylazo)xylyl]azo)-2-naphthol) (Sigma-Aldrich Co., Ltd.) at a ratio (weight basis) of

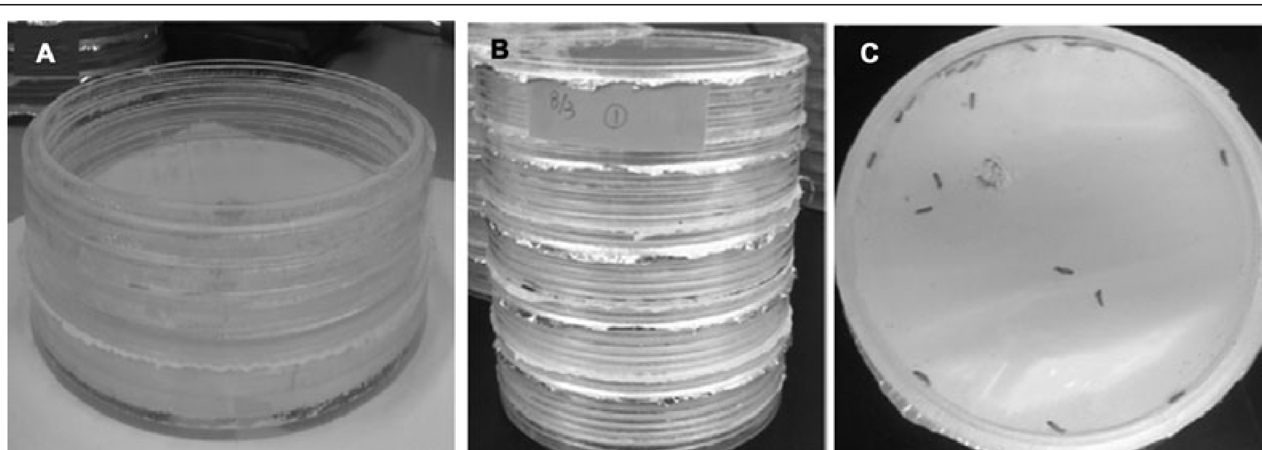

Figure 1–Photographs of the ring-shaped test apparatus (acrylic, 16-cm diameter, 1-cm thickness) used for the insect repellency test (A), a set of ring test apparatuses (B), and unrepelled pupae remaining on a film (C).

1.00:0.40:0.01 was heated in a water bath at 80 °C. The hydrated gum arabic, WPI/MD, or PVA (1.12, 1.24, or 1.07 g, respectively) was mixed with the oil mixture. The ratio of each encapsulant to CO in the mixture was identical to that used for preparing the microemulsions listed in Table 1. Each encapsulant mixture was then homogenized (T 25 digital Ultra-Turrax®, Janke & Kunkel GmbH & Co., IKA® Labortechnik, Staufen, Germany) under the conditions shown in Table 1. The homogenate was sprayed into 200 mL of distilled water using a spray gun (WAGNER W180P, Markdorf, Germany). The homogenate-sprayed solution (10 mL) was mixed with 20 mL of *n*-hexane (Samchun Pure Chemical Co., Ltd., Pyeongtaek, Korea) at 300 rpm for 10 min using an orbital shaker (JS Research, Inc., Gongju, Korea). Absorbance of the mixture was measured at 510 nm and the encapsulation efficiency was determined using the following equation (Choi and others 2010b):

$$\text{Encapsulation efficiency (\%)} = \frac{1 - \textit{absorbance}}{1} \times 100$$

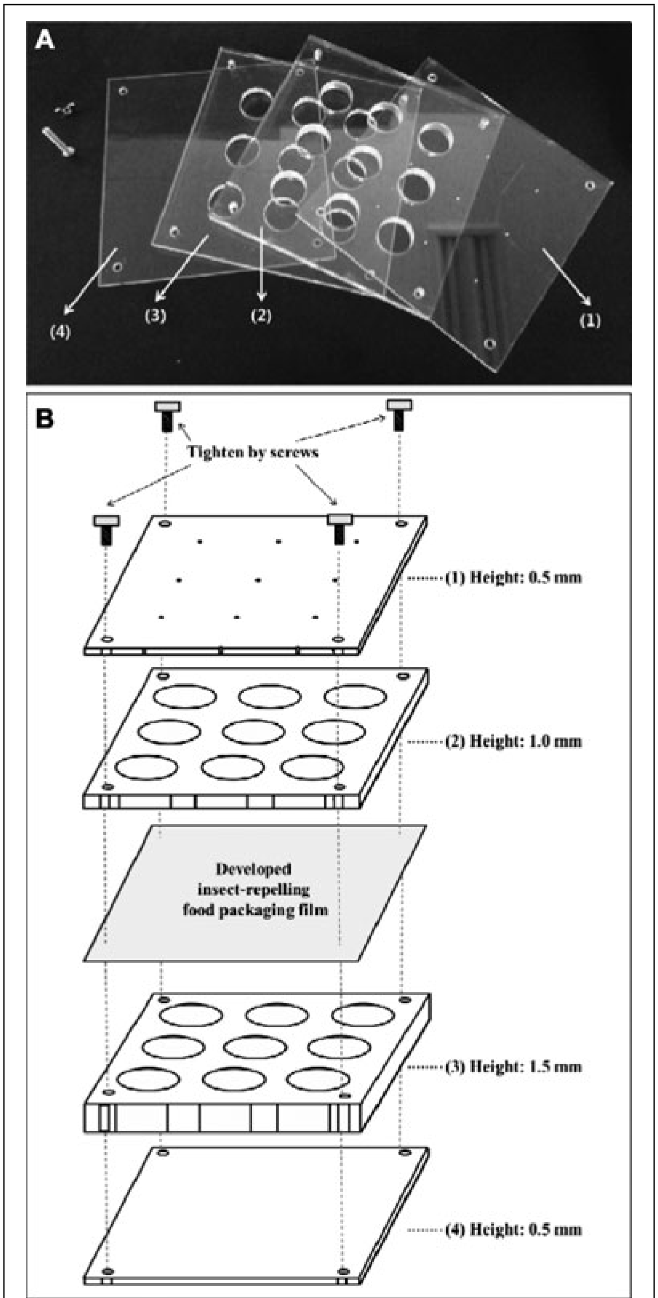

Figure 2–Photograph (A) and exploded diagram (B) of the apparatus used for the penetration test (25 cm × 25 cm) with *P. interpunctella* larvae. Labels (1), (2), (3), and (4) denote the plates. Nine holes (3-cm diameter) were drilled into plates (2) and (3).

Preparation of microcapsule powders

Microcapsule powders of gum arabic, WPI/MD, and PVA were prepared by spray drying (Eyela SD-1000, Tokyo Rikakikai Co., Tokyo, Japan). The inlet and outlet air temperatures used for drying the gum arabic and PVA emulsions were 178 ± 5 °C and 85 ± 5 °C (Krishnan and others 2005), respectively, and those for WPI/MD were 120 ± 3 °C and 60 ± 3 °C, respectively (Toure and others 2007).

Formation of LDPE films coated with inks incorporating microcapsule emulsions

The microcapsule emulsions prepared with gum arabic, WPI/MD, and PVA were mixed with the ink by vortexing for 1 min, 1 min, and 15 min, respectively. The concentration of the oil in the mixture was adjusted to 2% (w/w), accounting for the encapsulation efficiency of each microcapsule emulsion. That is, 2.4 g of the gum arabic emulsion, 2.2 g of the WPI/MD emulsion, or 2.5 g of the PVA emulsion was mixed with the ink, resulting in a total weight of 6 g for the mixture. The LDPE film was cut into a 25 × 25 cm sheet and coated with the ink incorporating each microcapsule emulsion by using a 20.6-μm-deep bar coater (COAD. 401 No. 9, Ocean Science, Uiwang, Korea). The thickness of the ink-coated LDPE film was 60 μm after drying.

Formation of LDPE films coated with PP incorporating microcapsule powders

The microcapsule powders prepared with gum arabic, WPI/MD, and PVA were mixed with the PP solution by vortexing for 1 min to adjust the concentration of CO to 2% (w/w). For example, 0.7 g of the gum arabic microcapsule powder, 0.6 g of the WPI/MD powder, or 0.8 g of the PVA powder was mixed with the PP solution, resulting in a total weight of 6 g for the mixture. The mixture was then spread on the surface of the LDPE film (25 × 25 cm) using the bar coater to form 60-μm-thick LDPE films coated with PP incorporating microcapsule powders.

Microscopic imaging

Microcapsule emulsions were investigated using a microscope (Versus, Medline, Oxfordshire, UK) equipped with a telecamera (Telecamera DV1300, Bel Engineering, Monza, Italy). Equal volumes of the emulsions were placed onto microscope slides, coverslipped, and imaged at 200× magnification. The morphologies of the microcapsule powders, the LDPE films coated with PP incorporating microcapsule powders, and the holes made on the films by larvae during the penetration test were examined by a field emission-scanning electron microscope (FE-SEM, S-4700, Hitachi, Chiyoda-ku, Tokyo, Japan). The powders and the films were attached to SEM stubs using double-sided carbon tape coated with platinum.

Repellency test

The insect repellency of the developed films was studied using 30 third-instar Indian meal moth larvae. The larvae were placed on the coated side of the film, which was mounted on a

16-cm-diameter, 1-cm-thick acrylic ring (Figure 1A). The open side of the ring-shaped container was then capped with 5 layers of aluminum foil. The capped containers were stacked (Figure 1B) and incubated at 30 °C for 7 d. The numbers of pupae from the larvae found on the surface of the film, the wall of the container, and the aluminum foil surface were counted to quantify the insect-repelling effect of the films (Figure 1C). Fewer pupae on the film indicated better insect repellency.

Release profile determination

Cinnamaldehyde is an active repellent of CO (Na and others 2008). The values for diffusion coefficient (D) of cinnamaldehyde for the diffusion in the ink incorporating the microcapsule emulsion of PVA and CO and the diffusion in the PP incorporating the microcapsule powder of WPI/MD and CO were determined. The values of D were also determined with LDPE films coated with ink or PP incorporating CO without microencapsulation (controls). The diffusion through the film was determined from the data obtained using a relationship derived from the solution to Fick's law for a plane sheet (Crank 1975). In the case of a film with initial uniform concentration equal to C_0 and both surfaces at a constant concentration equal to C_1, the following solution for diffusion in a plane sheet can be used as shown in Crank (1975)

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left\{-D(2n+1)^2\pi^2 t/4l^2\right\}$$

where M_t is the amount of cinnamaldehyde absorbed by the ink or PP at time t; and M_∞ (M_{inf}) is the amount of cinnamaldehyde absorbed by the ink or PP at equilibrium; l = thickness of film.

The release profile of cinnamaldehyde from the film was obtained with the determined value of D. Fitting the mathematical solution to the diffusion data was done using a nonlinear function from Matlab (Version 7.1.0.246 (R14), The Mathworks Inc., Natick, Mass., U.S.A.).

The cinnamaldehyde in the film was quantified following the method of Sheung and others (2004) with modifications. A circular piece (1.8-cm diameter) from the film was placed in a 25-mL-gas tight bottle. The bottle was sealed with Teflon coated rubber septa and aluminum caps (Supelco, Inc., Bellefonte, Pa., U.S.A.) and incubated for 1 h at 70 °C for cinnamaldehyde desorption from the pieces of the film. The desorbed cinnamaldehyde in the headspace of the bottle was isolated by a SPME fiber with 50/30 μm divinylbenzene/carboxen/polydimethylsiloxane (DVB/CAR/PDMS) coating (Supleco), which was manually inserted into the headspace of the bottle and kept for 20 min. The SPME fiber was retracted from the bottle and injected into a gas chromatography (GC) injection port at 220 °C and kept for 2 min for the desorption of volatile compounds. The desorbed volatile compounds were separated by a GC (GC-2010AF, Shimadzu Corp., Kyoto, Japan) equipped with a DB-5 capillary column (30 m × 0.25 mm) and a flame ionization detector. Detector temperatures was 290 °C. Column temperature rose from 55 to 65 °C at a rate of 1 °C/min, was held for 3 min, rose again from 65 to 290 °C at a rate of 10 °C/min rate, and was held at this final temperature for 10 min. Nitrogen was used as the carrier gas flowing at 1 mL/min.

Tensile properties

The American Society of Testing and Materials (ASTM) standard method D 882-01 (1997) was used to measure tensile strength

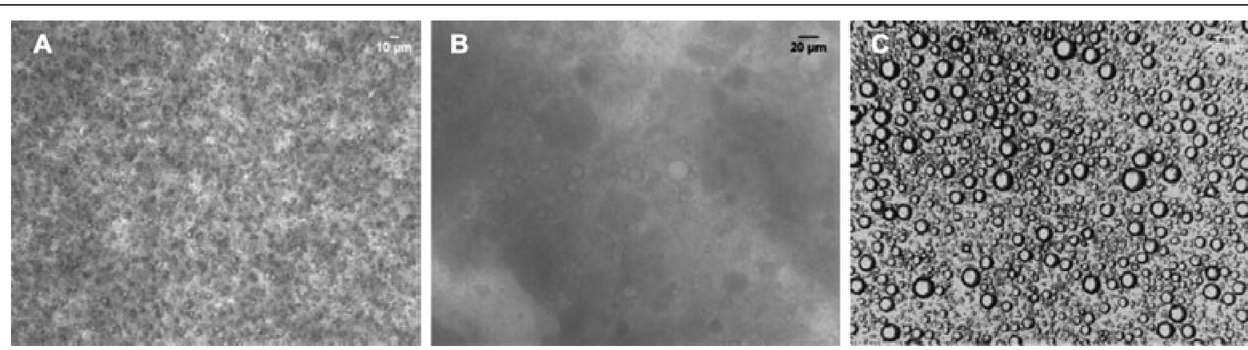

Figure 3–Micrographs of microcapsule emulsions made of gum arabic and cinnamon oil (CO) (A), whey protein isolate (WPI)/maltodextrin (MD) and CO (B), and poly(vinyl alcohol) (PVA) and CO (C).

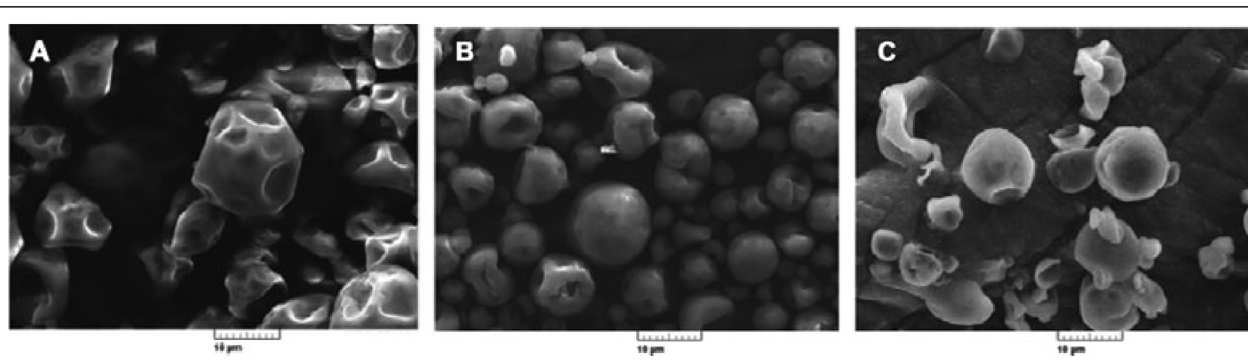

Figure 4–Micrographs of microcapsule powders made of gum arabic and cinnamon oil (CO) (A), whey protein isolate (WPI)/maltodextrin (MD) and CO (B), and poly(vinyl alcohol) (PVA) and CO (C).

(TS), percentage elongation at break (%E), and elastic modulus (EM) of films. Film samples were analyzed at 50 ± 2% RH, using a tensile property tester (TA-XT2, Stable Micro System Co. Ltd., Surrey, England) operated with a 5-kg load cell and a crosshead speed of 30 mm/min.

Penetration test

A penetration test investigating the ability of films to prevent invasion of insects through the packaging film was conducted following the method of Chung and others (2011). The test used 2 square acrylic plates (25 cm × 25 cm). Each plate had 9 wells, each 3 cm in diameter and 1 cm deep (Figure 2A). Cookie (1.5 g) (Pepperidge Farm, Inc., Norwalk, Conn., U.S.A.), a model food attractant, was placed in each well of one plate, and a third-instar larva was laid in each well of the other plate. The 2 plates were screwed together after placing a testing film between them (Figure 2B). The coated side of the film faced the larvae. The penetration test system was then incubated at 30 °C for 5 d. The number of wells that had been penetrated and the elapsed time to penetration were recorded for each film. Five types of films were tested: uncoated intact LDPE films, LDPE films coated with ink without microcapsules, LDPE films coated with ink incorporating the microcapsule emulsion of PVA and CO, LDPE films coated with PP without microcapsules, and LDPE films coated with PP incorporating the microcapsule powder of WPI/MD and CO.

Statistical analysis

The repelling test was replicated 15 times. Determination of tensile properties of the films was done in triplicate. Six repeated measurements in each replication were made to determine tensile properties. The penetration test was replicated 27 times for each kind of films. Analysis of variance was used to evaluate differences between means and if significant differences were observed, Duncan's multiple range tests were used to evaluate the means using PASW Statistics 18 (IBM Co., Ver. 18.0.0, N.Y., U.S.A.) to estimate the significant difference ($\alpha = 0.05$).

Results and Discussion

Encapsulation efficiency

The encapsulation efficiencies to form the microcapsule emulsions of gum arabic, WPI/MD, and PVA with CO were 90.4 ± 0.6, 94.6 ± 0.3, and 80.7 ± 1.9%, respectively. The encapsulation efficiency using PVA was relatively low, which may be due to a low ratio between the encapsulant PVA and CO (0.4:1). The

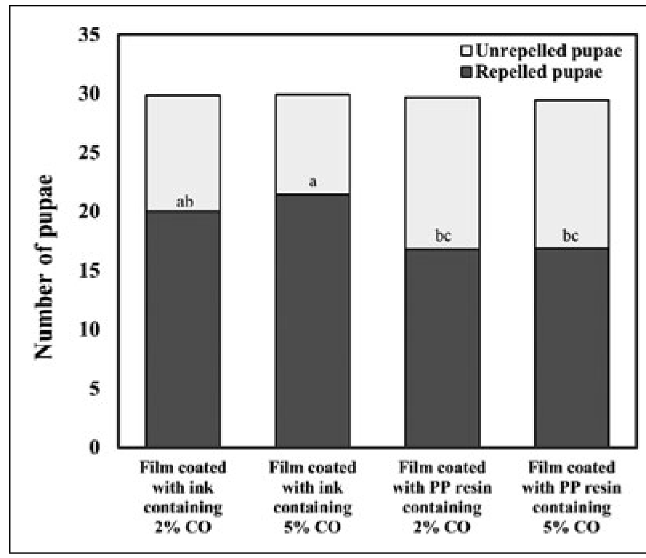

Figure 6–Comparison of insect-repelling effects of low-density polyethylene (LDPE) films coated with inks and PP incorporating cinnamon oil (CO) at 2% and 5% (w/w). Microencapsulation was not used in these experiments. The darker bar indicates the number of pupae found on the wall of the test container or on the aluminum foil surface; the difference between the value of a darker bar and 30 is the number found on the surface of each film. Means labeled with different letters are significantly different in the numbers of larvae found on the test film ($P < 0.05$).

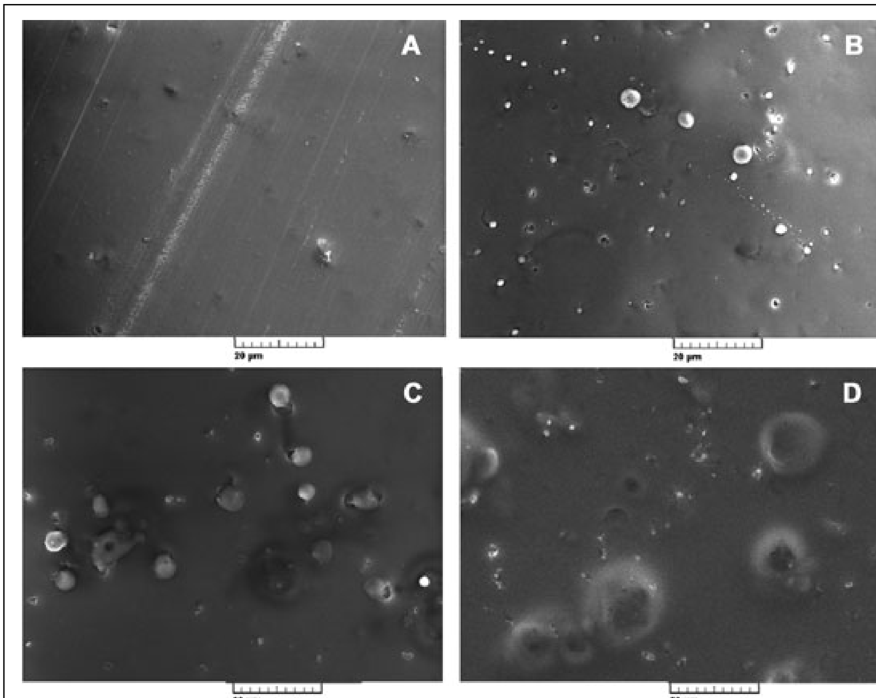

Figure 5–Micrographs of the polypropylene (PP) solution (control) (A), the PP solution incorporating microcapsule powders made of gum arabic and cinnamon oil (CO) (B), whey protein isolate (WPI)/maltodextrin (MD) and CO (C), and poly(vinyl alcohol) (PVA) and CO (D).

ratios for the gum arabic and WPI/MD encapsulants with CO were 5.5:1 and 4.0:1, respectively. At a lower ratio, the amount of wall material may be insufficient to fully cover the oil droplets, perhaps lowering the encapsulation efficiency (Minemoto and others 2002). However, a higher ratio or higher concentration of PVA could not be used because of its low solubility in water. At a higher concentration than 2% in water, undissolved PVA was precipitated.

The values for gum arabic and WPI/MD were higher than those reported for other encapsulants, including those for CO microcapsules made of soy protein isolate and MD (60%) and of modified starch and MD (33%) (Calvo and others 2010; Jun-xia and others 2011)

Microscopic imaging of microcapsule emulsions and powders and the PP incorporating microcapsule powders

Gum arabic, WPI/MD, and PVA formed stable microcapsule emulsions (Figure 3). The gum arabic emulsion droplet size was the smallest, and the WPI/MD one was the largest among the emulsions studied (Figure 3). Microcapsule powders were able to be prepared from those 3 emulsions and their morphologies are shown in Figure 4. The powder particles were generally bigger than 10 μm in diameter. Those made of WPI/MD and PVA were spherical, and those made of gum arabic were hollow (Figure 4). The hollow shape of cardamom oleoresin encapsulated with gum arabic was reported previously (Krishnan and others 2005). The formation of hollow particles is typical of the spray-drying process (Ré 1998) and is explained by the inclusion of a vapor bubble inside the emulsion. The formation of hallow particles is known to be affected by many factors, including the types and concentrations of the encapsulants and core materials (Ting and others 1992; Choi and others 2010a). Images of the PP incorporating microcapsule powder show that the powders were uniformly distributed throughout the PP matrix (Figure 5).

Repellency test

The results from the repellency test with LDPE films coated with either ink or PP incorporating oils without microencapsulation are given in Figure 6. The difference in the concentrations of the oils (2 and 5%) did not result in different repellencies ($P > 0.05$). Thus, a 2% concentration was chosen to study repellent release, film tensile properties, and insect penetration. The type of coating (ink compared with PP coating) also did not affect the insect repellency.

The insect-repelling effects of LDPE films coated with the inks incorporating different CO emulsions were compared with each other, with those of the uncoated LDPE film (control), and with the LDPE films coated with ink incorporating CO without microencapsulation (Figure 7). The number of pupae found on the wall of the test container and on the aluminum foil cap was significantly higher than the number of pupae found on the film in all cases except the control, indicating that all the coated films repelled the larvae. The repelling effects of the different films were not significantly different ($P > 0.05$). Incorporation of the

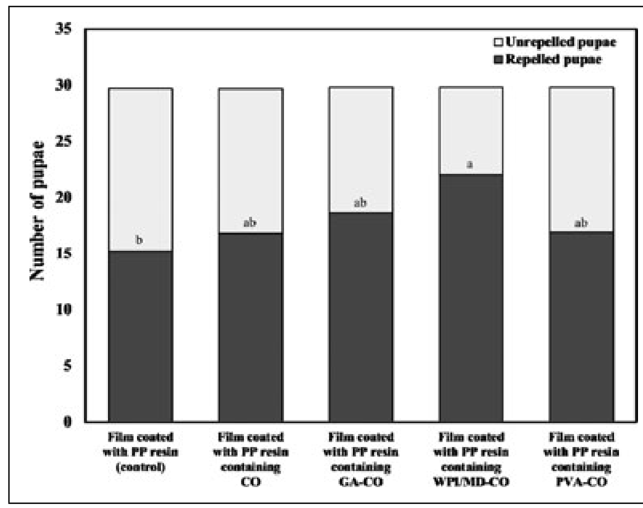

Figure 8–Insect-repelling effect of low-density polyethylene (LDPE) films coated with polypropylene (PP) incorporating cinnamon oil (CO) and microcapsule powders of gum arabic (GA)-CO, whey protein isolate (WPI)/maltodextrin (MD)-CO, and poly(vinyl alcohol) (PVA)-CO. The concentration of CO was 2% (w/w). The darker bar indicates the number of pupae found on the wall of the test container or on the aluminum foil surface; the difference between the value of a darker bar and 30 is the number found on the surface of each film. Means labeled with different letters are significantly different in the numbers of larvae found on the test film ($P < 0.05$).

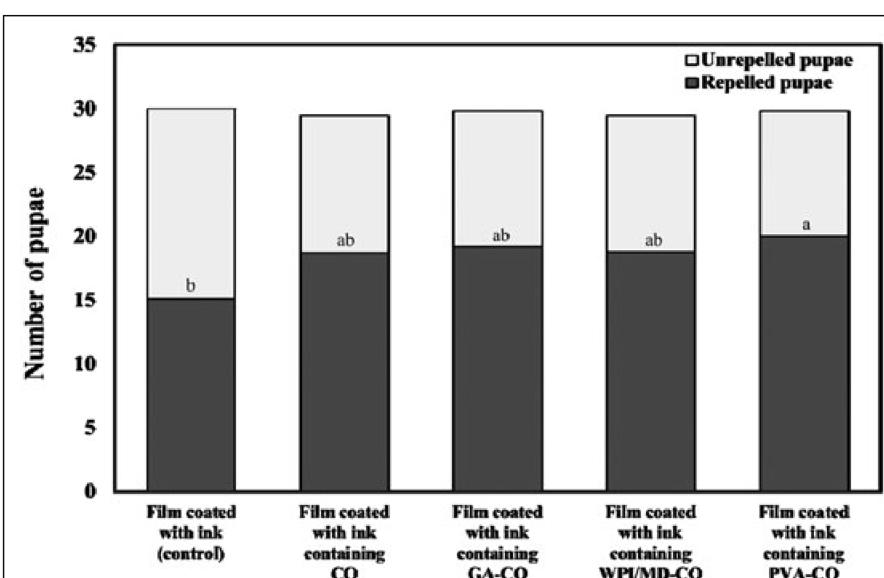

Figure 7–Insect-repelling effect of low-density polyethylene (LDPE) films coated with inks incorporating cinnamon oil (CO) and microcapsule emulsions of gum arabic (GA)-CO, whey protein isolate (WPI)/maltodextrin (MD)-CO, and poly(vinyl alcohol) (PVA)-CO. The concentration of CO was 2% (w/w). The darker bar indicates the number of pupae found on the wall of the test container or on the aluminum foil surface; the difference between the value of a darker bar and 30 is the number found on the surface of each film. Means labeled with different letters are significantly different in the numbers of larvae found on the test film ($P < 0.05$).

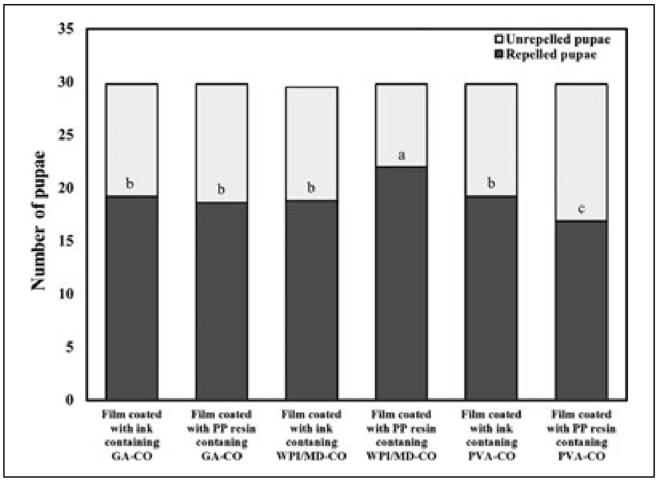

Figure 9–Comparison of insect-repelling effects of low-density polyethylene (LDPE) films coated with inks incorporating microcapsule emulsions made of cinnamon oil (CO) with gum arabic (GA), whey protein isolate (WPI)/maltodextrin (MD), and poly(vinyl alcohol) (PVA) and LDPE films coated with polypropylene (PP) incorporating microcapsule powders made of CO with GA, WPI/MD, and PVA. The darker bar indicates the number of pupae found on the wall of the test container or on aluminum foil surface; the difference between the value of a darker bar and 30 is the number found on the surface of each film. Means labeled with different letters reflect significantly different numbers of larvae found on the test film ($P < 0.05$).

microcapsule emulsions did not result in different repellencies than that for direct application of CO at 30 °C for 7 d (storage time). Microencapsulated active ingredients are released at controlled rates over prolonged periods of time (Madene and others 2006). Microencapsulation might be more effective over longer periods. The repelling effect of the film prepared with direct incorporation of the oil may have been stronger in the early days of storage but declined toward day 7 due to rapid release of repellent from the film.

The insect-repelling effects of the LDPE films coated with PP incorporating microcapsule powders are shown in Figure 8. The LDPE film coated with the PP incorporating microcapsule powder of WPI/MD demonstrated significantly greater repellency than the other films. The repellency was even stronger than that of the film coated with the ink incorporating the emulsion of PVA and CO ($P < 0.05$), which showed high repellency among the ink-coated films (Figure 7).

The repelling effects between the coatings with the ink and the coatings with the PP were compared (Figure 9) and were significantly different with WPI/MD and PVA ($P < 0.05$), but not with gum arabic ($P > 0.05$). When WPI/MD was used as the encapsulant, coating with the PP incorporating microcapsule powder resulted in greater repelling effect than did coating with the ink incorporating the microcapsule emulsion. However, when PVA was used, the coating with ink was more repellent than was that with PP. The results demonstrate that the repelling effect is influenced not only by the type of coating medium (ink or PP), but by the combination of coating medium and wall materials.

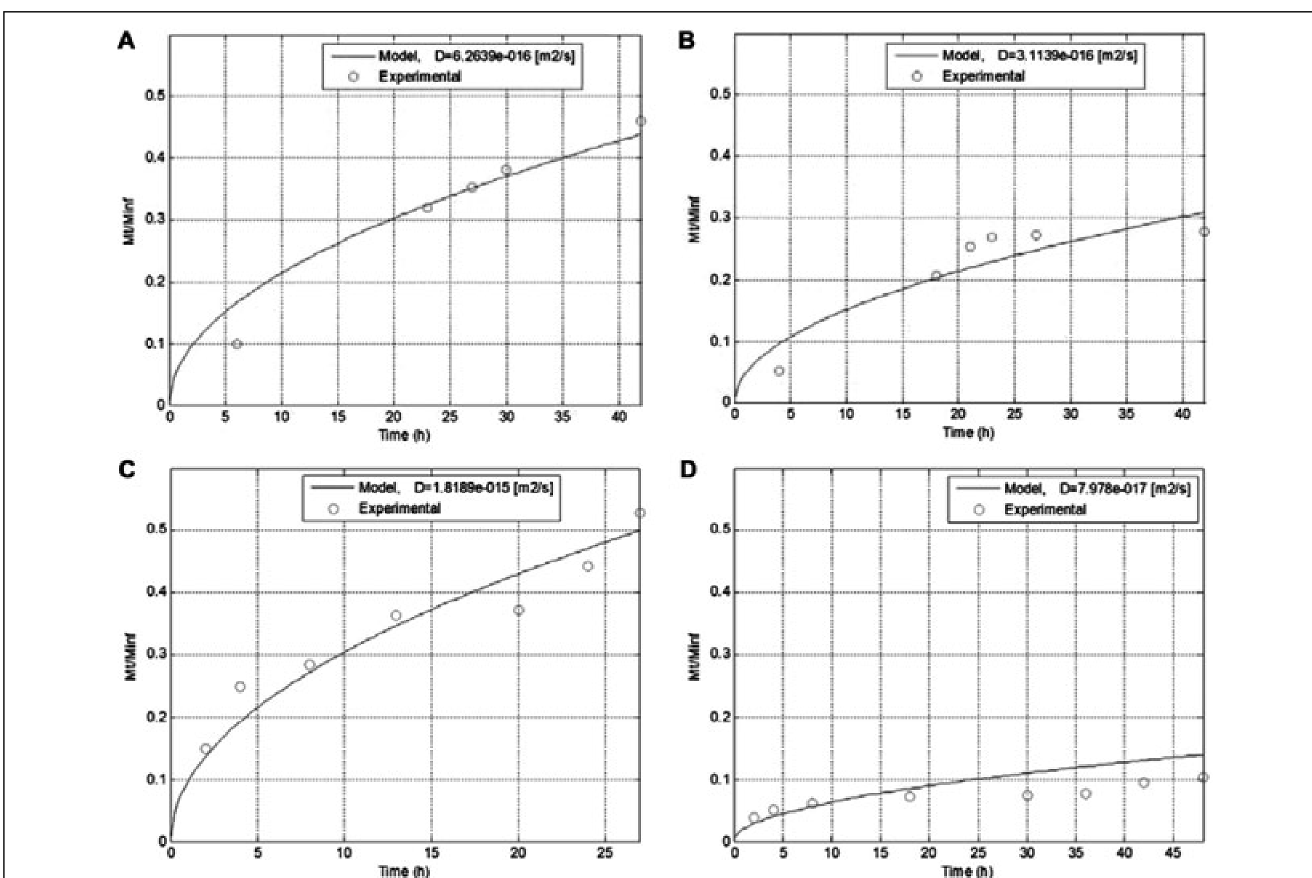

Figure 10–Release profiles of cinnamaldehyde in low-density polyethylene (LDPE) films coated with ink incorporating cinnamon oil (CO) without microencapsulation (A), ink incorporating the poly(vinyl alcohol) (PVA)-CO microcapsule emulsion (B), polypropylene (PP) incorporating CO without encapsulation (C), and PP incorporating the whey protein isolate (WPI)/maltodextrin (MD)-CO microcapsule powder (D).

Table 2–Effect of coating with cinnamon oil microcapsules on Indian meal moth larvae resistance.

Material	Nr of holes (percentage out of 27)	Time to penetration (h)
LDPE	25(92.6%)	20.8 ± 5.6^{b}
LDPE film coated with ink	25(92.6%)	20.4 ± 4.6^{b}
LDPE film coated with PP	24(88.9%)	20.4 ± 6.0^{b}
LDPE film coated with the ink incorporating microcapsules of PVA-CO emulsion	12(44.4%)	35.3 ± 18.9^{a}
LDPE film coated with the PP incorporating microcapsules of WPI/MD-CO powder	11(40.7%)	31.1 ± 10.4^{a}

Different superscript letters in the "Time to penetration" column indicate statistically significant differences between materials ($P < 0.05$).

Release profiles

Release profiles of cinnamaldehyde for different films were determined to assess the efficiency of the films at maintaining their repellency. The LDPE films coated with the ink incorporating the microcapsule emulsion of PVA and CO and coated with the PP incorporating the microcapsule powder of WPI/MD and CO, which demonstrated the highest repelling activity in the repelling test among the ink-coated LDPE films and the PP-coated LDPE films, respectively, were used as the microencapsulated CO-containing films. The profiles for LDPE films coated with inks and PP incorporating CO with and without microencapsulation are illustrated in Figure 10. The values of D for the diffusion of cinnamaldehyde in LDPE film coated with inks incorporating CO with and without microencapsulation were 3.1×10^{-16} and 6.3×10^{-16} and m^2/s, respectively. The values of D for the LDPE film coated with PP containing CO alone and that containing the microcapsule powder were 1.8×10^{-15} and 8.0×10^{-17} m^2/s, respectively. Using the microencapsulated emulsion with ink and using the microcapsule powder with PP, the rates of diffusion of cinnamaldehyde were reduced by approximately 2- and 23-fold, respectively. The release was approximately 4 times slower for the PP coating than for the ink coating. The different rate of release might be related to different structures of PP and ink. The components of ink include yellow organic pigment, toluene, and polyurethane. These materials have bulky structures, which could result in steric hindrance and thereby increase the resin-free volume. A faster migration of cinnamaldehyde could be allowed in the structure with a large free volume. The fact that the greatest repelling effect was found for the LDPE film coated with the PP incorporating microcapsule powder of WPI/MD might be related to its having the slowest release of cinnamaldehyde, resulting in long effectiveness retention. The microencapsulation developed in this study resulted in a slower release of cinnamaldehyde, implying adequacy for the method to be applied for extending effectiveness of insect repelling and controlling release of repellents from insect repellent films.

Tensile properties

The TS, %E, and EM of the uncoated LDPE film, the LDPE film coated only with ink without microcapsules, the LDPE film coated with ink incorporating the microcapsule emulsion of PVA and CO, the LDPE film coated with PP only, and the LDPE film coated with the PP incorporating microcapsule powder of WPI/MD were 10.1 to 21.5 MPa, 137.8 to 343.2%, and 208.4 to

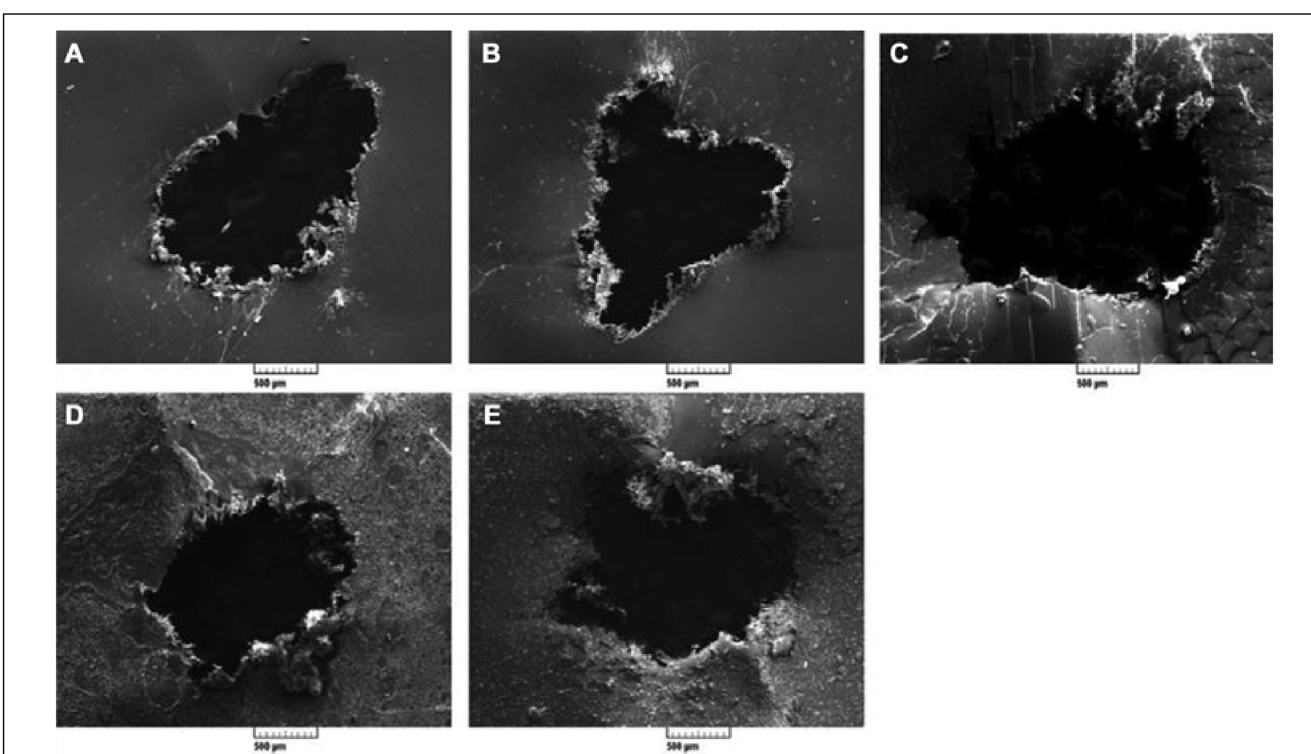

Figure 11–Micrographs of holes formed by *P. interpunctella* larvae on low-density polyethylene (LDPE) film (control) (A), LDPE film coated with ink incorporating cinnamon oil (CO) without microencapsulation (B), film coated with polypropylene (PP) incorporating CO without encapsulation (C), film coated with ink incorporating the poly(vinyl alcohol) (PVA)-CO microcapsule emulsion (D), and film coated with PP incorporating the whey protein isolate (WPI)/maltodextrin (MD)-CO microcapsule powder (E).

369.0 MPa, respectively. These values are similar to those reported previously for LDPE films (Briassoulis and others 1997; Han and others 2007). Each property was not affected by whether coatings were made with ink or PP ($P > 0.05$). The insect-repellent coatings did not modify the tensile properties of the LDPE, implying that replacement of uncoated with coated LDPE would not be an issue with current packaging equipment.

Penetration test

The numbers of test wells that had at least one hole and the time elapsed to form a hole are summarized in Table 2 Significantly less penetration occurred and shorter time elapsed to form a hole for the films coated with the microcapsule ink and PP ($P < 0.05$). The resistance of the microcapsule emulsion-coated film and the microcapsule powder-coated film were not different in terms of the number of holes and the times to penetration. Penetration was probably not affected by the difference in the rate of release of the repellent between the powder film and the emulsion film (Figure 10). Invasion was successfully prevented, that is, the cookies were protected, by the microcapsule-coated LDPE films. The holes made in the films by the larvae are shown in Figure 11. All the film samples displayed similar hole shapes and sizes as well as scratches and tears around holes. The sizes of the holes on all the samples were not significantly different ($P > 0.05$) and in the range of 980 to 1990 μm. The shapes of holes and edges formed on packaging materials by insects were reported influenced by tensile properties of the materials; more scratches and tears were observed with a material with high %E and low TS (Chung and others 2011). Insignificant differences in tensile properties of the film samples in this study could partly result in the undistinguishable shapes of holes and edges. The results from image analysis also imply that the released repellent did not alter mechanisms for the larvae in penetrating packages.

Conclusion

The LDPE-based films that effectively repelled Indian meal moth larvae were developed. The film coated with ink incorporating a microcapsule emulsion of PVA and CO and the film coated with PP incorporating a microcapsule powder of WPI/MD and CO were the most effective films among the ink-coated films and the PP-coated films, respectively, in terms of encapsulation efficiency and repellency. Microencapsulation resulted in a slower release of the insect repellent, showing opportunity for a controlled release of the repellent in insect-repelling food packaging. The ink- or PP-containing microcapsules did not modify the tensile properties of the LDPE film. The penetration of India meal moth to a model food (cookie) was successfully prevented by the test films. Thus, the insect-repellent films developed in this study could potentially be used to protect food products from invasion of Indian meal moth.

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