Diffusion of Linalool and Methylchavicol from Polyethylene-Based Antimicrobial Packaging Films

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Abstract

The diffusion of linalool and methylchavicol from thin (45-50 μm) antimicrobial low-density polyethylene-based films was evaluated after immersion in isooctane and the effect of temperature (4, 10, or 25 °C) on the diffusion rate was evaluated. The kinetics of linalool and methylchavicol release showed a non-Fickian behavior at the lowest temperature. An increase in temperature from 4 °C to 25 °C resulted in an increase in the diffusion coefficient from 4.2×10^{-13} m^2 s^{-1} to 2.5×10^{-12} m^2 s^{-1} for linalool and from 3.5×10^{-13} m^2 s^{-1} to 1.1×10^{-12} m^2 s^{-1} for methylchavicol. The effect of temperature on the diffusion coefficient followed an Arrhenius-type model ($r^2 = 0.972$) in relation to a time-response function with a Hill coefficient. Activation energies of 57.8 kJ mol^{-1} (linalool) and 42.8 kJ mol^{-1} (methylchavicol) were observed.

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1. Introduction

In solid and semi-solid foods, surface growth of microorganisms is one of the major causes of food spoilage (Maxcy, 1981). To overcome this problem, attempts are being made to develop antimicrobial (AM) packages in which AM agents are incorporated into the packaging material and slowly released onto the food surface (Han, 2000; Appendini & Hotchkiss, 2002; Suppakul Miltz, Sonneveld, & Bigger, 2003a). Such materials may have a crucial effect on the food quality and safety and/or on the shelf life extension of packaged food products. The controlled release of different AM agents from food packaging materials has been studied and reported in the literature (Mastromatteo, Mastromatteo, Conte, & Del Nobile, 2010).

Naturally-derived AM agents are perceived by consumers as having a low health risk. Therefore, there is an increasing interest in the evaluation and possible application of these compounds (Nicholson, 1998). The principal constituents of basil, linalool and methylchavicol, exhibit an AM effect against a wide range of microorganisms (Suppakul, Miltz, Sonneveld, & Bigger, 2003b). These compounds are generally recognized as safe (i.e. possess "GRAS" status), are relatively stable at high temperatures and therefore have the potential to be used in AM film applications. In recent studies (Suppakul, Miltz, Sonneveld, & Bigger, 2006; Suppakul, Miltz, Sonneveld, & Bigger, 2008), linalool and/or methylchavicol were incorporated into polyethylene-based films. The physical properties of the films (mechanical, barrier, optical and thermal) and the antimicrobial efficacy of the films were investigated. Apart from the properties and AM efficacy of the films, an understanding of the diffusion controlled release rate is an essential aspect for developing appropriate AM food packaging materials.

Antimicrobial films represent an application in which active substances (AM agents) present in the polymeric matrix migrate onto the surface of packaged products.

The release profile from an AM film occurs in the opposite direction to sorption (such as flavor scalping) (Sadler & Braddock, 1991). The diffusivity of the AM agent in the polymer is a characteristic parameter providing important information required for the prediction of the rate of release of the AM agent from the film (Han & Floros, 2000).

The present paper concentrates on evaluating the rate of diffusion of linalool and methylchavicol in AM low-density polyethylene-based films (LDPE films) and their migration into isooctane, simulating to some extant the migration of these agents onto the non-polar regions on the surface of hard cheeses that are created by fats, lipids and such species.

2. Materials and methods

2.1 Antimicrobial films

Low-density polyethylene-based films of 45-50 μm in thickness with and without linalool (MW = 154.25 g mol^{-1}, purity 97%, b.p. = 198.5C; L260-2, Aldrich Chemical Company, Inc., USA,) or methylchavicol (MW = 148.20 g mol^{-1}, purity 98%, b.p. = 216C; AUSTL 21320, Aurora Pty. Ltd., Australia) were prepared from commercially obtained LDPE pellets (Alkathene XJF 143, Qenos Pty. Ltd., Australia). A pre-blended master batch of ethylene vinyl acetate (EVA, ELVAXR®_3120, Dupont Ltd., Australia) copolymer powder containing approximately 15% w/w linalool or methylchavicol was mixed with virgin LDPE pellets and manufactured into films with a concentration of 1.5% w/w linalool or methylchavicol at a ratio of 10% w/w EVA to 90% w/w LDPE master batch by extrusion film blowing in a single screw extruder (Telford Smith, Australia). The temperature in the extruder was approximately 160°C (all zones). Films without linalool or methylchavicol were prepared under similar conditions by the same method and were used as controls.

2.2 Film thickness measurement

A hand-held micrometer (Hahn & Kolb, Stuttgart, Germany) was used for measuring film thickness. Five readings were taken for each sample, one at the sample center and four around the perimeter.

2.3 Quantification of agents by gas chromatography

The amount of linalool or methylchavicol in the samples was determined by gas chromatography (GC). The procedure was as follows: the film (5 g) was extracted for 18 h by Soxhlet extraction using 150 mL of isooctane. An aliquot of the extract with a precisely known volume was then sampled for GC analysis. A Varian Star 3400-CX GC equipped with a fused silica capillary column DB-5 (30 m × 0.25 mm i.d., film thickness 0.25 μm, J & W Scientific, USA) was used. The following conditions were applied: sample volume, 1.0 μL; initial column temperature, 80°C; heating rate, 5°C min^{-1} to 180°C that was then held for 5 min more; injector temperature, 250°C, split ratio, 1:100; FID detector temperature, 300°C; carrier gas, nitrogen. The linalool and methylchavicol contents of the samples were calculated from prepared standard curves.

2.4 Diffusion experiments

The release of linalool and methylchavicol from the AM LDPE-based films was investigated by immersing 4 pieces (5 × 5 cm) of the test film in 100 mL of isooctane (Unichrom 2516-2.5L, GL grade, APS Chemicals Ltd., Australia), as a fatty food simulant, in a closed system and storing at 4, 10 or 25°C in an incubation shaker (Innova™ 4230, New Brunswick Scientific, U.S.A.) with a continuously rotating speed of 30 rpm. The flasks were incubated with mild agitation, simulating agitation during storage

and transportation (Appendini & Hotchkiss, 2002). It is believed that under these conditions a steady-state transfer of AM agents from the film occurs. Aliquots were sampled at various times. Experiments were performed in triplicate.

The amount of linalool or methylchavicol in the aliquot was determined using GC. An aliquot of the extract of a precisely known volume was injected into the GC for analysis. The GC was operated using the conditions described above. The linalool and methylchavicol contents of the samples were calculated from previously prepared standard curves.

2.5 Kinetics analysis of linalool and methylchavicol release from LDPE-based films

The relationship between the sorption and the desorption of a given species within a polymeric matrix is given in Eq. 1:

$$[M_t/M_\infty]_{desorption} = 1 - [M_t/M_\infty]_{sorption} \quad (1)$$

where M_t is the total amount of a species that has migrated after time t, and M_∞ is the maximum amount of the species that can migrate after an infinite time, ($t = \infty$, namely, at equilibrium). The ratio M_t/M_∞ is known as the fractional mass release.

Several methods have been reported to be appropriate for measuring diffusion of small molecules in a polymer (Crank, 1975; Giannakopoulos & Guilbert, 1986; Miltz, 1987; Lim & Tung, 1997). Redl, Gontard & Guilbert (1996) suggested a relatively rapid and convenient method to determine diffusivity of a species in AM films by immersion in food simulants (Feigenbaum, Riquet & Scholler, 2000; McCort-Tipton & Pesselman, 2000) such as distilled water, buffer solution, isooctane, ethanol, acetic acid and rectified olive oil.

In the current study, the question of whether the fractional mass release ratio is directly proportional to $t^{1/2}$ was considered first, since such a linearity would indicate compliance with the general law of diffusion (Crank, 1975). The diffusion coefficient D (m^2 s^{-1}) of linalool and methylchavicol were later calculated using the half-time method given in Eq. 2 (Miltz, 1987; Lim & Tung, 1997; Han & Floros, 2000; Ouattara, Simard, Piette, Begin & Holley, 2000):

$$D = 0.0491 \times L^2/t_{0.5} \quad (2)$$

where L is the thickness of the film, and $t_{0.5}$ is the time required for 50% of the migrating species to be released into the simulant (i.e. when $M_t = 0.5M_\infty$).

Theoretical values of the fractional mass release as a function of time were calculated assuming an exponential rise to a maximum level as indicated in Eq. 3 (Schwartzberg, 1975; Lim & Tung, 1997):

$$M_t/M_\infty = 1 - \exp(-kt) \quad (3)$$

where k is the empirically obtained rate constant (s^{-1}) that dependents on the mass transfer properties, geometry and other conditions of the film material (Han & Floros, 2000).

In order to determine the temperature dependence of the diffusion coefficient, the well-known Arrhenius equation (Eq. 4) was used (Chatwin, 1996):

$$D = D_0\exp(-E_a/RT) \quad (4)$$

where D_0 is a pre-exponential factor, E_a is the activation energy, R is the ideal gas constant and T is the absolute temperature. The parameters D_0 and E_a can be obtained by curve fitting of the experimental data (Helmroth, Rijk, Dekker & Jongen, 2002).

The data were also analyzed by the time response function using a Hill coefficient in accordance with Eq. 5:

$$M_t/M_\infty = 1/[1 + (k/t)^n] \quad (5)$$

where k is a rate constant and n is the Hill coefficient, indicating the degree of "cooperativity" of the agent (Hill, 1984).

2.6 Data analysis

The initial part of the migration curves (i.e. values of $M_t/M_\infty < 0.6$), that has been defined as the "short-term migration" (Miltz, 1987), was plotted against the square root of time, $t^{1/2}$, and tested for linearity using a linear correlation procedure (KyPlot 2.0 for Windows, Kyence Inc, Japan). The kinetic results were also analyzed using a time-response function with a Hill coefficient to determine the rate constant of the kinetic equation. A two-way ANOVA with replication procedure was applied to evaluate the significance of the main effects of temperature and time as well as their interaction.

3. Results and discussion

3.1 Film preparation

A constant temperature of approximately 160°C was applied along the extruder in order to minimize the loss of active agents by evaporation, as recommended in the literature (Han, 2000). Although a loss of the active agents was observed during the

extrusion process, it was significantly lower than the losses observed in a previous study with linear low-density polyethylene (LLDPE) alone (Suppakul et al., 2006). The actual amount of linalool or methylchavicol in the extruded films was found to be 0.34% w/w in each film. This increased retention of the active agent (compared to 0.05% w/w in the previous study) may be attributable to the lower extruder temperature and/or the interaction between the active agent and the EVA copolymer. This copolymer may assist in solubilizing or partially "anchoring" the active molecules within the polymeric matrix. Linalool-LDPE-based and methylchavicol-LDPE-based films were 47.6 μm and 48.1 μm thick, respectively.

3.2 *Migration of linalool and methychavicol from LDPE into isooctane*

The experimental migration data of linalool and methylchavicol from the LDPE-based films immersed in isooctane (used as a fatty food simulant) at different temperatures are shown in Fig. 1. The migration curves at 4°C for linalool and methylchavicol using curve fitting involving Hill coefficients of 1.92 and 1.72 respectively are shown in Fig. 2. It can be seen that the migration rate is at a maximum immediately after a lag time of *ca*. 60 s and declines progressively thereafter until the extent of migration becomes nearly complete after *ca*. 1800 s for both AM agents. The linearity achieved in all cases when the data associated with the initial portions of the curves (i.e. $M_t/M_\infty < 0.6$; Miltz, 1987) in Figure 1 were fitted with respect to the $t^{1/2}$ model of the initial portion of the curve was quite good (r^2 ranging from 0.899 to 0.985). However, the kinetics of linalool and methylchavicol release from the films was fitted considerably better ($r^2 = 0.994$ and $r^2 = 0.993$ respectively) with a nonlinear, least-squares fit of the time-response function using a Hill coefficient (Eq. 5).

206 In view of the latter, the release of linalool and methylchavicol from LDPE-based
207 films immersed in isooctane, might be described by the "swelling-controlled" model for
208 drug release that was previously reported by Armand, Magbard, Bouzon, Rollet, Taverdet,
209 & Vergnaud (1987). According to this model, a simulant such as isooctane penetrates first
210 into the polymer matrix and dissolves the AM agents thereby enabling their subsequent
211 release. Indeed, it is expected that an isooctane uptake will cause polymer swelling
212 (Feigenbaum et al., 2000) because the solubility parameter of isooctane is close to that of
213 LDPE (Brydson, 2000). The migration of linalool and methylchavicol is thus expected to
214 increase with an increase in isooctane penetration into the LDPE-based film, reaching a
215 plateau when the matrix is saturated with isooctane (Armand et al., 1987). The
216 experimental results obtained in the current study are described well by this model and
217 evidence for this is the slight lag time that is apparent in the release curves shown in
218 Figure 2. Nonetheless, the importance of swelling could be further investigated by
219 following its extent as a function of the temperature in order to more fully characterize the
220 lag time. In reality, the situation may be more complex and the "swelling-controlled"
221 model may only be valid in some cases. Many interactions take place during the migration
222 of species from polymers into liquids. Moreover, Lim & Tung (1997) reported that a
223 time-dependent relaxation process occurs as a result of the swelling that takes place during
224 the diffusion of the liquid into the polymer. As a consequence, release rates change
225 continuously and the accurate mathematical analysis of the migration is difficult
226 (Gnanasekharan & Floros, 1997).
227 In the present study, the initial portion of the migration curves was found to be,
228 more or less, in accordance with the predictions of Fick's law for diffusion. However,
229 evidence for the non-Fickian nature of the diffusion appears in the sigmoidal shape of the
230 migration curves, especially at low temperatures. This indicates interactions that cause the

migration curves to display sigmoidal kinetics. The upward curvature of the experimental sorption curve shows a constant increase in the diffusion coefficient. The penetration of isooctane molecules facilitates further penetration by the plasticization of the polymer matrix, until a plateau is reached (Feigenbaum et al., 2000). This suggests that the release of linalool and methylchavicol from LDPE-based films is not determined by diffusion alone (Peppas, 1985). Furthermore, the fractional mass release, plotted as a function of time, was better fitted by a time-response function with a Hill coefficient (Eq. 5) than by an exponential rise of M_t/M_∞ to a maximum level (Eq. 3). These findings are in agreement with those of Ouattara et al. (2000) who reported a non-Fickian behavior for the diffusion of acetic and propionic acids from chitosan-based films into buffer solutions. Consequently, the non-Fickian behavior observed in the present study is most likely due to simultaneous swelling (due to isooctane uptake) and outward diffusion of linalool or methylchavicol (Ouattara et al., 2000).

3.3 Effect of temperature on diffusion

The migration data showed a significant effect of temperature on the release of linalool and methylchavicol from the polymeric matrix, as qualitatively indicated in Fig. 1 where raising the temperature from 4 to 25°C clearly causes a faster rate of migration for both agents. In particular, the time required to release half the amount of linalool contained initially in the LDPE-based film decreases from 238 s at 4°C to 165 s at 10°C and to 42 s at 25°C, whereas the corresponding times for methylchavicol at the respective temperatures are 327 s, 231 s, and 97 s. Furthermore, the diffusion coefficient, D, of linalool calculated from the half-time method (Eq. 2) increased from 4.2×10^{-13} m^2 s^{-1} to 2.5×10^{-12} m^2 s^{-1}, and the corresponding rate constant k (Eq. 5) decreased from 251 to 44

s^{-1}, when the temperature was increased from 4 to 25°C. Similar behavior is observed in the case of methylchavicol (see Table 1).

At all temperatures both linalool and methylchavicol showed a positive affinity for isooctane as indicated by the Hill coefficients being greater than unity. Furthermore, in the case of linalool there is no statistically significant difference ($p > 0.05$) in the Hill coefficient within the temperature range of 4 to 25°C. This is in agreement with the notion that the Hill coefficient of a given system is temperature-independent. However, at 10°C, the Hill coefficient of methylchavicol was found to be 1.35 which lies outside the expected range of between 1.67-1.72. The reasons for this apparent anomaly remain unclear at present.

In order to further explore the effect of temperature on the kinetics of migration, Arrhenius plots of the data presented in Table 1 were constructed and these appear in Figure 3. It can be seen from the plots that each of the analysis methods indicates the rate of linalool migration is more temperature-sensitive than that of methylchavicol within the temperature range investigated. The temperature dependence of the diffusion coefficient is well described by an Arrhenius relation with activation energies of 58.0 kJ mol^{-1} and 38.2 kJ mol^{-1} obtained for linalool and methylchavicol respectively. The activation energies obtained from the analysis of a time-response function with a Hill coefficient were found to be 57.8 kJ mol^{-1} and 42.8 kJ mol^{-1} for linalool and methylchavicol respectively. Taken collectively, these data confirm the consistency between the two methods of analysis used in this case. In particular, the activation energy is a measure of the sensitivity of the diffusion coefficient to temperature (Chung, Papadakis & Yam, 2001) and the values of the activation energies derived from the diffusion coefficient data are close to those derived from the half-time method equation. The latter is normally used for the evaluation of the approximate diffusion coefficients (Lim, & Tung, 1997; Ouattara

et al., 2000; Teerakarn, Hirt, Acton, Rieck & Dawson, 2002). These data also reflect the expected doubling of the diffusion coefficient for approximately every 10°C rise in temperature.

The dependency of the rate of diffusion of linalool and methylchavicol from LDPE-based films from the point of view of a pure diffusion model is in many cases explained by temperature effects on the solubility of the diffusing molecules in films, on the nature of adhesive forces at interfaces (Brydson, 2000), and on the molecular mobility (Myint, Daud, Mohamad, & Kadhum, 1996). As the molecular weight of linalool is only slightly higher than that of methylchavicol, it is likely that the different mobility of these species within the polymer matrix may be due to either their different shapes or polarities. Indeed the higher polarity of the linalool molecule compared with methylchavicol may explain its greater mobility and sensitivity of its diffusion coefficient to temperature. This is because the exudation of a polar species from a non-polar matrix such as LDPE occurs more readily compared to a non-polar species that will tend to be retained in the matrix. The fact that the relationship between diffusion and temperature is well described in the present study by the Arrhenius equation, suggests that the effect of temperature is thermodynamic in nature, regulated essentially by the proportion of energy provided to the activation energy (Daniels, & Alberty, 1972).

4. Conclusions

Low-density polyethylene-based films containing linalool and methylchavicol have been proposed as AM packaging materials. In migration studies of the AM agents into isooctane, used as a fatty-food stimulant, the diffusion coefficient and the temperature sensitivity of migration of linalool were found to be higher than those of methylchavicol. Sigmoidal-shape diffusion curves, especially at low temperatures, indicated that diffusion

of the AM agents in the polymer was not purely Fickian in nature. The fractional mass release, plotted as a function of time, was better fitted by a time-response function with a Hill coefficient than by an exponential rise in this value to a maximum.

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Table 1: Effect of temperature on the migration of linalool and methylchavicol from LDPE-based films into isooctane

Agent	Temperature T/°C	Thickness[1] L × 10^6/m	Diffusion Coeff.[2] D × 10^{12}/m^2 s^{-1}	Rate Constant[3] k/s^{-1}	Hill Coeff.[4] n
linalool	4	47.0±1.8	0.42[a]	250.7[c]	1.92
	10	47.3±2.0	0.68[b]	167.2[b]	1.87
	25	48.4±1.4	2.46[c]	44.5[a]	1.93
methylchavicol	4	48.0±1.6	0.35[a]	346.0[c]	1.72[b]
	10	48.7±1.1	0.44[b]	296.7[b]	1.35[a]
	25	47.5±0.3	1.10[c]	99.1[a]	1.67[b]

[1] For each AM agent, thickness values are non-significantly different ($p > 0.05$).

[2] For each AM agent, *D* values with different letters are significantly different ($p \leq 0.01$).

[3] Rate constant obtained by nonlinear regression. For each AM agent, *k* values with different letters are significantly different ($p \leq 0.01$).

[4] For each AM agent, *n* values with different letters are significantly different ($p \leq 0.05$).