

Interactions between Aroma and Edible Films. 1. Permeability of Methylcellulose and Low-Density Polyethylene Films to Methyl Ketones

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This work contributes to the study of aroma transfers through edible and plastic packaging films. Permeability, sorption, and diffusivity of three methyl ketones (2-heptanone, 2-octanone, and 2-nonanone) in and through low-density polyethylene and methylcellulose-based edible films have been determined. Permeability was measured using a dynamic method coupled with a gas chromatograph. The methyl ketone permeability of polyethylene films mainly depends on diffusivity of the penetrant in the polymer. In the case of 2-heptanone, a saturation of the polymer network is observed at high vapor concentrations. The formation of clusters could take place when concentrations are higher in the vapor phase. Physicochemical interactions between aroma compounds and components of the methylcellulose-based film induce structural changes such as plasticization. Therefore, the diffusion step depends on the aroma concentration differential, and permeability is essentially driven by the sorption.

Keywords: Methyl ketones; permeability; sorption; diffusivity; methylcellulose; LDPE

INTRODUCTION

The loss of quality in foodstuff can be related to the transport of small molecules (water, salts, pigments, or aroma compounds), which is why the food industry uses glass, metal, and plastic packaging to protect food products. However, the selective loss of aroma compounds through the plastic materials induces either a reduction of the flavor intensity or a change in the food's aroma. Packaging must limit these modifications, particularly the change in the sensory quality.

The performance of packaging materials depends on their efficiency to reduce transfers between foodstuff and the environment such as the transport of volatile compounds from food to the outer atmosphere or from the surrounding medium to the packaged product (permeation) and the adsorption of volatile compounds at the surface of the packaging (sorption or scalping) (Reineccius, 1991; Benet et al., 1992; Blumenthal, 1997). Technological progress in synthetic packaging materials allows an important and selective reduction of noncondensable gas and water vapor exchanges such as that obtained with barrier polymers (Chomon, 1992). Nevertheless, most plastic packagings have a strong affinity toward hydrophobic volatile compounds, such as aroma compounds (Reineccius, 1991; Leufvén and Stöllman, 1992; Miller and Krochta, 1997). Moreover, plastic materials cannot be used to protect all kinds of foods: for example, to separate two different parts in a heterogeneous product such as a pie or a pizza (Kamper

and Fennema, 1985; Kester and Fennema, 1989). Therefore, the combination of edible and plastic materials has been proposed to improve their functional properties.

The barrier efficiency against vapor transfer through an edible or a synthetic film can be determined by measuring the mass transfer of different substances. This transfer depends mainly on both sorption and diffusion. The sorption mechanism consists of adsorption, absorption, and/or desorption of penetrant molecules and reveals the polymer–volatile compound affinity, whereas diffusion is related to their mobility within the polymeric network of the material.

The volatile compound and film polymer characteristics must be taken into account to explain the transfer process. However, other structural factors exist that can affect sorption and diffusion, that is, the type of polymerization, the polymer tridimensional structure, the material cohesion maintained by weak energy bonds, or the polymer's glass transition temperature (Hernandez et al., 1992; Matsui et al., 1992; Giacin, 1994; Gavara et al., 1996). In addition, the physicochemical characteristics of volatile compounds influence the film permeability: an aroma compound's shape and size affect its diffusivity, whereas solubility is influenced by the compound's nature, polarity, and ability to condense (Rogers, 1985).

The solute polarity is an important factor in sorption process. Some authors showed that aroma compounds are adsorbed more easily in the polymeric film if their polarities are similar (Shimoda et al., 1987; Arora et al., 1991; Matsui et al., 1992). However, very few authors studied the effect of this factor on transfer through hydrophilic edible packaging.

Previous works showed that hydrophobicity of some films has a determining effect on the transfer, but this relationship seems to be complex. Indeed, some authors

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Table 1. Physicochemical Characteristics of Methyl Ketones

characteristic	2-heptanone	2-octanone	2-nonanone
formula	CH ₃ -CO-(CH ₂) ₄ -CH ₃	CH ₃ -CO-(CH ₂) ₅ -CH ₃	CH ₃ -CO-(CH ₂) ₆ -CH ₃
mol wt (g mol ⁻¹)	114.18	128.21	142.24
density at 20 °C (g mL ⁻¹)	0.8166	0.8185	0.8317
solubility in water at 25 °C (ppm v/v)	5200	2000	470
log <i>K</i> ^a	1.8	2.3	2.9
reported concn in food products ^b (μg mL ⁻¹)	2.7–25.0	0.1–4.0	0.1–4.0

^a Calculated according to the method of Rekker (1977). ^b Fenaroli (1975).

studied the influence of the distribution and physicochemical characteristics of the hydrophobic part in composite films on their barrier efficiency against water transfer (Martin-Polo, 1991; Debeaufort and Voilley, 1994; Johansson and Leufven, 1994), but they did not find a model describing both transfer and physicochemical phenomena. Furthermore, aroma barrier properties of edible films have not been as thoroughly studied as water and oxygen transfer have been (Miller and Krochta, 1997).

The aim of this research was to better understand the effect of the physicochemical properties of the aroma compounds, mainly their hydrophobicity, on their transfer through edible membranes and to compare the barrier performance of low-density polyethylene (LDPE) films.

MATERIALS AND METHODS

Aroma Compounds. The selected volatile compounds are three methyl ketones; their physicochemical characteristics are summarized in Table 1. They were chosen because they are present in many food products (Fenaroli, 1975) at relatively high concentration, mainly in dairy products.

The hydrophobicity is expressed by log *K* (where *K* is the partition coefficient of the aroma compound between octanol and water); 2-heptanone can be considered as a hydrophilic compound (log *K* < 2), whereas 2-octanone and 2-nonanone are hydrophobic ones (log *K* > 2).

Plastic Film. A 25 ± 1 μm thick LDPE film Riblene FF30 was provided by EniChem Polymères France S.A. (France). Its density at 25 °C is 0.924 g mL⁻¹.

Model Edible Film. This film was prepared with methylcellulose, a hydrosoluble cellulose derivative with an average molecular weight of 17000 Da and corresponding to 110 glucose units, a 40 mPa·s viscosity for a 2% solution at 25 °C, and a 27.5–31.5% degree of methyl substitution (Benecel MO21, Aqualon, France). The edible films contained 25% (m/m of dry matter) of polyethylene glycol 400 (PEG400, Merck, Darmstadt, Germany) used as plasticizer.

Preparation of Films. Edible films were obtained by solubilization of 4.25 g of methylcellulose in 75 mL of a water/ethanol solution (3:1 v/v) at 75 °C under magnetic stirring for 10 min. Afterward, 1.3 mL of PEG 400 was added and the viscous solution was kept for 5 min under the same conditions. The solution was spread using a thin-layer chromatography spreader onto glass plates covered by poly(vinyl chloride) to facilitate the unsticking of films. Films were dried at 50 °C and 7% relative humidity for 12 h. The final thickness of the films was 25 ± 3 μm.

Vapor Permeability Measurement. A dynamic measurement method of aroma vapor fluxes through films or membranes was used. The apparatus has been described by Debeaufort and Voilley (1994). The permeation cell was composed of two chambers divided by the film to be studied. The film area exposed to transfer was 15.9 cm². The two chambers were continuously swept by a 30 mL min⁻¹ helium flow. The aroma concentrations in the vapor phase on the upper side of the cell were obtained by mixing two flows: one containing the volatile compound and the other dry helium. Flows containing vapors were obtained from bubbling dry helium through pure compounds. Organic volatiles were

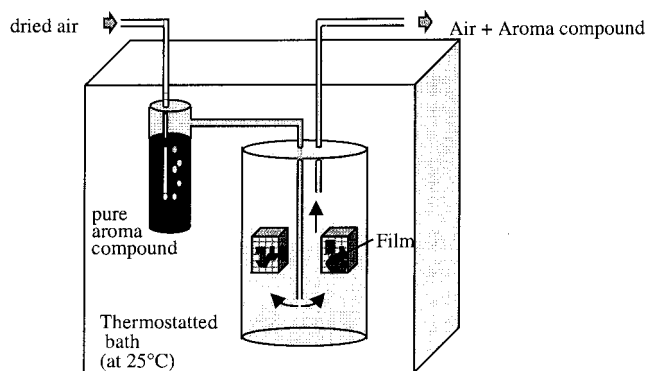


Figure 1. System to determine aroma compound sorption kinetics.

analyzed with a flame ionizing detector (FID). Samples were weighed before and after permeation measurements to determine the amount, *Q*, of volatile sorbed within the films, expressed as micrograms of volatile per milliliter of dry film (μg mL⁻¹ d.f.). The gravimetric method used to determine *Q* was compared for some samples to values obtained according to the method described further in this paper, and they were considered as enough accurate.

Films were equilibrated at 0% relative humidity at 25 °C before permeability determinations. Permeation measurements were carried out at 25 °C. Vapor concentration differentials were between 0 and 2.5 μg mL⁻¹ He for 2-nonanone, between 0 and 7.0 μg mL⁻¹ He for 2-octanone, and between 0 and 23 μg mL⁻¹ He for 2-heptanone. The highest concentration of aroma in the vapor phase (saturation) was obtained by bubbling a carrier gas through pure aroma at 25 °C and atmospheric pressure, the concentration of which was checked and measured by gas-liquid chromatography (GLC).

Vapor Sorption Measurements. The quantity of aroma compounds adsorbed in the film at the steady state of the mass transfer was obtained using a modified microatmosphere method: 0.3 g of dried films cut in small pieces (0.25 cm²) was exposed to atmospheres saturated with pure aroma compound (Figure 1). This atmosphere was conditioned at 0% relative humidity and continuously swept with a carrier gas (helium) containing known vapor concentration of aroma compounds. The atmosphere inside the flask containing the film was kept at a constant aroma concentration. The total amount of volatile compound sorbed at a given time until constant *Q* (3–5 days) was determined by extraction with *n*-hexane in a sample of the film and by injection of the resulting aroma solution in a gas-liquid chromatograph (GLC). The extraction yield was 97%. The quantities of methyl ketone adsorbed in the films are expressed as μg mL⁻¹ of d.f.

Calculations. The aroma flux was defined as the ratio of the weight of permeated vapors (g) to the product of exposed area (m²) and time (s). The flux was expressed as g m⁻² s⁻¹.

Each measurement was made at least in triplicate, and differences between means were tested statistically at *p* < 0.05 level using the Student–Newman–Keuls *t* test of SAS (Statistical Analysis System version 6.09, 1991).

RESULTS AND DISCUSSION

Figures 2 and 3 present the evolution of methyl ketone flux through polyethylene and methylcellulose

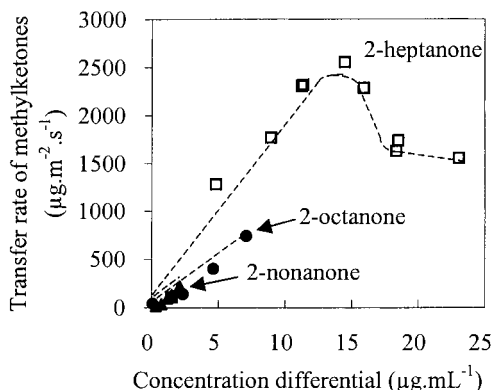


Figure 2. Transfer rate of three methyl ketones through LDPE films as a function of their concentration rate at 25 °C.

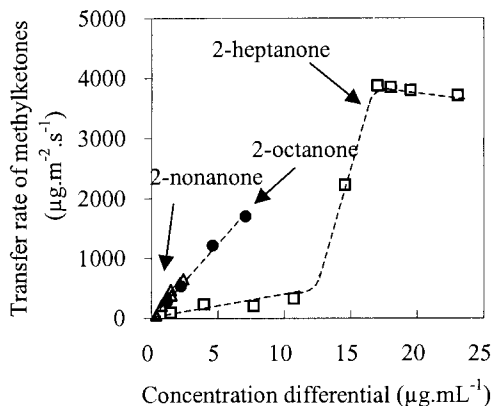


Figure 3. Transfer rate of three methyl ketones through methylcellulose-based films as a function of their concentration rate at 25 °C.

films versus aroma vapor concentration differential at 25 °C. Up to a concentration differential of 10 $\mu\text{g mL}^{-1}$, the flux increases for the three methyl ketones in both methylcellulose and polyethylene films ($r^2 > 0.98$, at $\alpha < 0.05$). Over 10 $\mu\text{g mL}^{-1}$ the 2-heptanone flux does not increase linearly and depends on the nature of the film. This phenomenon is not observed for 2-octanone and 2-nonanone because of their lower volatilities. Indeed, the maximal concentrations obtained in the vapor phase for 2-octanone and 2-nonanone are, respectively, 7 and 2.5 $\mu\text{g mL}^{-1}$ at 25 °C and atmospheric pressure, whereas the maximal concentration is 23 $\mu\text{g mL}^{-1}$ for 2-heptanone.

Transfer of Methyl Ketones through LDPE Films.

The 2-heptanone flux through LDPE film increases linearly with the concentration gradient up to a value of 13 $\mu\text{g mL}^{-1}$. For concentration gradients $> 13 \mu\text{g mL}^{-1}$, the transfer rate decreases until a value of 2000 $\mu\text{g m}^{-2} \text{s}^{-1}$ is reached. Several hypotheses can be proposed to explain this behavior.

(A) A mechanism of antiplasticization of the polymer could be caused by 2-heptanone. Thus, the aroma compound probably modifies the polymer structure and increases the T_g of the LDPE. The absence of functional groups able to react with the methyl ketone and the great stability of the C–C bond in the polyethylene string lead us to eliminate this possibility. Indeed, polyethylene is frequently used to protect foodstuffs for its nonreactivity with some compounds and particularly water (Reineccius, 1991; Hernandez, 1994).

(B) Transfers are in general described by the model of sorption–diffusion, which defines permeability as the

Table 2. Vapor Partial Pressure (p) of Three Methyl Ketones and Their Permeability (P), Diffusion (D), and Solubility (S) Coefficients through a Film of Polyethylene

	2-heptanone	2-octanone	2-nonanone
$p_{760\text{mmHg}}^{25^\circ\text{C}}$ (Pa)	470	129	39
P ($10^{-6} \mu\text{g m}^{-1} \text{s}^{-1} \text{Pa}^{-1}$) for $\Delta C < 10 \mu\text{g mL}^{-1}$	239.5	133.1	164.7
D ($10^{-13} \text{m}^2 \text{s}^{-1}$) for $\Delta C < 10 \mu\text{g mL}^{-1}$	4.5 ± 0.5	0.20 ± 0.03	0.22 ± 0.007
S ($10^6 \mu\text{g m}^{-3} \text{Pa}^{-1}$)	24.3	129.0	334.1

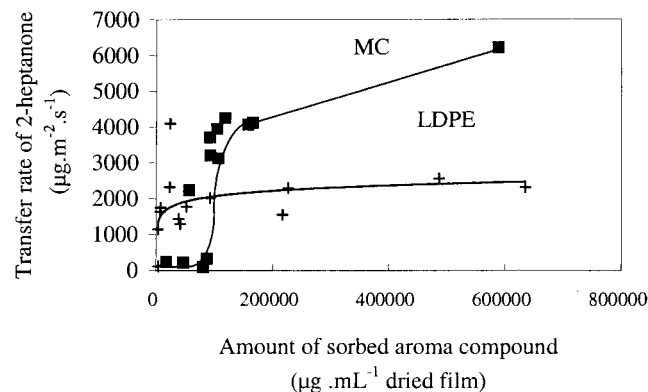


Figure 4. Transfer rate of 2-heptanone through a methylcellulose-based film and an LDPE film as a function of its sorption in the membrane at 25 °C.

product of solubility (S) and diffusivity (D) (Rogers, 1985). This model can be applied only in the absence of strong interactions between the volatile and the polymer, that is, for a constant diffusivity and a linear sorption isotherm. In practice, permeability and solubility coefficients are calculated from the equations

$$P = (F/\Delta p)e \quad (1)$$

$$S = Q/\Delta p \quad (2)$$

where F is the transfer rate ($\mu\text{g m}^{-2} \text{s}^{-1}$), Δp is the vapor partial pressure gradient, e is the film thickness (m), and Q is the quantity of volatile compound sorbed in the film ($\mu\text{g mL}^{-1}$).

The diffusion coefficient has been calculated with the half-time method, designating the time $t_{1/2}$ at which the transfer rate is equal to half of the transfer rate at the steady state obtained by a differential permeation method (Crank, 1975; Felder, 1978; Chao and Rizvi, 1988). The diffusion coefficient (D) was calculated following the eq 3:

$$D = e^2/7.199t_{1/2} \quad (3)$$

In Table 2 are reported the saturated vapor pressures of the methyl ketones (p) calculated from the ideal gas equation for an atmosphere at 25 °C and a pressure of 760 mmHg along with their permeability (P), diffusion (D), and solubility (S) coefficients in LDPE. No direct relationship between permeability and the other parameters was observed. Nevertheless, P varies in the same way as diffusivity (D). Because S has not been directly correlated with permeability, we can suppose that there is no physicochemical interaction between the polymer and the volatile compounds. Whatever the amount of methyl ketone sorbed in the film, the transfer rate remains constant (Figure 4).

(C) A third hypothesis could be a mechanical process for 2-heptanone. If the LDPE film is dense in a microscopical level, the film structure possesses a tridimensional network (Pascot, 1985). When this network is saturated with an aroma compound, the transfer rate does not increase with the concentration rate. In this case, only diffusion can have an effect on transfer at higher concentrations. Indeed, the diffusion coefficient of 2-heptanone is $(4.5 \pm 0.5) \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ for a concentration differential $< 10 \mu\text{g mL}^{-1}$, whereas D is $(2.0 \pm 0.2) \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ when the transfer rate decreases with an increasing concentration gradient ($\Delta C > 15 \mu\text{g mL}^{-1}$) (Figure 2). This phenomenon can be explained by a friction process between the molecules of 2-heptanone and the polymer network induced by the clustering effect occurring at high concentration of aroma compound. Indeed, Hernandez et al. (1992) showed that polar compounds at high activities may form clusters, which reduces the volatile diffusivity. Hernandez (1994) has reported a decrease in the water diffusivity in LDPE films when water concentration increases, and Debeaufort et al. (1994) revealed the same phenomenon in a methylcellulose-based film. These authors have explained this decrease in diffusivity by clusters formation. Otherwise, the permeability P for the three methyl ketones varies linearly with the logarithm of the diffusion coefficients:

$$P = 1.08 \times 10^{-3} + 29.7 \times 10^{-6} \ln D$$

$$(r^2 = 0.93 \text{ significant at } \alpha = 0.05) \quad (4)$$

The results show an unusual diffusion of 2-heptanone in LDPE film. The diffusion coefficient of this methyl ketone is 20 times higher than those of 2-octanone and 2-nonanone for the lower concentration differentials ($< 10 \mu\text{g mL}^{-1}$). As this behavior was observed for the smaller and the more volatile molecule, we could think that the size of the molecule would play an important role on mass transfer. Johansson and Leufvén (1994) found that permeability and diffusion do not always decrease when the carbon atom number of the volatile molecule increases. They studied the transfer of a group of aldehydes and alcohols (carbon atom number from 4 to 10) through a linear LDPE (LLDPE). For cross-linked natural rubber and organic vapors, Fujita (1968) reported the nonregular size dependence of D for hydrocarbons from 1 to 5 carbon atoms. In summary, the transfer of methyl ketones through LDPE displays a high effect of mechanical factors such as free volume of the membrane and molar volume of the volatile compound, the latter having an effect on the diffusion coefficient.

Transfer of Methyl Ketones through Methylcellulose-Based Films. Contrary to results obtained for LDPE, the transfer of 2-heptanone through methylcellulose-based films increases strongly for a concentration differential $> 10 \mu\text{g mL}^{-1}$. This phenomenon could be explained by a plasticization of polymer in the presence of 2-heptanone. Moreover, when ΔC is $> 15 \mu\text{g mL}^{-1}$, films become opaque and sticky. Indeed, the opacity and the swelling of polar polymers is typical of a plasticization mechanism, which results in a decrease in T_g of the polymer and in a reduction in its barrier properties (Ashley, 1985). This behavior has been observed by Donhowe and Fennema (1993) when plasticizer concentration exceeds the limit of compatibility between polymer and plasticizer.

Table 3. Vapor Partial Pressure (p) of Three Methyl Ketones and Their Permeability (P) and Solubility (S) Coefficients through a Film of Methylcellulose

	2-heptanone	2-octanone	2-nonanone
$P_{760\text{mmHg}}^{25^\circ\text{C}}$ (Pa)	470	129	39
P ($10^{-6} \mu\text{g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$)	39.2	338.0	420.8
for $\Delta C < 10 \mu\text{g mL}^{-1}$			
D ($10^{-13} \text{ m}^2 \text{ s}^{-1}$)	2.1 ± 0.5	0.7–4.1	1.1–8.7
for $\Delta C < 10 \mu\text{g mL}^{-1}$			
S ($10^6 \mu\text{g m}^{-3} \text{ Pa}^{-1}$)	139.3	599.4	1139.0

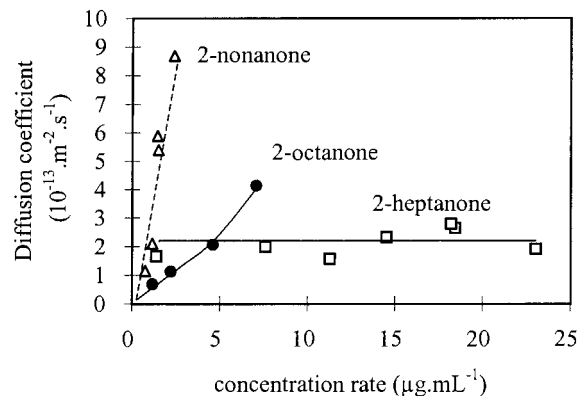


Figure 5. Diffusion coefficient of three methyl ketones through a methylcellulose-based film versus the concentration rate in the vapor phase at 25 °C.

In Table 3, relationships between sorption coefficients (S) or partial pressures (p) of volatile compounds and transfers can be observed in the presence of aroma-saturated atmosphere. These relationships are given in the following equations:

$$P = -3.434 \times 10^{-3} + 185.82 \times 10^{-6} \ln S$$

$$(r^2 = 0.99 \text{ significant at } \alpha = 0.05) \quad (5)$$

$$P = 0.453 \times 10^{-3} - 0.88 \times 10^{-6} p$$

$$(r^2 = 0.99 \text{ significant at } \alpha = 0.05) \quad (6)$$

Several authors have determined relationships between permeability of polymeric films and vapor pressure of volatile compounds such as aroma compounds (Shimoda et al., 1988; Arora et al., 1991). The transfer rate for 2-heptanone significantly increases with the amount of aroma compound sorbed (Figure 4). The affinity of volatile compound to the film seems to be the main factor governing the transfer, which relates physicochemical interactions between volatile compound and polymer. Inversely to the polyethylene film, methylcellulose-based films had 25% d.m. of PEG 400. This constituent allows one to establish weak interactions (hydrogen bonds, van der Waals interactions, hydrophobic interactions) with ketones, due to the polar groups (OH). Moreover, Debeaufort and Voilley (1994) reported that the transfer rate of 1-octen-3-ol through edible methylcellulose-PEG400 films increases with concentration in PEG 400. Donhowe and Fennema (1993) noted the same effect with the oxygen transfer through the same polymer. Otherwise, the diffusion coefficient for 2-heptanone remains constant [$(2.1 \pm 0.3) \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$] whatever the concentration differentials. However, the diffusivity of 2-octanone and 2-nonanone increases with aroma compound concentration in the film (Figure 5). The behavior of these two aroma compounds agrees with the report of Fujita (1968) stating that the concentration dependence of the diffu-

sion coefficient in the case of organic vapors must be taken into account as an additional variable to obtain *D*.

These results confirm that diffusivity of volatile compounds through methylcellulose-based films is not the main factor affecting transfer through methylcellulose films. The volatile compound with the shortest chain (2-heptanone) has the highest diffusivity and the smaller permeability. Moreover, several authors have shown that the diffusivity of volatile compounds in edible films depends not just on concentration when sorbant-polymer interactions exist, so the transfer has a non-Fickian behavior (Debeaufort et al., 1994; Habig McHugh et al., 1994).

CONCLUSION

This work has shown the transfer behavior of three methyl ketones through edible or plastic packagings. The transport mechanism depends on the polymer nature and concentration differential of the volatile compound; both polymers showed different barrier properties against the methyl ketone transfer.

In the case of LDPE films, transfer rates vary in the same way as diffusion coefficients for the three methyl ketones. However, diffusivities are not proportional to the chain length of the volatile compounds. The transfer of 2-heptanone through LDPE could be explained by a saturation of the polymer at concentration gradients $<10 \mu\text{g mL}^{-1}$ and by the cluster formation between 2-heptanone molecules.

Transfer of three methyl ketones through methylcellulose-based films is more complex than for LDPE. Their transfer rate increases with the chain length of volatile compound while their diffusion coefficient decreases. There is a relationship between permeability and sorption and between permeability and saturated vapor pressure. Mass transfer seems to depend mainly on the affinity between the volatile compound and the polymer. This relationship reveals the presence of physicochemical interactions that modify the structure of the polymer (plasticization). Confirmation of the polymer-aroma compound interactions affecting the film structure should be made by rheological and spectrometric studies.

Comparing the performances of both the plastic and the edible films studied, we observed that at concentrations of methyl ketones close to those of foodstuffs, edible films have better aroma barrier properties than LDPE films. The application of edible coatings or films to improve plastic packaging properties could be envisaged.

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