The Effect of Relative Humidity Gradient on Water Vapor Permeance of Lipid and Lipid-Hydrocolloid Bilayer Films

I. Greener Donhowe[1] **and O. Fennema***
Department of Food Science, University of Wisconsin, Madison, Wisconsin 53706

The water vapor (WV) permeance of lipid and lipid-hydrocolloid films exposed to relative humidity (RH) gradients of 100-0%, 100-50%, 100-65% and 100-80% RH were determined. The lipids used were beeswax (BW) or a blend of BW and acetylated monoglycerides (AG). Hydrocolloids used were methylcellulose, carboxymethylcellulose or ethylcellulose (EC). All films, except those containing EC, exhibited increased water vapor permeance as the RH gradient was reduced by raising the low-end RH. This increase in permeance was apparently caused by hydration and swelling across the entire film thickness, thus facilitating water movement through the film. Because of its hydrophobicity, EC likely lessened this swelling. Knowledge of the WV properties of edible films at relatively small gradients in the upper half of the RH spectrum, such as those used in this study, is useful because these conditions are far more common to foods than are the 100-0% gradients that are often used when evaluating films. Even though the WV permeance of BW and BW/AG films increased greatly at the 100-80% RH gradient, as compared to gradients ranging from 100-65% to 100-0%, they still possess WV barrier properties sufficient to be useful for foods.

KEY WORDS: Beeswax, films, hydration, hydrocolloids, permeability, permeance, relative humidity, water vapor.

Moisture migration will occur in foods when a water activity (a_W) differential exists within the food or between the food and its environment. During storage of foods, moisture migration is commonly associated with deleterious changes in quality. Katz and Labuza (1) determined that crackers, potato chips and popcorn, all with initial a_W in the range of 0.062–0.074, became unacceptable when their a_W increased to 0.39, 0.51 and 0.49, respectively. Similarly, the crusts of frozen pizzas and pies can become soggy during frozen storage because of moisture migration from the filling or sauce.

Moisture absorption from the environment can make some foods more susceptible to deteriorative chemical and enzymatic reactions. Some enzymatic reactions attain maximum reaction rates near an a_W of 0.7, while nonenzymatic browning attains a maximum reaction rate in the range of 0.75–0.85 a_W (2). Food safety can also become of concern if a_W is altered because microbial growth is strongly dependent on a_W.

Moisture interchange between a food product and its environment can be controlled in some instances by adjusting the a_W of the food. Lowering of a_W can be accomplished by the addition of humectants, such as sugars and salts or by drying. Alternately, a food can be enveloped in a film with good barrier properties to water vapor (WV) transmission, and this will effectively retard moisture loss.

To retard internal movement of water among food components, the a_W of the components can be equalized or an edible film or coating with good barrier properties to WV can be situated at the interface of the components.

The use of edible films to lessen internal migration of moisture in foods has been studied by several investigators. Some of these films have been shown to be effective barriers with considerable promise for commercial use (3–6). Lipid or lipid-hydrocolloid films are especially good barriers to water vapor (4–13). However, the performance of these films as influenced by variable water vapor gradients has received little attention.

Much of the research on WV barrier properties of films has involved exposing one side of the film to 0% relative humidity (RH), and the other to a high RH, usually 100%. Furthermore, it is commonly stated that the WV permeability of hydrophilic, but not hydrophobic, polymers is dependent on WV pressure. For example, Barrer (14) stated that pure rubbers and waxes absorb little water and that their permeabilities are little affected by variations in humidity. Supporting data were, however, not presented.

Sufficient work has been done to establish that the WV permeability obtained by testing a film at one RH gradient is not always a reliable indicator of what the WV permeability will be at another RH gradient. Several investigators have found that WV permeabilities of some edible films are significantly greater when tested at a 97-65% RH gradient than they are when tested at a 100-0% RH gradient (7,9,10). This is important because the former gradient is more likely to be encountered in foods than the latter.

In the present study, edible films were prepared as lipid-hydrocolloid (L-H) bilayers. The hydrocolloid film provided structural support for the lipid film and the lipid provided the WV barrier properties. Also, the hydrocolloid film provided a smooth surface, which facilitated formation of a lipid film with good integrity (5). The objectives were to determine the effect of RH gradient on the WV permeance of lipid and L-H films and to evaluate how the supporting hydrocolloid film affects WV permeance of L-H films.

EXPERIMENTAL PROCEDURES

(i) Preparation of L-H films. Types of hydrocolloids tested, the compositions of solutions used to prepare films, and the properties of the resulting dried films are listed in Table 1. Solutions of hydrocolloid films were prepared as follows:

Methylcellulose (MC). All of the MC (A15LV, Dow Chemical, Midland, MI) was suspended in half the final volume of water (90°C, twice distilled). The remaining amount of water (10°C) was incorporated, and the solution was cooled to 10–15°C. Air bubbles were removed by gentle mixing. A measured volume of solution was poured onto a level glass plate that had been previously cleaned with acetone. A plastic frame (17.5 cm × 17.5 cm) was used to confine the solution to 306 cm^2. MC films were allowed to dry overnight at ambient conditions.

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[1] Current address: Griffith Laboratories, 1 Griffith Center, Alsip, IL 60658.

*To whom correspondence should be addressed.

TABLE 1

Properties of Hydrocolloid Films and the Solutions from Which They Are Prepared

Film solution				Properties of dried film	
Product	Hydrocolloid (% wt/vol)	Solvent (%)	Volume poured (mL/306 cm^2)	Thickness[a] (mm) [mil]	Film density[a] (mg/cm^2)
Methylcellulose	4.5	Water, 100 Ethanol, 0	35.0	0.038 ± 0.005 [1.5 ± 0.2] A	3.6 ± 0.3 A
Carboxymethylcellulose	4.0	Water, 85 Ethanol, 15	27.0	0.043 ± 0.003 [1.7 ± 0.1] B	3.7 ± 0.3 A
Ethylcellulose	3.5	Water, 0 Ethanol, 100	40.0	0.051 ± 0.005 [2.0 ± 0.2] C	3.4 ± 0.2 A

[a]Mean ± SD. Values followed by identical letters do not differ significantly at the $\alpha = 0.05$ level.

Sodium carboxymethylcellulose (CMC). CMC (7 JF, Aqualon, Wilmington, DE) was dispersed in aqueous ethanol (water, twice distilled; ethanol, 100%) and agitated until solution was achieved. After removal of air bubbles by gentle mixing, the solution was spread on a glass plate and dried as described for MC films.

Ethylcellulose (EC). EC (Standard 45, Dow Chemical) was dispersed in ethanol (100%) at 50°C and stirred until the EC dissolved. The EC film was formed and dried as described previously. When the RH of the room was greater than 65%, the films were dried for 30 min in a vacuum oven at 30 mm Hg gauge pressure and ambient temperature. There was no noticeable difference in the properties of films dried by the two procedures.

Lipid layer. Two lipid types were used in L-H films. Beeswax (BW, Select Australian white beeswax, National Wax, Skokie, IL) was chosen because of its excellent barrier properties to WV, good film-forming ability, low cost, and because it can be legally added to some foods. A blend of 80% BW and 20% acetylated monoglycerides (AG, Myvacet 5-07, Eastman Chemical Products, Kingsport, TN), referred to hereafter as BW/AG, was selected because it has, for food film applications, properties that are generally better, except for moisture barrier properties, than those of BW alone. Addition of AG to BW reduces its melting point and tensile strength, both of which can lessen the likelihood of sensory detection in heated foods.

Both BW and BW/AG were heated to 100°C before being applied to the hydrocolloid film. Molten lipid (2 mL) was deposited in a thin line adjacent to the edge of the dried hydrocolloid film and spread evenly over the surface with a prewarmed thin-layer chromatography spreader. This method deposited 4.0 ± 0.4 mg lipid/cm^2 film, resulting in a lipid layer of 0.046 mm (1.8 mil) thickness and a total film thickness of 0.084–0.097 mm (3.2–3.8 mil).

Preparation of pure lipid films. To prepare pure lipid films, lipid-MC films were first prepared as described. The hydrocolloid layer was then removed by soaking the film in warm water (30–35°C). After 3–4 min, the MC layer was washed off, and the lipid film was blotted dry with towels and more completely dried overnight under vacuum at room temperature. Samples were stored in a desiccator containing $CaSO_4$ prior to testing.

(ii) Determination of moisture absorption. A modified TAPPI method (15) was used to determine the amount of water absorbed by hydrocolloid films at various RH. Hydrocolloid film samples (7 cm × 7 cm) were held at 25 ± 0.5°C and exposed to 50, 65 or 80% RH for 48 h before hydrated weight was determined. Hydrated samples were then placed in a convection oven at 105°C for 2 h, and moisture content was calculated. Four replicates of each hydrocolloid type were tested at each RH.

Pure BW films, prepared and dried according to the method just described, were weighed and placed in small, air-tight containers at 30, 50, 65 or 80% RH and 22–23°C. Constant RH was achieved in the containers by using appropriate concentrations of glycerol in water (16). BW samples were removed periodically to measure weight gain and were stored until a constant weight was achieved. Unless otherwise noted, four replicates were tested at each RH.

(iii) Gravimetric water vapor permeance. Film samples were sealed to glass cups in accordance with the ASTM standard method for WV transmission (17). Each L-H film sample was affixed (lipid side down) to the rim of a glass cup with a blend of molten paraffin and microcrystalline wax. Distilled water was added to the cup through a hole in its side. The final water level inside the cup was 1.3 cm (0.5 in) below the lower surface of the film. The hole was then sealed with the paraffin-microcrystalline wax. The film area exposed for WV transmission was 33 cm^2.

Pure lipid films could not be sealed by the method described because the molten sealing wax melted the film. Therefore, a 38-cm^2 disk of lipid film was cut and placed between two rings of tape (Transmask S polyvinyl chloride tape, MacTac, Stow, OH). A 20-cm^2 circular area was cut from the center of each piece of tape, leaving a ring with an outer diameter of 6.7 cm and inner diameter of 5.0 cm (Fig. 1). The vinyl-lipid-vinyl film was then sealed to the cup in the same manner previously mentioned. This technique proved to be satisfactory.

Film samples sealed to the glass cups were then placed in a room maintained at 50, 65 or 80 ± 2% RH and 25 ± 0.5°C. A balance, with a precision of ±0.0001 g, was used to measure weight losses from the cups. Film samples were weighed 4–5 times over a 24-h period. Correlation coefficients for weight loss *vs.* time were 0.99 or greater.

For testing at a 100-0% RH gradient, samples were prepared as previously described, then placed in desiccators containing calcium sulphate (Drierite, W.A. Hammond Drierite Co., Xenia, OH) to maintain a 0% RH environment. During these measurements, the room was maintained at 50% RH, the lowest controllable RH. Exposure

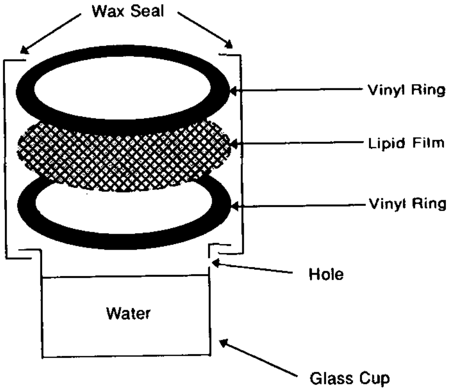

FIG. 1. Schematic view showing how lipid film was sealed to a cup with the aid of vinyl tape.

time of film samples during weighing was minimal (less than 30 s). WV transmission through these samples was determined by periodic weighing.

WV permeance of the pure lipid films was determined by considering the 13-cm^2 ring and the 20 cm^2 of pure lipid as resistances in parallel. With that assumption, the following equation applies:

$$J_t = J_r + J_l \qquad [1]$$

where J_t is the flux of WV through all materials, J_r is the flux of WV through the 13-cm^2 ring, and J_l is the flux of WV through the 20-cm^2 film (18). To determine the amount of WV passing through the ring, samples were prepared as described except the 20-cm^2 area was not removed from the vinyl tape. The value obtained, adjusted to a basis of 13 cm^2, was then subtracted from values of total weight loss obtained from samples tested as shown in Figure 1. This resulted in a value for WV transmission through the lipid film only.

Unless otherwise noted, at least five replicates were tested for each sample at a given condition.

(iv) Automated water vapor permeance. A Permatran W-1A (Mocon Instruments, Minneapolis, MN) was used to determine WV transmission values for films that could not be sealed effectively to the glass cups used for the gravimetric method. For example, when L-H films were tested by the gravimetric method in a 100-0% RH gradient, the seals failed.

At least four replicates of EC, MC and CMC L-H films were tested at a 100-0% RH gradient, a temperature of 25°C and an air flow of 20 mL/min across the dry (0% RH) side of the film. The lipid surface of the film was always positioned toward the high RH side. Stainless steel masks were used to decrease the transmission area of the film to 20 cm^2. Film samples were equilibrated overnight in the Permatran Conditioning Rack (Mocon Instruments, Minneapolis, MN) prior to determining WV transmission.

(v) Hydrocolloid film densities. Densities of dried hydrocolloid films were calculated from the weights of samples cut to an area of 49 cm^2. Reported densities are means of at least eight measurements.

(vi) Amount of lipid deposited. The amount of lipid deposited on the hydrocolloid film was determined by subtracting the mean weight of the hydrocolloid film (7 cm × 7 cm) from the final weight of the bilayer film (7 cm × 7 cm).

(vii) Film thickness. A micrometer was used to measure film thickness. Reported thickness values are means of at least eight measurements.

(viii) Calculations of water vapor permeance. WV permeance of L-H films and pure lipid films was calculated by the following equation:

$$\text{permeance} = J/(C_i - C_a) = \text{m} \cdot \text{s}^{-1} \qquad [2]$$

where C_i is WV concentration inside the cup in $\text{g} \cdot \text{m}^{-3}$, C_a is WV concentration of the room in $\text{g} \cdot \text{m}^{-3}$, and J is WV transmission through the film in $\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$. J was determined by the gravimetric cup method or with the Permatran.

Permeance values have been used to describe WV transmission through heterogeneous barriers and membranes (8,19–22). Use of the standard permeability expression [with units of $\text{g}(\text{m} \cdot \text{s} \cdot \text{Pa})^{-1}$] for composite films is considered inappropriate because total film thickness does not accurately reflect the barrier properties of the individual film components. Because film thickness is disregarded in the permeance expression, this expression is especially well suited for use with composite films and coatings, such as the L-H films in this study, provided film thickness is reasonably consistent among samples being compared (23).

(ix) Statistical analysis. One-way ANOVAs (SAS® Statistical Package, SAS Institute, Inc., Cary, NC) were calculated for permeance data at each humidity gradient. When the one-way ANOVA indicated a significant (α = 0.05) difference among permeance means, a least significant difference (LSD) test was used to identify specific differences.

One-way ANOVA and LSD test were also used to identify significant differences among moisture contents of BW films, as well as to evaluate statistical significance among thicknesses and densities of hydrocolloid films.

RESULTS

(i) Film thicknesses and film densities. Significant differences (α = 0.05) existed among thicknesses of the three hydrocolloid films (Table 1), with the thickness of the EC films being greatest. This occurred because the EC film developed a rippled character during drying. When thickness was measured, the crests in the film surface caused the measured thickness to be artificially large. Surfaces of the MC and CMC films were smooth.

(ii) Moisture absorbed by hydrocolloids films. The amount of water absorbed by the hydrocolloid films at various RHs is shown in Figure 2. Differences in the moisture contents of the hydrocolloid films at 50, 65 and 80% RH are the result of differences in substituent groups on the anhydroglucose units of the cellulose molecule.

In CMC, an average of 0.7 of the three hydroxyl hydrogens on each anhydroglucose unit has been replaced by carboxymethyl groups ("degree of substitution" [DS] is 0.7). Of the three hydrocolloids used in this study, CMC had the largest number of hydrophilic substituent groups per anhydroglucose unit.

The moisture content of MC was less affected by RH than that of CMC. The methyl groups on MC are more hydrophobic than the carboxymethyl groups of CMC and would, therefore, decrease MC's affinity for moisture. In

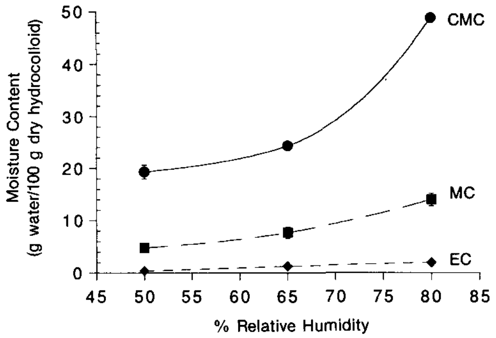

FIG. 2. Effect of relative humidity on moisture content of hydrocolloid films. Each datum point is a mean of four replicates. When standard deviation bars are not shown, the standard deviation is less than the size of the symbol.

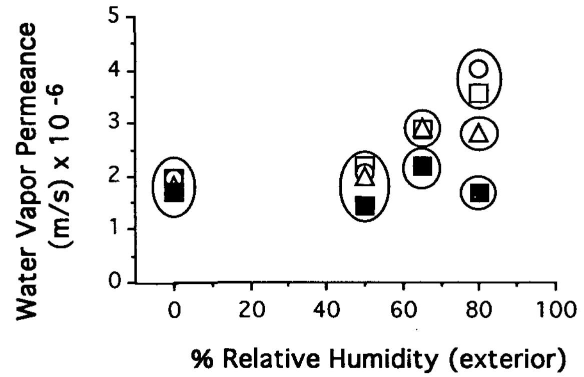

FIG. 4. Effect of relative humidity (RH) gradient on water vapor (WV) permeance of beeswax (BW) and beeswax-hydrocolloid films at 25°C. RH inside the cup was 100% and the lipid surface of the L-H (lipid-hydrocolloid) film was in contact with this RH. Means located in the same ellipse do not differ significantly ($\alpha = 0.05$). The permeance value for BW-CMC (carboxymethyl cellulose) at the 65% exterior RH was identical to that of the BW-EC (ethylcellulose) film at that same RH. Each datum point is a mean of at least five replicate except for BW-EC (100-0% RH) and BW-CMC (100-0% RH), which are means of four. □, BW; ■, BW-EC; △, BW-MC (methylcellulose); ○, BW-CMC.

addition, the DS for MC, 1.6–1.7, is greater than that of CMC.

The moisture content of EC was the smallest of the hydrocolloids at each RH and was least affected by changes in RH. Of the three hydrocolloids tested, EC had the largest DS, 2.47–2.58, and the substituents are quite hydrophobic.

Because the moisture contents shown in Figure 2 were determined from a single weighing of each film sample (four replicates), after it had been stored for 2 d at each RH, no claim is made that the values are equilibrium values. However, the moisture contents reported for EC are in good agreement with those reported by Wellons and Stannet (24).

(iii) Moisture absorbed by BW films. The amount of water absorbed by BW films at various RHs is shown in Figure 3. The moisture content of BW films at 80% RH was significantly ($\alpha = 0.05$) greater than that of films stored at the other RHs.

(iv) Water vapor permeance of films. With BW and BW-hydrocolloid films, significant differences ($\alpha = 0.05$) among permeance values of the films at a given RH gradient did not occur until the exterior RH was increased to 65% (Fig. 4). These differences became more pronounced at the 100-80% RH gradient, with BW-EC being the least permeable and BW-CMC the most.

With BW/AG films, few significant differences in permeance occurred at the 100-0% RH gradient, none at the 100-50% gradient, and many at the 100-65% and 100-80% RH gradients (Fig. 5). The pattern of differences in permeance at the 100-80% RH gradient was the same for the BW and BW/AG films.

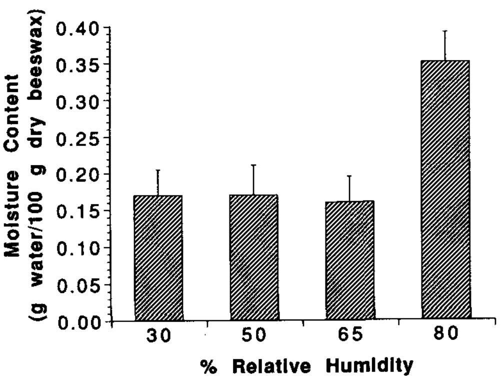

FIG. 3. Effect of relative humidity (RH) on moisture content of beeswax at 22–23°C. Each datum point is a mean of four replicates except points at 50% and 80% RH, which are means of three. Error bars represent standard deviations.

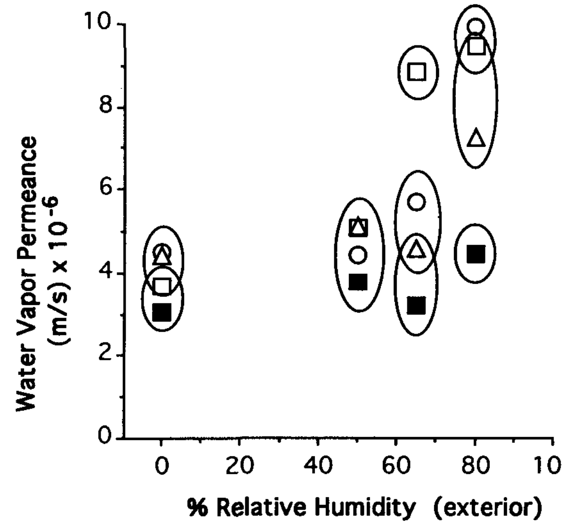

FIG. 5. Effect of relative humidity gradient on WV permeance of beeswax/acetylated monoglyceride and beeswax/acetylated monoglyceride-hydrocolloid films at 25°C. The RH inside the cup was 100% and the lipid side of the L-H film was in contact with this RH. Means located in the same ellipse do not differ significantly ($\alpha = 0.05$). Each datum point is a mean of at least five replicates except for BW/AG and BW/AG-CMC (100-0% RH), which are the means of four. □, BW/AG; ○, BW/AG-CMC; △, BW/AG-MC; ■, BW/AG-EC. Abbreviations as in Figure 4.

Differences in permeance of a given film as a function of RH gradient could not be statistically analyzed because the gradients were not replicated. However, data in Figures 4 and 5 appear to indicate a trend of increasing WV permeance with increasing exterior RH for all films except for those containing EC.

DISCUSSION

In this study, lipid and L-H films were tested at RH gradients of 100-0%, 100-50%, 100-65% and 100-80%. All of these gradients, with exception of the 100-0% RH gradient, are reasonably common in foods.

It should be noted that all constituents used in this study, with the exception of EC, can be legally added to foods, although use restrictions apply in the case of BW (25,26).

Pure lipid films. Literature values pertaining to the WV permeance of pure BW were not found. The study most analogous to ours is that of Kester and Fennema (27). They studied BW mounted on filter paper and found its WV permeance to be 4.69×10^{-6} $m \cdot s^{-1}$ as compared to our value of 1.95×10^{-6} $m \cdot {}^{-1}$. Both of these values were obtained in a 100-0% RH gradient. A difference is understandable because the BW of Kester and Fennema (28) was partially embedded in filter paper, thereby increasing WV permeance of the total sample.

The WV permeance of BW tested at the 100-80% RH gradient is approximately 83% greater than that at the 100-0% RH gradient. This trend was observed previously in this laboratory with L-H films (7,9,10). These earlier results with the L-H films were attributed to hydration and swelling of the hydrocolloid film, with accompanying fractures of the adhering lipid layer. Obviously, this theory is not relevant for films of pure lipids.

Incorporation of 20% AG in BW resulted in a WV permeance of 3.69×10^{-6} $m \cdot s^{-1}$, an 89% increase as compared to the permeance of a pure BW film (100-0% RH). WV permeance values for BW/AG blends were not found in the literature.

The WV permeance of BW/AG films, like that of BW films, was dependent on RH gradient and especially on location of the gradient in the RH spectrum. It is likely that the same mechanism responsible for the increase in WV permeance of BW films at RH gradients located at the high end of the RH spectrum is responsible for the increased permeance of BW/AG films under the same circumstances. However, the increase that occurred in WV permeance of BW/AG films when the RH on the dry side was increased from 0 to 80% was much greater than that observed with BW films. Under these circumstances, permeance of the BW/AG film increased 257%, whereas that of the BW film increased only 83%. This difference was attributed to the larger proportion of polar components in BW/AG than in BW.

Other studies involving barrier properties of lipids at variable RH gradients have yielded diverse results. Lovegren and Feuge (29) found that WV permeability of AG films increased from 44.5×10^{-12} $g(m \cdot s \cdot Pa)^{-1}$ at a 97-0% RH gradient (21.1°C) to 105×10^{-12} $g(m \cdot s \cdot Pa)^{-1}$ at a 100-75% RH (21.1°C). They attributed the increase to the high concentration of water in the films at elevated RH.

Landmann *et al.* (30), however, found no difference in the WV permeability of fully hydrogenated coconut seed oil when tested at RH gradients of 75.4-22.5% (26.7°C) and 100-0% (26.7°C). Failure to detect a difference in WV permeability at the two RH gradients may have resulted because of the low WV permeability of this lipid or the insensitivity of the procedure used.

Still another result was achieved by Landmann *et al.* (30) in their study of WV permeability of a sweet milk chocolate coating. They observed an increase in WV permeability from 10.7×10^{-12} $g(m \cdot s \cdot Pa)^{-1}$ at 76.4-22.5% RH to 892×10^{-12} $g(m \cdot s \cdot Pa)^{-1}$ at 100-0% RH (26.7°C). The large increase in permeability at the 100-0% RH gradient was attributed to absorption of water by hydrophilic constituents of the coating (protein, milk solids and sugar), and destruction of film integrity. Both of Landmann's studies involved a dry-side RH that was well below the 65% RH (100-80% or 100-65%) needed in our studies to produce lessening of the WV barrier properties of the lipids studied.

Hydrophilic films become increasingly permeable as the RH to which they are exposed is increased (31). According to Rouse (32), this behavior is attributed to swelling and plasticization of sorbed water in the hydrophilic films at high RHs. As a hydrophilic film absorbs water and swells, the newly added water molecules are presumably more mobile, causing the average mobility of the water molecules to increase, and permeability to increase as well (32).

One might suspect that BW would not mimic the behavior of hydrocolloids at elevated RH gradients because of BW's hydrophobic nature. BW consists of 71% wax esters, 1-1.25% free alcohols, 13.5-14.5% free fatty acids, 10.4-13.6% hydrocarbons, 1-2% moisture, and foreign matter (33). But it is not totally devoid of hydrophilic groups. The ester groups in the wax are hydrophilic as are the hydroxyl groups of the free alcohols and the carboxyl groups of the free fatty acids. Although the polar groups of fatty acids and fatty alcohols in plant waxes are sandwiched between aliphatic chains (34), at high RH they may be able to hydrate sufficiently to influence WV permeability. This possibility is substantiated by the significant ($\alpha = 0.05$) increase in moisture absorbed by BW at 80% RH, as compared to that absorbed at lower RHs (Fig. 3). This increase in hydration would be expected to cause matrix swelling, increased water mobility and an increase in the diffusion coefficient.

Higuchi and Aguiar (35) stated that the internal concentration gradient of water in a wax barrier exposed to a differential WV pressure is probably not linear, as might be expected, but is instead sigmoidal. Thus, with BW it is reasonable to schematically depict gradients as shown in Figure 6. When the external gradient is 100-0% RH (Fig. 6A), the dry side of the film would be low in water content and unswollen. As the dry-side RH is elevated (Fig. 6B), the moisture content in the dry side of the film would rise, and the matrix in this area would become progressively more swollen. If the depiction in Figure 6 is valid, an external gradient of 100-80% RH would cause the matrix to be swollen across the entire film, and the water in this swollen area would have enhanced mobility. This depiction is in accord with the observed increase in the water content of BW films when exposed to 80% RH (Fig. 3). The result of a swollen matrix would be increased permeability, and the mechanism would be the same as that exhibited by hydrophilic films except on a reduced scale.

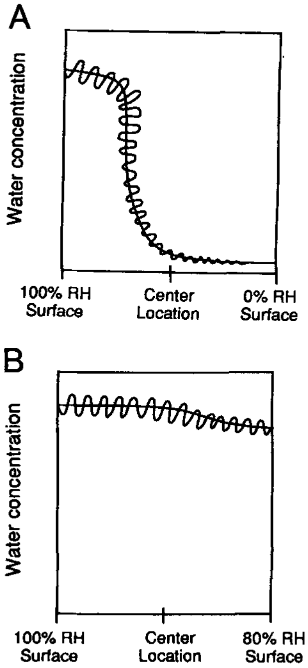

FIG. 6. Hypothetical water concentration gradients in a hydrophobic film containing hydrophilic groups (such as beeswax) at two external relative humidity (RH) gradients. Height of the serpentine lines depict the relative degrees of matrix swelling. A = 100-0% RH gradient; B = 100-80% RH gradient.

Lipid-hydrocolloid (L-H) films. With respect to WV permeance of BW-H films, the only other literature values are those from this laboratory. Those values are 2.98 $\times$ 10^{-6} m$\cdot$sec^{-1} (3.2 mg BW/cm^2) reported by Greener and Fennema (10) and 1.58 $\times$ 10–6 m$\cdot$s^{-1} (4.0 mg BW/cm^2 film and 0.65 mg fatty acid/cm^2 film) reported by Kester and Fennema (9). These small discrepancies in permeance values can be attributed to differences in the fabrication procedure and thickness of the lipid layer.

The WV permeance of L-H films, with the exception of those containing EC, followed the same trend as that of the pure lipid films—increasing WV permeance as the RH gradient was altered from 100-0% RH to 100-80% RH.

It is interesting that L-H films prepared from EC behaved much differently than did films prepared from the other hydrocolloids. When the dry-side RH was raised (Figs. 4 and 5), permeance was independent of RH gradient only when EC was used. Although EC is not a good barrier to WV transmission when compared to BW, it is a much better barrier than MC or CMC. At the 100-0% RH gradient, the WV permeability of EC is 4.9 $\times$ 10^{-12} g(m$\cdot$s$\cdot$Pa)$^{-1}$ (24,36), that of MC is 50 $\times$ 10^{-12} g(m$\cdot$s$\cdot$Pa)$^{-1}$ (37), and that of CMC is still greater. Also, as shown in Figure 2, EC has little ability to absorb water as compared to the other two cellulose ethers. Therefore, at high humidities, the mobile water phase present in EC would be much smaller than that existing in the other hydrocolloids. Also, water present may have limited mobility because it has been reported to exist as clusters in EC films, a state that lessens diffusion (24).

Because of MC's and CMC's hydrophilicity, the concentration gradient across the lipid component of the BW-MC, BW/AG-MC, BW-CMC, and BW/AG-CMC films at 100-80% and 100-65% RH presumably would be similar to those for the pure lipid films (Fig. 6B). However, substitution of EC for MC or CMC would lessen water absorption and swelling in the L-H film. As a result, the water gradient for the lipid-EC films at 100-65% and 100-80% RH gradients probably would be intermediate in character between the two gradients in Figure 6.

There was no significant ($\alpha = 0.05$) difference in the WV permeance of the BW-H and BW films when tested at the 100-0% RH gradient (Fig. 4). This is not surprising because the WV permeance of the cellulosic materials is large in comparison to BW. The WV permeance of BW/AG-EC films was significantly ($\alpha = 0.05$) less than those of the BW/AG-MC and BW/AG-CMC films at the 100-80 RH gradient (Fig. 5). This slight difference is probably attributable to the additional barrier properties of EC in the BW/AG-EC film.

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