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Water vapor transport parameters of a cast wheat gluten film

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Abstract

Understanding the mode of transport of water vapor through the film is important for improving the moisture barrier properties of wheat gluten (WG) films. Effective permeability (P_{eff}), solubility (S_{eff}), and diffusion (D_{eff}) coefficients of a hydrophilic cast WG film were determined at 25°C within the relative humidity (RH) range of 0–84% (with a 9–13% RH gradient between upstream and downstream water vapor flux). P_{eff}, S_{eff}, and D_{eff} increased substantially as the RH gradient moved upwards in the RH spectrum. P_{eff} increased by four orders of magnitude from the lowest RH condition of 0–11% (3.8×10^{-11} g·m/m^2·s·Pa) to the highest RH condition of 75–84% (4.1×10^{-7} g·m/m^2·s·Pa). A moisture sorption isotherm of the film at 25°C was constructed. Both the Guggenheim–Anderson–DeBoer (GAB) and the Kuhn moisture sorption isotherm models showed a good fit to the experimental adsorption data. Testing of WG films at the expected conditions of actual use is necessary to quantify the water vapor permeation through the films.

Keywords: diffusion, permeability, protein films, sorption isotherm, wheat gluten

1. Introduction

Concerns over solid packaging waste have increased interest in biopolymer films and coatings. Such films, comprised of protein, polysaccharide, and/or lipid materials, are renewable, possibly edible, and can lead to innovative packaging applications. Due to its unique cohesive and elastic properties, wheat gluten (WG) has been studied as a film former (Gennadios et al., 1994a). WG

films are effective oxygen barriers, but poor water vapor barriers (Gennadios et al., 1994a). Limited improvements in water vapor barrier properties of WG films have been achieved (Gennadios et al., 1993; Gontard et al., 1994; Ali et al., 1997). The poor resistance of WG films to water vapor is due to the hydrophilic nature of the protein and to the substantial amount of hydrophilic plasticizer added to impart adequate film flexibility. Understanding the mode of transport of water vapor through the film is important for improving the moisture barrier properties of WG films.

Water vapor transport through polymer films proceeds through: (i) absorption of water vapor on to the polymer surface; (ii) solution of water vapor into the polymer matrix; (iii) diffusion of water vapor through the polymer; and (iv) desorption of water vapor from the other surface of the polymer (Debeaufort et al., 1994). Water vapor permeability (P) is defined as (Rogers, 1985):

$$P = D \cdot S \tag{1}$$

where P is permeability coefficient ($g \cdot m/m^2 \cdot s \cdot Pa$); D is diffusion coefficient (m^2/day); and S is solubility or sorption coefficient ($g/m^3 \cdot Pa$).

Ideally, when no interaction occurs between a polymer film and the permeating water vapor, P is independent of the apparent equilibrium water vapor pressure corresponding to the water activity (a_w) of the film (Ashley, 1985). Hydrophobic films, such as polyethylene, have water vapor permeabilities independent of the water vapor pressure (Myers et al., 1962). However, permeation of water vapor through hydrophilic films deviates substantially from the ideal behavior. Protein-based films, similar to other hydrophilic films, exhibit water vapor pressure-dependent permeability (Ashley, 1985). Water vapor permeability measurements of WG films have been reported (Aydt et al., 1991; Gontard et al., 1992; Gennadios et al., 1994b; Herald et al., 1995; Park and Chinnan, 1995). However, such water vapor permeability measurements were limited to one or two relative humidity (RH) gradient conditions and these values cannot be used to predict transport properties of these films at different RH gradient conditions. Water vapor transmission rates (WVTR) of WG films determined by Gontard et al. (1993) in the range of 10–90% RH at a RH gradient of 10 ± 1% across films, showed the dependence of P on RH. Schwartzberg (1986) observed that the failure to account for air resistance across the upstream and downstream surfaces of hydrophilic films leads to a substantial underestimation of the film diffusion coefficient. Gontard et al. (1993) did not separate upstream and downstream air resistances from that of the WG film itself and did not determine the film diffusion coefficients. Our study was aimed at determining the effect of moisture concentration on water vapor transport parameters (permeability, solubility, and diffusion coefficients) of a cast WG film.

2. Materials and methods

2.1. Reagents

Films were prepared using vital WG (Wheat-Pro-80®, Ogilvie Mills, Quebec, Canada) with an approximate protein content of 80% (dry basis); ethyl alcohol, 95% (v/v) (J.T. Baker, Phillipsburg, NJ); ammonium hydroxide, 5 N (Aldrich, Milwaukee, WI); and glycerol (Fisher Scientific, Fair Lawn, NJ). Salts used to create different RH conditions (Table 1) were anhydrous calcium sulfate (W.A. Hammond Drierite, Xenia, OH), lithium chloride, potassium acetate, magnesium chloride, potassium carbonate, magnesium nitrate, sodium bromide, strontium chloride, sodium chloride, and potassium chloride (reagent grade, Mallinkrodt, Paris, KY).

Table 1. Relative humidity (RH) gradients created at 25°C by anhydrous calcium sulfate and various saturated salt solutions (Wolf et al., 1984) for the determination of water vapor permeability

Salt inside cups	Salt outside cups	RH gradient (%)
$CaSO_4$, anhydrous	LiCl	0.00–11.15
LiCl	CH_3COOK	11.15–22.60
CH_3COOK	$MgCl_2$	22.60–32.73
$MgCl_2$	K_2CO_3	32.73–43.80
K_2CO_3	$Mg(NO_3)_2$	43.80–52.86
NaBr	$SrCl_2$	57.70–70.83
NaCl	KCl	75.32–84.32

2.2. Film preparation

The film preparation method described by Gennadios et al. (1993) was followed. WG (15 g) was added to 95% ethanol (72 ml) and glycerine (6 g). WG dispersion was facilitated by adding 14 ml of 5 N ammonium hydroxide and 48 ml of distilled water, while warming and stirring on a magnetic stirrer/hot plate. A notable decrease in viscosity marked WG dispersion. The suspensions were heated to 75°C and then cooled to 45°C, strained through cheese cloth to remove any small lumps, and cast on level flat glass plates with a thin-layer chromatography spreader (Brinkman, New York, NY). Castings were kept at ambient temperature for ≈20 h before the films were peeled off the plates.

2.3. Film thickness and density

Thickness of dried films was measured to the nearest 2.54 µm (0.1 mil) with a hand-held micrometer (B.C. Ames, Waltham, MA). Five measurements were taken on each specimen and their mean was used in calculations. Densities were determined by weighing the film specimens (3×3 cm) after drying in a desiccator over anhydrous calcium sulfate (0% RH) for 7–10 days. Films were considered dry when constant weight was recorded between two consecutive days of weighing. Film specimens had a mean thickness of 83±2 µm. Density was determined by dividing film weight by film volume. A mean dry film density value of 0.925±0.038 g/m^3 was obtained.

2.4. Determination of water vapor permeability

Seven different RH gradients (Table 1) were used to study the water vapor permeation through films. Permeability was measured gravimetrically using a variation of the method described by Gennadios et al. (1994c). Test cups consisted of a cylindrical well (2.1 cm in depth) bored in a polymethylmethacrylate cylinder and a lid (with an opening in the center) of the same material. Both the cup well and the lid openings were 4.6 cm in diameter. The cups were filled with a saturated salt solution to 0.6 cm below the brim of the well. Film specimens were placed on top of the wells and secured beneath the lids by four screws, symmetrically placed around the lid perimeter. To ensure airtight sealing, high vacuum silicone grease was applied around the edge of the wells and under the lids.

The cups were placed on a weighing balance inside a sorbostat. The sorbostat was maintained at a lower RH than that in the cups (Table 1). The downstream resistance of air (Schwartzberg, 1986) was minimized by operating a fan inside the chamber to provide an airflow of 259 m/min over the cups. McHugh et al. (1993) observed an increase in water vapor permeability of hydrophilic films with an increase in air flow rate up to 152 m/min, after which the increase was negligible. The rate of weight loss, which was constant at steady state, was used to calculate P. When Henry's law is obeyed, the loss rate or WVTR is (Rogers, 1985):

$$\mathrm{WVTR} = DS(p_{W1} - p_{W2})/l \qquad (2)$$

where WVTR is water vapor transmission rate (g/m^2·s); p_{W1} is partial pressure of water vapor at the underside of the film (Pa); p_{W2} is partial pressure of water vapor at the film surface outside the cup (Pa); and l is film thickness (µm).

Partial pressure of water vapor at the underside of the film (p_{W1}) was calculated from (Gennadios et al., 1994c):

$$p_{W1} = p_T - (p_T - p_{W0}) \cdot \exp[(R \cdot T \cdot \mathrm{WVTR} \cdot h_i)/(p_T \cdot D_{air})] \qquad (3)$$

where p_T is total atmospheric pressure (Pa); p_{W0} is partial pressure of water vapor in air at the surface of distilled water or saturated salt solution in the cup (Pa); R is universal gas constant (8,306,600 Pa·cm^3/gmol·K); T is absolute temperature during testing (K); h_i is the gap between the film underside and the surface of saturated salt solution in the cup (cm); and D_{air} is the diffusion coefficient of water vapor in air (cm^2/s), which is 0.26 cm^2/s at 25°C.

Equations (1) and (2) were combined to obtain Equation (4):

$$P = \text{WVTR} \cdot l/(p_{w1} - p_{w2}) \quad (4)$$

Due to the hydrophilic nature of films, both D and S vary with concentration and pressure gradient. Therefore, P also depends on those variables. Thus, P is replaced by P_{eff} (effective water vapor permeability coefficient in g/m·s·Pa) in Equation (4), and Equation (1) becomes:

$$P_{eff} = D_{eff} \cdot S_{eff} \quad (5)$$

where D_{eff} is effective diffusion coefficient (m^2/day) and S_{eff} is effective solubility coefficient (g/m^3·Pa).

2.5. Determination of moisture adsorption isotherm

Prior to the moisture adsorption experiments, film specimens (3×3 cm) were brought to zero moisture content by equilibrating them over anhydrous calcium sulfate (0% RH) in a desiccator. The samples were weighed daily and equilibrium was assumed to have been reached when the weight change of samples between two consecutive days was less than 0.001 g water/g of dry matter (Gennadios and Weller, 1994). Equilibrium was observed within 10 days. The desorbed samples were weighed into aluminum dishes and suspended inside glass sorbostats containing saturated salt solutions. Prior to introducing the samples, the targeted RHs of the sorbostats were verified using a thermohygrometer (model HI 8564, Hanna Instruments, Woonsocket, RI). Sorbostats containing saturated salt solutions of LiCl, CH_3COOK, $MgCl_2$, K_2CO_3, $Mg(NO_3)_2$, $SrCl_2$, NaCl, or KCl had equilibrium RH values close to 11, 23, 33, 44, 53, 71, 75, or 84%, respectively (Wolf et al., 1984). The samples reached equilibrium within 9 days (as observed by a weight change of less than 0.001 g water/g of dry matter in two consecutive daily weighings). The sorption study was replicated three times. Experimental water adsorption data were fitted with six different sorption isotherm models (Table 2). The parameters of the models were estimated with the nonlinear regression (NLIN) procedure in SAS (1993) software. Accuracy of fit was evaluated by the mean of the relative percent difference between the experimental and predicted values of the moisture content or mean relative deviation modulus (G) defined as (Gencturk et al., 1986):

$$G = (100/n) \cdot \sum(|M_a - M_p|/M_a) \quad (6)$$

where n is number of observations; M_a is experimentally determined moisture content (g/g dry solids); and M_p is predicted moisture content (g/g dry solids).

A G value lower than 5 corresponds to extremely good fit, a G value between 5 and 10 shows a reasonably good fit, and a G value greater than 10 is considered a poor fit (Gencturk et al., 1986).

2.6. Calculation of solubility and diffusion coefficients

The fitted GAB equation was used to estimate the moisture content of each film specimen surface in the permeability study. The S_{eff} value for each pressure gradient was obtained from:

$$S_{eff} = [(M_1 - M_2)/(p_{w1} - p_{w2})]d \quad (7)$$

where M_1 is moisture content of film at its underside surface (g/g solids); M_2 is moisture content of film at its surface outside the cup (g/g solids); and d is dry film density (g/cm^3).

Table 2. Moisture sorption isotherm models fitted to experimental adsorption isotherm data of a wheat gluten film[a]

Name	Model	Reference
Smith	$M = A - [B \ln(1 - a_w)]$	Smith (1947)
Oswin	$M = A[a_w/(1 - a_w)]^B$	Oswin (1946)
Halsey	$M = A[-1/\ln(Ta_w)]^B$	Boquet et al. (1978)
Flory–Huggins	$M = A \exp(Ba_w)$	Rogers (1985)
Kuhn	$M = [A(-1/\ln(a_w))^B] + C$	Kuhn (1964)
GAB	$M = (M_0 kCa_w)/[(1 - ka_w)(1 - ka_w + kCa_w)]$	Bizot (1984)

a. M, equilibrium moisture content (g water/g dry matter); M_0, monolayer moisture content (g water/g dry matter; a_w, water activity; A, B, C, k, constants.

Table 3. Water vapor permeability coefficients (P_{eff}), solubility coefficients (S_{eff}) and diffusion coefficients (D_{eff}) of wheat gluten films at different RH gradients

RH gradient (%)	Film thickness (μm)	P_{eff} (g×m/m²×s×Pa)	S_{eff} (g/m³×Pa)	D_{eff} (m²/day)
0.00–11.15	81	$3.8 \pm 0.4 \times 10^{-11}$	37.91 ± 0.01	$8.7 \pm 1.0 \times 10^{-8}$
11.15–22.60	83	$7.7 \pm 1.0 \times 10^{-11}$	44.68 ± 0.02	$1.5 \pm 0.2 \times 10^{-7}$
22.60–32.73	84	$1.4 \pm 0.1 \times 10^{-10}$	54.81 ± 0.02	$2.1 \pm 0.2 \times 10^{-7}$
32.73–43.80	85	$2.7 \pm 0.3 \times 10^{-10}$	70.34 ± 0.15	$3.3 \pm 0.4 \times 10^{-7}$
43.80–52.86	79	$5.2 \pm 0.3 \times 10^{-10}$	92.58 ± 0.18	$4.8 \pm 0.3 \times 10^{-7}$
57.70–70.83	83	$2.4 \pm 0.2 \times 10^{-9}$	147.98 ± 0.98	$1.4 \pm 0.1 \times 10^{-6}$
75.32–84.32	83	$4.1 \pm 1.7 \times 10^{-7}$	308.78 ± 0.31	$1.1 \pm 0.5 \times 10^{-4}$

D_{eff} for each pressure gradient studied was then calculated from:

$$D_{eff} = P_{eff}/S_{eff} \tag{8}$$

3. Results and discussion

3.1. *Effective water vapor permeability*

Experimentally determined P_{eff} values for WG films at different RH gradients are shown in Table 3. Because water vapor permeability of hydrophilic protein films is affected by film thickness, an effort was made to reduce film thickness variability to a minimum (79–85 μm). As expected with a hydrophilic film (Schwartzberg, 1986; McHugh and Krochta, 1994), P_{eff} increased substantially with increasing RH. Specifically, P_{eff} increased by four orders of magnitude from the lowest RH condition (0–11%) to the highest RH condition (75–84%). Most likely, at the high RH, extensive swelling of the protein network caused by sorbed water enhanced water molecule diffusion, thus substantially reducing the water vapor barrier ability of films (Gontard et al., 1993). For the sake of comparison, Briston (1988) reported a P value of 7.3–9.7×10^{-13} g/m·s·Pa for low density polyethylene film (25 μm thick at 38°C and 90% RH gradient). At a low RH condition, WG films exhibited 40-fold greater P_{eff} values than low-density polyethylene. The P_{eff} obtained in our study at 0–11% RH was similar to that (5.6×10^{-11} g·m/m²·s·Pa) reported by Gennadios et al. (1993) for WG films at comparable temperature and RH conditions. The slightly greater P_{eff} reported by Gennadios et al. (1993) was probably due to the thicker films (≈100 μm) used in that study. Generally, P_{eff} of hydrophilic films increases with film thickness, as shown for protein films from sodium caseinate (McHugh et al., 1993) and soy protein isolate (Ghorpade et al., 1995).

3.2. *Moisture adsorption isotherm*

Moisture adsorption isotherm data of WG films at 25°C within the 11–84% RH range are presented in Table 4. Estimated parameters and goodness of fit for the six fitted moisture sorption models are shown in Table 5. Both three-parameter models (Kuhn and GAB equations) showed better fits (G values of 3.54 and 7.24, respectively) than the two-parameter models. The GAB moisture sorption model is used widely for foods. It is an extension of the two-parameter BET (Brunauer–Emmett–Teller) model, taking into account the modified

Table 4. Experimental and predicted (by the Kuhn and GAB moisture sorption models) equilibrium moisture contents (g water/g dry matter) for adsorption isotherm of wheat gluten films at 25°C

a_w	Experimental	Kuhn model	GAB model
0.11	0.0100 ± 0.0015	0.0082	0.0145
0.23	0.0292 ± 0.0027	0.0309	0.0322
0.33	0.0515 ± 0.0024	0.0523	0.0514
0.44	0.0800 ± 0.0030	0.0803	0.0783
0.53	0.1114 ± 0.0024	0.1096	0.1076
0.71	0.2073 ± 0.0071	0.2064	0.2071
0.75	0.2482 ± 0.0204	0.2476	0.2494
0.84	0.3783 ± 0.0213	0.3865	0.3860

Table 5. Parameter (*A*, *B*, and *C*) estimates of various moisture sorption isotherm models and their goodness of fit (mean relative deviation modulus *G*) to moisture adsorption isotherm data of wheat gluten films at 25°C[a]

Model	*G*	*A*	*B*	*C*
Smith	18.20	–0.02374	0.20284	
Oswin	12.81	0.09791	0.82103	
Halsey	31.83	0.06612	1.01216	
Flory-Huggins	15.47	0.00901	4.54389	
Kuhn	3.54	0.12267	0.73149	–0.06088
GAB	7.24	0.09327	0.94428	1.37330

a. For GAB model, $A = M_0$ (monolayer moisture content), $B = k$ (constant correcting properties of multilayer molecules with respect to the bulk liquid); *C*, Guggenheim constant.

properties of the sorbate in the multilayer region through the introduction of a third parameter, *k*. This model showed a remarkable fit over a wide range of a_w values and a better evaluation of water tightly bound by the primary adsorption sites (Bizot, 1984). Also, the GAB model is simpler than the Kuhn model, since the GAB model does not include a logarithmic term. Therefore, although a slightly better fit was shown by the Kuhn equation, the GAB equation was used for estimating the film moisture contents (M_1 and M_2) at various RH gradients.

The monolayer water content (M_0) estimated from the GAB equation (0.0933 g/g dry matter) was slightly lower than that (0.1052 g/g dry matter) estimated by Gennadios and Weller (1994) for a similar WG film. The lower value was probably due to the higher protein content (minimum 80% on dry basis), and therefore lower starch content, of the WG product used in the present study than the WG product (75% protein on dry basis) used by Gennadios and Weller (1994). Starch is more effective in depressing a_w than protein. The parameter *k* (presented as *B* in Table 5) in the GAB model is a constant correcting the properties of the multilayer molecules with respect to the bulk liquid. The lower the *k* value from unity, the lower the sorption of water. The *k* value obtained in our study (0.9443) was greater than reported *k* values (~0.84) for proteins (Chirife et al., 1992). Most likely, the large amounts of hygroscopic glycerol incorporated into the films (40%, w/w, of WG) resulted in higher moisture sorption than that reported for proteins. Debeaufort et al. (1994) also observed increased monolayer water content with increased level of a_w depressants (plasticizers) in hydrophilic methylcellulose films.

3.3. *Effective solubility and diffusion coefficients*

For each studied RH gradient, S_{eff} was calculated (Table 3) from Equation (7) after estimates for M_1 and M_2 were obtained from the fitted GAB model. Also, Equation (8) was used to calculate D_{eff} values at each RH gradient (Table 3). As expected, S_{eff} and D_{eff} also increased, similar to P_{eff}, as the RH gradient applied across films moved upwards in the RH spectrum. Similar behavior has been documented for other hydrophilic films from calcium sodium pectinate (Schultz et al., 1949), hydroxypropyl methylcellulose/ethylcellulose (Woodruff et al., 1972), and hydroxypropyl methylcellulose/fatty acids (Kamper and Fennema, 1984).

4. Conclusions

P_{eff}, S_{eff}, and D_{eff} values of cast hydrophilic WG film increased substantially as the RH gradient applied across the films increased in the RH spectrum. Sorbed water induced film plasticization, increased polymer chain mobility and, thus, facilitated water vapor diffusion through the film. It was shown that water vapor transport parameters of WG films (and likely of other hydrophilic protein-based films) at given RH gradient conditions cannot be predicted from available data obtained at different RH gradient conditions. Instead, testing of such films at the expected conditions of actual use in packaging applications is necessary to quantify the water vapor permeation through the films.

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