Food Sci. Technol. Int. Tokyo, **3** (2), 163–169, 1997

Technical paper

Simulation of Water and Flavor Migration during Storage of Tobacco Products

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Received September 20, 1996

To propose an optimum packaging design and storage conditions for a tobacco product, which is composed of various solid components, a simulation model has been developed for predicting the amounts of water and flavor distribution among components during storage. The model is based on the adsorption equilibria of volatile vapors for the tobaccos, papers, filter and activated carbon as well as the permeation rate through the packaging films. The results of the simulation were in sufficient agreement with the experimental data to warrant the practical application of the model. The "Ethanol Adsorption Treatment" (EAT) for flavors with poor water solubility was confirmed to be remarkably effective when cigarettes were packaged with a film having a thin polyvinylidene chloride layer. When the products packaged with this film were stored under normal conditions for commercial marketing, such as inside a vending machine during the summer season, the water loss was predicted to be greater than the flavor loss, leading to a deterioration in the quality of the tobacco products. The optimum packaging designs and required conditions were obtained as follows: (1) the EAT for the filter tip, (2) storage conditions at lower temperatures, (3) selection of packaging film having a high vapor barrier against flavors.

Keywords: tobacco, water, flavor, migration, adsorption, permeation, storage, quality

Various volatile compounds are playing an increasingly important role in the enhancement of taste and smoke odors for cigarettes. However, a number of these volatile compounds are well known to migrate within the tobacco and its packaging materials, depending on their characteristics of adsorption equilibria or permeation through each packaging material. In this way, undesirable migrations of volatile compounds have caused serious deterioration in the quality of taste and odors. In particular, it is widely recognized as a serious problem that the migration of water and various flavors may fail to maintain desirable smoke odor and taste during storage inside a vending machine in the summer. In the food technologies field, many papers have been published with reference to the migrations or losses of flavors caused by sorption, as well as adsorption and permeation (Ayres *et al.*, 1983; Farrell, 1988; Mohney *et al.*, 1988; Ikegami *et al.*, 1991; Letinski & Halek, 1992).

In these papers, only the sorption and permeation for a specified packaging material were treated. However, for the solid products such as the tobaccos, the migrations or losses of flavors and water through the packaging material were governed by the vapor pressure of the volatile compounds. Therefore, it was apparent that we had to systematically measure the adsorption characteristics of all the materials used in the products and calculate the vapor pressure of the volatile compounds within the packages. Furthermore, for the plant material, i.e., tobacco, the binary adsorption equilibria of flavors and water must be studied, because the adsorbed water exerts a significant influence on the adsorption phenomena of the flavors. In our previous studies (Miyauchi *et al.*, 1995a, b, 1996a), the adsorption equilibria of various flavors and water for typical materials used in tobacco products were measured, and the vapor pressure of water and the flavor within the stored package were predicted based on their adsorption equilibria. The permeation coefficients of water and the various flavors through the packaging films were then published (Miyauchi *et al.*, 1996b). Also, the authors have developed a new technique, called the Ethanol Adsorption Treatment (EAT), for reducing flavor migration (Miyauchi *et al.*, 1995c). The fundamental concept of the EAT is based on the increase in the amount of flavor distribution in a tobacco column due to the increase in the amounts of adsorbed ethanol on a filter, as well as the ethanol vapor existing in the spaces between the cigarettes. The technique is performed by applying a mixture of water and ethanol vapors to the filter tip before combining it with the tobacco column. Therefore, the authors demonstrated the fundamental effects of the EAT on the migrations and losses of volatile compounds for solid products, based on the coupled properties of adsorption and permeation.

The objectives of this study were to (a) develop a simulation model for predicting the amount of water and flavor distribution among materials depending on their migration, (b) confirm the validity of the model by comparing the calculated amount of water and flavor distribution with the experimental data during normal storage conditions, (c) demonstrate the useful application of the model for considering an optimum packaging design after the EAT, and (d) present important factors of optimum packaging designs and conditions during commercial marketing storage.

Simulation of Water and Flavor Migration

In the box of the tobacco product, the water and volatile flavors adsorbed on the solid components such as tobacco were vaporized and sublimated, leading to the existence of their vapors in the spaces between the cigarettes during storage. Parts of the vapors were then adsorbed on each material contained in the box of the tobacco product and transferred outside the tobacco product through the packaging film. Because the manufacturing process of the tobacco product is carried out in an atmosphere having a relative humidity of 60%, the water content of each component was adjusted to the amount of adsorbed water in equilibrium with this relative humidity. The flavor was applied to the shredded tobacco, and then the tobacco products were stored for over 2 weeks until an equilibrium was attained within the package before shipping them for commercial marketing. In the calculation, the initial amounts of water and flavor distribution at $\theta=0$ were held in equilibrium with the relative humidity of 60% and the specific vapor pressure of the flavor. Furthermore, the water content was not affected by the flavor vapors because the flavors have been applied to the products in the lower vapor pressure range. Thus, the developed simulation model was easily applied to the individual water and flavor migrations. The amounts of water and flavor distribution were simulated on the basis of the adsorption equilibria and the permeation rate through the film, assuming that each component of the tobacco product was held in equilibrium with the vapors in the spaces between the cigarettes. As a result of the simulation, the amounts of water and flavor distribution were calculated within the total storage time of 28 days. To evaluate these calculated amounts based on the assumed condition of equilibrium, they were compared with data which had been obtained from experiments carried out under normal storage conditions.

Conditions

Tobacco product The components of a typical commercial tobacco product listed in Table 1 were used in the calculation. The structure of films A, B and C were the same as that reported in the previous study (Miyauchi *et al.*, 1996b). Films A and B were mainly made of polypropylene layers, while film B was coated with a thin polyvinylidene chloride layer. Film C consisted of some complex polymer layers, including an aluminum layer. Films A, B and C were 22.5, 23, and 24 μm in thickness, respectively, except that film A used in the experiment (see Fig. 3) had a 30 μm thickness.

Storage conditions The storage conditions are shown in Table 2. In cases A and B, the temperature, vapor pressure of water as well as the flavor were kept constant, while in the case C, the temperature made a cyclic change as follows:

$$\text{at } \theta<8 \text{ or } \theta>16 \quad T=303$$
$$\text{at } 8\leq\theta\leq16 \quad T=20\sin\left(\frac{\theta-8}{8}\pi\right)+303, \qquad (1)$$

while the vapor pressures were kept constant as:

$$p_w=3.0$$
$$p_f=0 \qquad (2)$$

Case C mimicked the condition inside the vending machine in the summer (see Fig. 1); this was found to be appropriate by a preliminary test.

Modeling In this simulation, we assumed the steady-state conditions of the vapor pressure of the volatile compounds within (p_{in}) and outside (p_{out}) the package, respectively. After the incremental stored time (Δt), the total amount (ΣW_i) of the volatile compounds within the package was obtained by adding or subtracting the permeation amount

Table 1. Components of a tobacco product.

Components of cigarette			
Tobacco column		Filter tip	
Bright yellow tobacco	5.3 g	Acetate filter	4.2 g
Burley tobacco	5.3 g	Tipping paper	0.6 g
Cigarette paper	0.8 g	Wrapping paper	0.3 g
		Activated carbon	
		(Charcoal filter)	0.8 g
		(Non-charcoal filter)	0 g
Components of package			
Hard pack	4.7 g	Aluminum foil	0.9 g
Film	3 types[a]		
Dead space	8.0×10^{-5} m^3	Film area	1.7×10^{-2} m^2

[a] The films used are described elsewhere (Miyauchi *et al.*, 1996b).

Table 2. The conditions at which the prediction of water and flavor migration were estimated and measured.

Conditions	Temperature [K]	Relative pressure of water [−]	Relative pressure of flavor [−]	Storage time [day]
A	303[a]	0	—	28
	319[b]	0	—	28
B	303[a]	0.6	0	28
	319[b]	0.6	0	28
C	Eq. (1)	*c*)	0	28

[a] Temperature was a value for a product of mentholated tobacco.

[b] Temperature was a value for other products.

[c] Values are obtained from Eq. (2) and the Antoine equation.

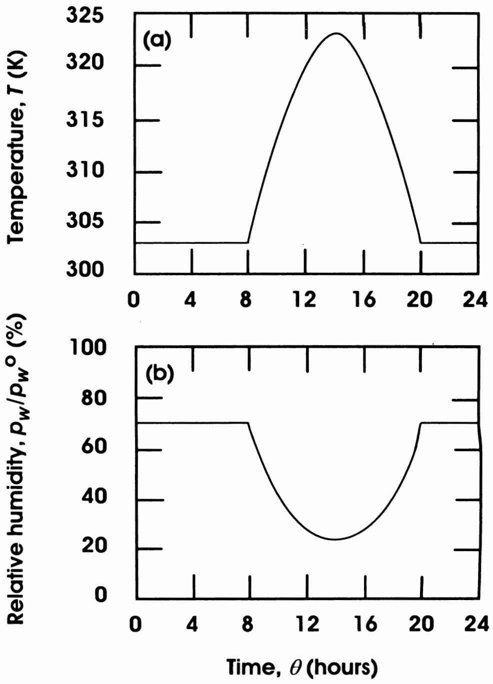

Fig. 1. Assumed conditions of (a) temperature and (b) relative humidity inside vending machines in the summer using Eqs. (1), (2).

(Q_i) from the total amount (ΣW_i) at $\Delta t=0$. The permeation rate ($Q_i/\Delta t$) through the packaging film is given as:

$$\left(\frac{Q_i}{\Delta t}\right)=P_i\frac{a}{l}(p_{in}-p_{out}) \tag{3}$$

where P_i is the permeation coefficient of the packaging film. The value of p_{in} was calculated using the following adsorption equilibrium.

Adsorption equilibrium Water adsorption equilibrium: A modified Dubinin-Astakhov (DA) equation (Kame-oka & Horibe, 1994) was used as follows:

$$w_i=w_i^0\exp\left(-\left(\frac{A}{E}\right)^n\right) \tag{4}$$

The values of n, E and w_i^0 for the tobacco and its packaging materials used in the calculation are listed in Table 3.

Binary adsorption equilibria: For tobaccos, papers and filter, the amount of adsorbed water was assumed constant (except that some of the water adsorbed on the filter was slightly desorbed as the ester was adsorbed), while the amount of adsorbed flavors was calculated by:

$$q_i=k\times p_i \tag{5}$$

For activated carbon, the amount of adsorbed water was assumed to be zero and that of the flavor was calculated using the DA equation. The values of k, w_i^0 and E of the flavor presented for the binary adsorption equilibria of the water and flavor are listed in Table 4.

Multi-component adsorption equilibria: The amounts of a mixture of more than two organic compounds adsorbed on activated carbon were correlated by the following equation (Lewis *et al.*, 1950):

$$\Sigma\left(\frac{q_i}{q_i^*}\right)=1 \tag{6}$$

where q_i^* is the adsorbed amount, as in a pure volatile adsorption isotherm, at the vapor pressure corresponding to the total vapor pressure of the volatile compounds (Σp_i). The effect of the EAT was evaluated using the flavor distribution ratio (D_t), which was defined as the amount of flavor for the tobacco column to that for the total tobacco products; it was estimated on the basis of the following approximations (Miyauchi *et al.*, 1995c): (a) for tobaccos, papers, or filters; the adsorbed amounts could be evaluated by the binary adsorption equilibria, and (b) for activated carbon; those were determined based on the multi-component adsorption equilibria. As the amounts of water vapor are greater than those of any other compound, the value of q_w^* could be assumed to be nearly constant. Therefore, the (q_w/q_w^*) value in Eq. (6) was assessed to be 0.1 based on the data of the ternary adsorption (Miyauchi *et al.*, 1995c).

Table 3. The constants[a] of DA equation (4) for adsorption equilibrium of water which were used in the calculation of this work.

Material	n	w^0 [cm³/g]	E [kJ/mol]
Tobacco column			
Bright yellow tobacco	0.43	2.15	0.12
Burley tobacco	0.33	1.86	0.07
Cigarette paper	0.86	0.10	3.57
Filter tip			
Acetate filter	0.42	0.28	0.30
Tipping paper	0.60	0.16	1.66
Wrapping paper	0.46	0.26	0.97
Package			
Hard pack	0.58	0.17	1.74
Aluminum foil	0.94	0.10	2.54

[a] Miyauchi *et al.* (1995a).

Differential adsorption equilibria: The variation in the amounts of water and flavor adsorbed was estimated as a function of vapor pressure using the following differential equation obtained from Eq. (4) or (5), respectively:

$$\frac{\mathrm{d}q}{\mathrm{d}p}=-q\frac{n\left(RT\ln\frac{p^0}{p}\right)^{n-1}}{E^n}\frac{RT}{p} \tag{7}$$

$$\frac{\mathrm{d}q}{\mathrm{d}p}=k \tag{8}$$

Permeation coefficient For films A and B, the permeation coefficient (P) was estimated using the following equation:

$$P=P_0\exp(\alpha p) \tag{9}$$

For film C, the permeation coefficient was given as (Kagaku Kogaku Kyokai (ed.), 1988):

$$P=\frac{2r}{3}\frac{c^{\frac{1}{2}}}{RT}n_p\pi r^2=C_3T^{-\frac{1}{2}} \tag{10}$$

The parameters to estimate P_0, α and the parameter C_3 are given in Table 5.

Calculation procedure The calculation procedure is illustrated in Fig. 2. The number following corresponds to the step number shown in the frame.

(1) The initial value of $q_i(0)$ for each material was calculated utilizing the adsorption equilibria.

(2) By assuming that the change in the vapor pressure (Δp_i) was 0.1% of the vapor pressure, i.e., $\Delta p_i=0.001P_i$, the change in the amount of the volatile compound (ΔW_i) was given by

Table 4. The constants of Eqs. (4) and (5) for adsorption equilibrium of ethanol and flavors which were used in the calculation of this work.

Material	Ethanol	Ethyl acetate[b]	Ethyl butyrate[d]	D-Limonene[d]	L-Menthol[d]
	k[mol/(kg·kPa)]				
Tobacco column					
Bright yellow tobacco	0.20[a]	0.022	0.042	0.073	1.76
Burley tobacco	0.13[a]	0.022	0.054	0.217	2.61
Cigarette paper	0.04[c]	0.011	0.009	0.045	0.55
Filter tip					
Acetate filter	0.51[c]	0.285	0.627	0.261	2.59
Tipping paper	0.05[c]	0.010	0.009	0.025	0.55
Wrapping paper	0.04[c]	0.009	0.009	0.006	0.55
Package					
Hard pack	0.05[c]	0.012	0.016	0.057	0.38
Aluminum foil	0.03[c]	0.009	0.009	0.020	0.55
Activated carbon	w^0[cm³/g]				
	0.22[c]	0.26	0.25	0.20	0.13
	E[kJ/mol]				
	11.5[c]	12.4	22.5	22.0	16.0
Activated carbon	w^0[cm³/g]				
	0.25[c]				
	E[kJ/mol]				
	12.7[c]				

[a] Nakanishi & Kobari (1989), [b] Miyauchi *et al.* (1995b), [c] Miyauchi *et al.* (1995c), [d] Miyauchi *et al.* (1996a).

utilizing the differential adsorption equilibria.
(3) The value of the incremental stored time (Δt) corresponding to the change in the vapor pressure was calculated using the permeation rate and the obtained ΔW_i value.
(4) The value of Δp_i decreased until Δt was small enough to accept the assumed condition of equilibrium, i.e., $\Delta t < 1$ h. The procedure for assuming Δp_i was selected depending on the types of volatile compounds, films and storage conditions.
(5) If the results satisfy the limitation of step (4), the total storage time (t) was calculated by adding the Δt obtained in step (4).
(6) At the obtained total storage time, the values of $q_i(t)$ for each material were determined by utilizing the adsorption equilibria.
Steps (2)–(6) were repeated until the total storage time of 28 days was reached.

Case of EAT: For the ethanol vapor, the Δt, p_e and q_e values of ethanol were calculated as already mentioned. Next, the value of ΔW_f for the flavors was given using the permeation rate of the flavor and the obtained Δt. For the activated carbon, the values of p_f and q_f of the flavor were estimated by the multi-adsorption equilibria, i.e., Eq. (6).

Experimental

Materials To confirm the practical validity of the application of the model, experiments were designed to measure the amounts of water and flavor distribution during the storage of the tobacco products. The components of the samples to be tested are shown in Table 1 and packaged using the films A and B.

Storage test The amounts of water adsorbed on the tobacco were measured at intervals when the boxes of tobacco products were stored under constant storage condition A. To investigate the amounts of flavor migration, the amounts of ethanol for the filter tip or L-menthol for the tobacco column were measured at intervals under storage condition B. The apparatus used in the storage test was the same flow-type multi-component adsorption system as described in a previous study (Miyauchi *et al.*, 1995a). The tested samples were placed into the vessels with a 1000 cm^3 volume, and they were then attached to the adsorption system. The gas whose relative humidity was at 0 or 60% flowed through the vessels at 100 cm^3/min under the condition of a constant temperature of 303 or 319 K. The amounts of water or flavor desorbed were analyzed with the TCD or FID of a gas chromatograph, respectively.

Results and Discussion

Performance of the model For the products packaged with film A or B, the calculated and experimental amounts of (a) water adsorbed on the tobacco, (b) ethanol adsorbed on the filter tip and (c) L-menthol adsorbed on tobacco column were plotted against the storage time, as shown in Figs. 3 or 4, respectively. The calculated results indicated as the solid curves were in good agreement with the experimental data. These results demonstrated that the assumed condition of equilibrium was acceptable. In addition, the motive force of the water and flavor migrations between the product and its surroundings is due to two effects of permeation through the packaging film, as well as the diffusion through holes existing in parts pasted together for the packaging film. This agreement between the calculated and experimental results indicated that the migration between the product and its surroundings was mainly governed by the permeation through the packaging film. Therefore, these results allowed the practical

Table 5. Parameters[a] of permeation coefficient used in the calculation of this work.

$$P_0 = C_1 \exp\left(-\frac{C_2}{RT}\right)$$

$$P = C_3 T^{-\frac{1}{2}}$$

$$\alpha = -C_4 \times T + C_5$$

Films	Permeants	C_1 [mol/(m·s·kPa)]	C_2 [kJ/mol]	C_4 [1/(kPa·T)]	C_5 [1/(kPa)]
Film A					
	Water	2.7×10^{-7}	27		
	Ethanol	9.0×10^{-2}	64	0.002	0.576
	Ethyl acetate	1.4×10^{-2}	59	−0.002	−0.418
	Ethyl butyrate	7.7×10^{-2}	62	0.031	10.388
	D-Limonene	1.6×10^{5}	98	1.010	325.63
	L-Menthol	$P_0 = 2.5 \times 10^{-11}$		$\alpha = 9.74$	
Film B					
	Water	2.7×10^{-5}	38		
	Ethanol	2.0×10^{6}	115	0.008	2.727
	Ethyl acetate	5.2×10^{11}	149	0.024	7.984
	Ethyl butyrate	5.9×10^{16}	184	0.068	23.071
	D-Limonene	5.3×10^{7}	140	2.013	641.92
	L-Menthol	$P_0 = 1.3 \times 10^{-13}$		$\alpha = 19.74$	

		C_3 [mol/(m·s·kPa·T$^{1/2}$)]
Film C	Water	1.92×10^{-10}

[a] Miyauchi *et al.* (1996b).

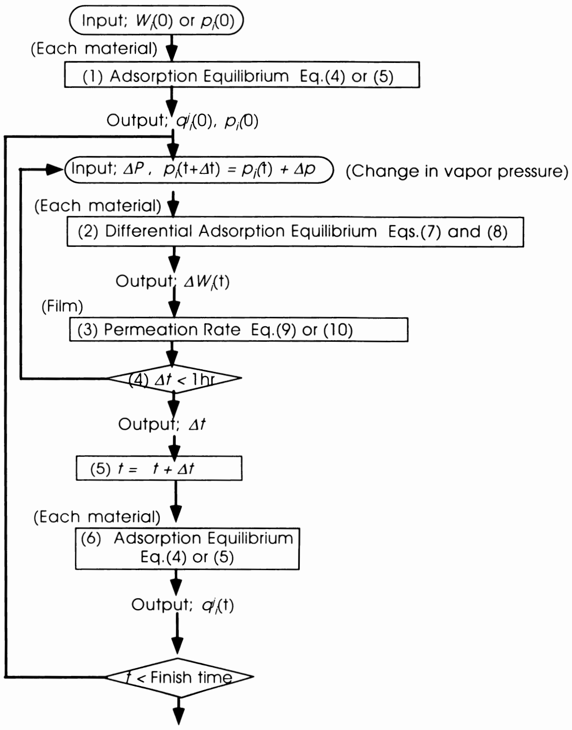

Fig. 2. Schematic diagram showing calculation procedure for water and flavor migrations.

use of the model.

Effect of the EAT In Fig. 5 the flavor distribution ratio for the tobacco column (D_t/D_{t0}) is represented as a function of the total amount of ethanol in the tobacco products. For flavors having relatively poor water solubility, the EAT was found to be remarkably effective for increasing the flavor content in the tobacco column, in terms of the value of D_t/D_{t0} as well as the minimum amounts of ethanol needed for the EAT. The ratio for D-limonene reached the highest ratio of over 1000 among the flavors tested. It was also indicated that the tendency for minimum amounts of ethanol needed in the EAT decreases with an increase in the amounts of the added flavors.

As shown in Fig. 6, the variation in the amounts of ethyl acetate adsorbed on tobacco was simulated for storage condition B after the EAT, compared with their variation in the product without treatment. Because the vapor pressure of the flavors within the package increased after the EAT, the permeation rate as well as loss increased. In the application of the EAT, it was extremely important to select a film having the characteristics of a higher barrier against flavor.

During storage in the vending machine The simulated amounts of the water and flavor distributions during storage condition C are presented in Figs. 7 and 8, respectively, and Fig. 9 shows the amounts of ethyl acetate distribution for the same conditions after the EAT was carried out. In these figures, the adsorbed amounts of water and flavor at $\theta=0$ were plotted versus storage time. As shown in Fig. 7, it was recognized that water migrates from the tobacco to the surroundings inside the vending machine in the summer, leading to an undesirable drying of the products. The percentage of the amounts of water content in tobacco to that of the initial water content was assessed to be 73% for film B after 28 days of storage. Therefore, this film is unable to

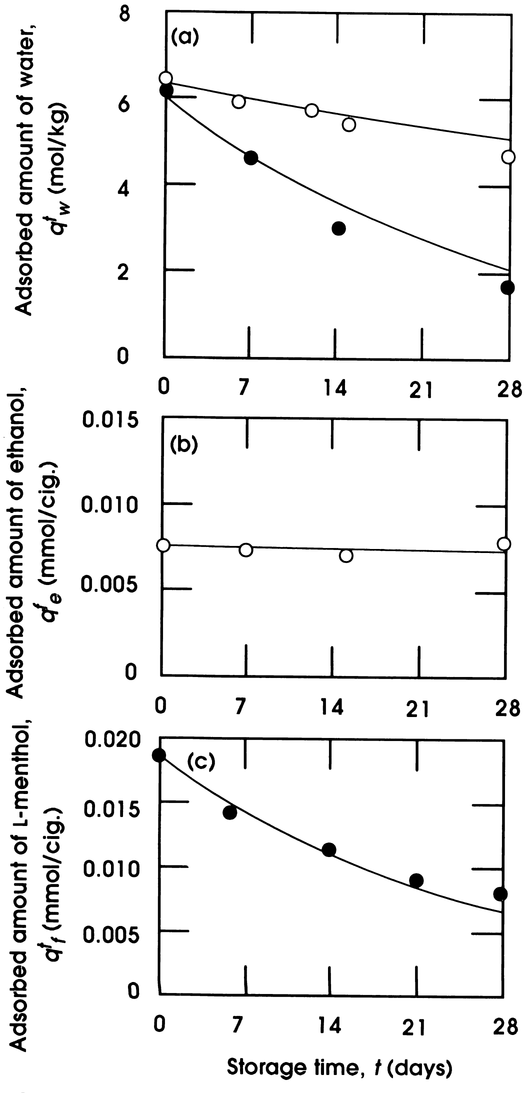

Fig. 3. Comparison of calculated and experimental values for amounts of (a) water adsorbed on tobacco, (b) ethanol adsorbed on filter tip in charcoal filter products and (c) L-menthol adsorbed on tobacco column in non-charcoal filter products, packaged with film A (film thickness 30 μm) during storage. ——, calculated; ○, experimental in charcoal filter products at 303 K; ●, experimental in non-charcoal filter products at 319 K.

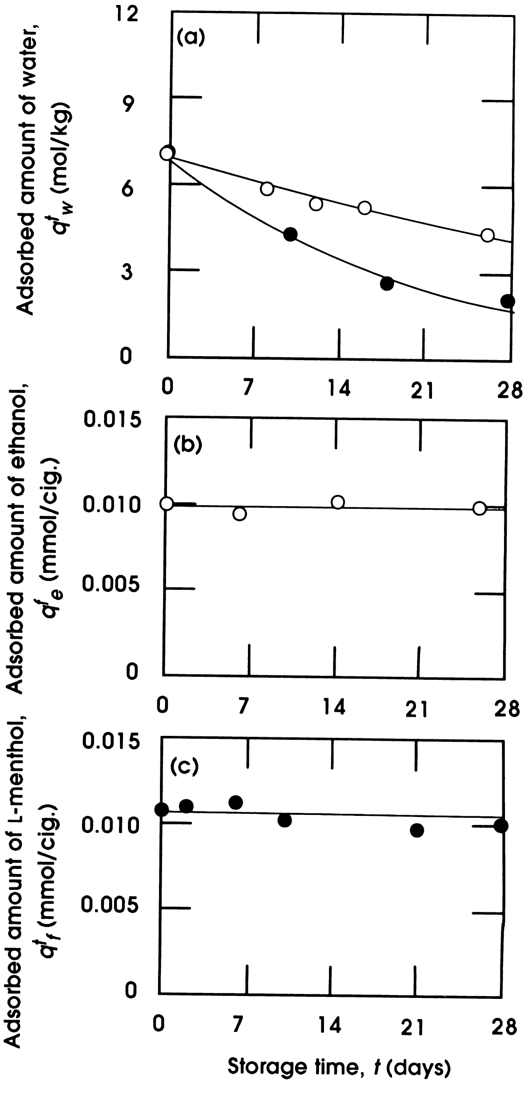

Fig. 4. Comparison of calculated and experimental values for amounts of (a) water adsorbed on tobacco, (b) ethanol adsorbed on filter tip in charcoal filter products and (c) L-menthol adsorbed on tobacco column in non-charcoal filter products, packaged with film B during storage. ——, calculated; ○, experimental in charcoal filter products at 303 K; ●, experimental in non-charcoal filter products at 319 K.

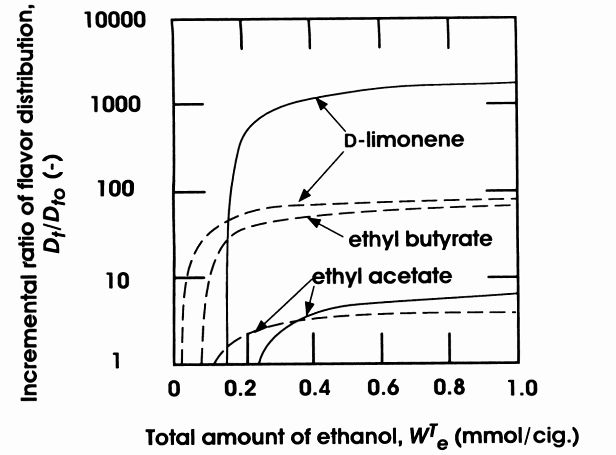

Fig. 5. Incremental ratio of flavor distributed amounts as a function of total amount of ethanol. ——, flavor of 50 mg in loading amount; ------, flavor of 100 mg in loading amount.

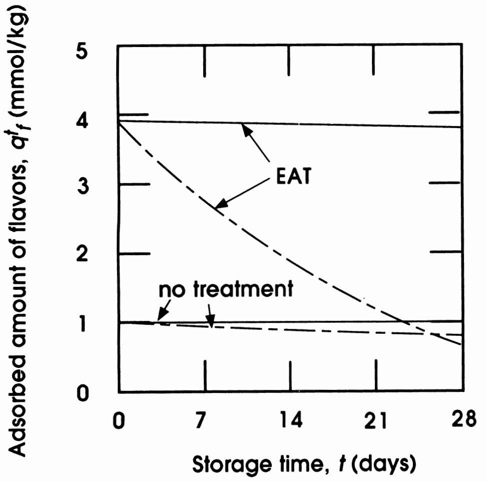

Fig. 6. Variation in amounts of ethyl acetate adsorbed on tobacco in products during storage after Ethanol Adsorption Treatment and no treatment at a temperature of 303 K and relative humidity of 60%. -·-·-, film A; ——, film B.

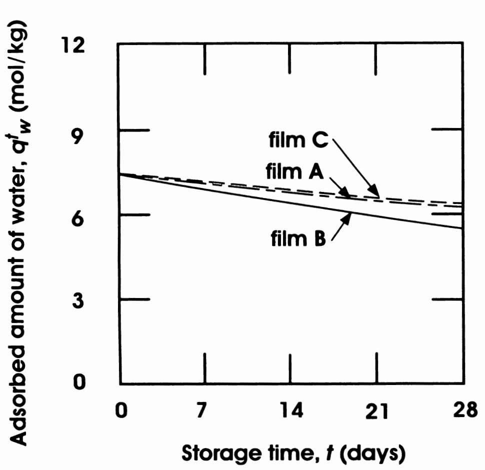

Fig. 7. Adsorbed amounts of water simulated for tobacco in products during storage inside vending machines in the summer.

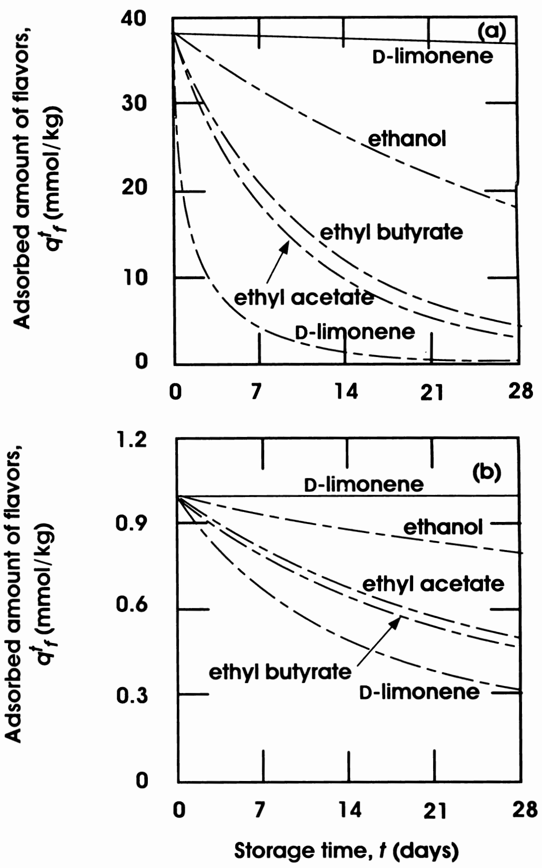

Fig. 8. Adsorbed amounts of various flavors simulated for tobacco in (a) non-charcoal and (b) charcoal products during storage inside vending machines in the summer. -·-·-, film A; ——, film B.

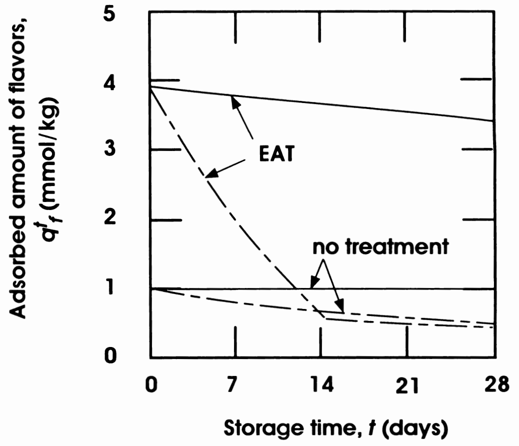

Fig. 9. Adsorbed amounts of ethyl acetate simulated for tobacco in products during storage inside vending machine in the summer after Ethanol Adsorption Treatment and no treatment. -·-·-, film A; ——, film B.

function as a high-barrier film to maintain the initial water content. The amounts of flavor losses through film A were found to be great, as shown in Fig. 8. Furthermore, as shown in Fig. 9, the effect of the EAT disappears for products packaged using film A stored longer than 14 days. Therefore, a serious flavor loss through film A took place, while the amount of flavor loss through film B was negligible over the whole range of storage conditions for commercial marketing. These results indicated that when a product packaged with a film having the characteristics of a high barrier against flavor, i.e., film B, was stored inside the vending machine in the summer, water loss affected the deterioration of the tobacco products, compared with the flavor dissipation.

Important factor for optimum packaging designs and conditions The proposed model provided useful information for considering the optimum packaging design as well as the storage conditions for commercial marketing. The important findings to improve the quality of the tobacco products during storage are summarized as follows: (1) the EAT for the filter tip can accomplish the desired amount of water and flavor distribution within the tobacco column, (2) the storage at a lower temperature enables the water content of the tobacco column to maintain the initial content, and (3) the selection of a packaging film which provides a high barrier against flavor can realize a more effective reduction of the flavor loss than the storage temperature.

Nomenclature

A =free energy of adsorption [J/mol]
a =exposed surface area of film [m^3/mol]
c =mean molecular velocity [m/s]
D_t =ratio of flavor distributed amount [—]
D_{t0} =ratio of flavor distributed amount when ethanol does not exist in products [—]
E =characteristic free energy of adsorption [J/mol]
k =constant in Eq. (5) [mol/(kg·kPa)]
l =film thickness [m]
n =constant in Eq. (4) [—]
n_p =pore number in aluminum layer [—]
P =permeation coefficient [mol/(m·s·kPa)]
P_0 =permeation coefficient extrapolated to zero concentration [mol/(m·s·kPa)]
p =vapor pressure [Pa]
p^0 =saturated vapor pressure [Pa]
Q =amount of permeated compound [mol]
q =amount of adsorbed compound [mol/kg]
q^* =amount of adsorbed compound, as in pure isotherm [mol/kg]
R =gas constant [J/(K·mol)]
r =pore radius in aluminum layer [m]
T =temperature [K]
t =storage time [day]
W =amount of volatile compounds [kg]
w =volume of adsorbed compound [m^3/kg]
w^0 =constant in Eq.(4) (=micropore volume) [m^3/kg]
α =concentration dependence factor of permeation coefficient [1/kPa]
θ =time [h]

⟨Subscripts⟩
e =ethanol
f =flavor
i =vapor compound
w =water

⟨Superscripts⟩
f =filter
j =material
T =total (=the sum of the tobacco column and filter tip)
t =tobacco

References

Ayres, J.L., Osborne, J.L., Hopfenberg, H.B. and Koros, W.J. (1983). Effect of variable storage times on the calculation of diffusion coefficients characterizing small molecule migration in polymers. *Ind. Eng. Chem. Prod. Res. Dev.*, **22**, 86-89.

Farrell, C.J. (1988). Characterization and control of organic flavor molecule absorption into polyolefin containers. *Ind. Eng. Chem. Res.*, **27**, 1946-1951.

Ikegami, T., Nagashima, K., Shimoda, M., Tanaka, Y. and Osajima, Y. (1991). Sorption of volatile compounds in aqueous solution by ethylene-vinyl alcohol copolymer films. *J. Food Sci.*, **56**, 500-503, 509.

Kagaku Kogaku Kyokai (ed.) (1988). "Kagaku Kogaku Binran, 5th ed." Maruzen, Tokyo, pp. 158-164 (in Japanese).

Kameoka, T. and Horibe, K. (1994). Equilibrium moisture content equation for food (part 1)—DA equation and Weibull distribution—. *Nogyokikai-Gakkaishi*, **56**, 57-62 (in Japanese).

Letinski, J. and Halek, G.W. (1992). Interactions of citrus flavor compounds with polypropylene films of varying crystallinities. *J. Food Sci.*, **57**, 481-484.

Lewis, W.K., Gilliland, E.R., Chertow, B. and Cadogan, W.P. (1950). Adsorption equilibria. Hydrocarbon gas mixtures. *Ind. Eng. Chem.*, **42**, 1319-1326.

Miyauchi, M., Miyake, A., Nakanishi, Y. and Sagara, Y. (1995a). Characteristics of water adsorption isotherms for materials contained in a box of a tobacco product. *Drying Technol.*, **13**, 351-370.

Miyauchi, M., Miyake, A., Nakanishi, Y. and Sagara, Y. (1995b). Binary adsorption isotherms of water and ethyl acetate vapor for materials contained in a box of a tobacco product. *Drying Technol.*, **13**, 1741-1761.

Miyauchi, M., Miyake, A., Nakanishi, Y. and Sagara, Y. (1995c). A new ethanol adsorption treatment to decrease flavor and water migration during storage of a tobacco product. *Food Sci. Technol., Int.*, **1**, 44-51.

Miyauchi, M., Miyake, A., Nakanishi, Y. and Sagara, Y. (1996a). Binary adsorption equilibria of various flavors and water for materials contained in a box of a tobacco product. *Food Sci. Technol., Int.*, **2**, 65-71.

Miyauchi, M., Nakanishi, Y. and Sagara, Y. (1996b). Permeation of water and volatile flavors through packaging films. *Food Sci. Technol., Int.*, **2**, 217-222.

Mohney, S.M., Hernandez, R.J., Giacin, J.R., Harte, B.R. and Miltz, J. (1988). Permeability and solubility of *d*-limonene vapor in cereal package liners. *J. Food Sci.*, **53**, 253-257.

Nakanishi, Y. and Kobari, M. (1989). Binary adsorption equilibria of water and ethanol vapor on contractive plant material. *Kagaku Kogaku Ronbunshu*, **15**, 968-976 (in Japanese).