

A New Ethanol Adsorption Treatment to Decrease Flavor and Water Migration during Storage of a Tobacco Product

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Received May 15, 1995

A new "Ethanol Adsorption Treatment" (EAT) for a filter has been presented to depress the migration of volatile flavor and water in a box of a tobacco product during storage and marketing. The EAT is performed by applying the binary vapor of water and ethanol to the filter tip before connecting it with a tobacco column. During the EAT, ethanol was adsorbed physically on the filter as an additive agent, and thus the water content within the filter tip was controlled to a desirable level at which the water does not migrate. Then during storage, the presence of ethanol vapor depressed the flavor migration mainly by decreasing the amounts of flavors adsorbed on the activated carbon within the filter tip. The EAT was confirmed to be effective for decreasing both the water and flavor migration simultaneously.

Keywords: tobacco product, activated carbon, migration, flavor, water, ethanol, binary adsorption isotherm

Various volatile flavors are being applied to food and tobacco products for the enhancement of taste and smoke odor. In the food industry, it is widely recognized that significant loss of flavor occurs in stored products when polymer films have been used as outer packaging materials. Many papers have been published with reference to their losses caused by diffusion or sorption as well as adsorption (Shimoda *et al.*, 1984; Mohny *et al.*, 1988; Halek & Meyers, 1989; Sadler & Braddock, 1990). In the box of a tobacco product, the volatile flavor components migrate and distribute among each material during storage and marketing (Samejima, 1983). The depression of undesirable flavor loss has been realized by surface modification of the packaging materials (Corbin *et al.*, 1985; Matsui *et al.*, 1991; Fukamachi *et al.*, 1993) or application of a flavor component such as a glycoside to the tobacco product (Herron, 1989). In our previous study (Miyachi *et al.*, 1995a, b), it was found that water migrates from the filter tip to the tobacco column, while flavors transfer backwards, depending on the selectivity as well as the mechanism of adsorption. Especially, the flavor migration to the charcoal filter which contains the activated carbon is greater than that to the plain filter without it.

A manufacturing process for a tobacco product is shown in Fig. 1. In the flavoring process, a mixture of ethanol and various flavors is sprayed onto the shredded tobacco leaves. Next, the tobacco is rolled with cigarette paper to make a tobacco column. On the other hand, the filter tip is made by composing cellulose acetate fiber or tow, activated carbon and wrapping paper. The tobacco column is then connected to the filter tip using tipping paper to make a cigarette, and the final product is obtained by packaging the cigarettes into a box. In this paper, a new "Ethanol Adsorption Treatment" (EAT) of a charcoal filter has been proposed to decrease both water and flavor migration during storage and marketing, and

it is applied to the filter tip before the connection of the charcoal filter to the tobacco column, as shown in Fig. 1.

The purpose of the EAT is to add some substance to the charcoal filter for controlling the adsorption equilibria between the activated carbon and the volatile components. Ethanol was selected and examined as an adsorbed agent for the EAT because it has been used as a solvent in the flavoring process as mentioned above. The EAT has two beneficial characteristics; i.e., (a) the control of the water content within the filter tip and (b) the depression of flavor migration due to the increase in ethanol vapor in the spaces between the cigarettes. To evaluate the effectiveness of the EAT, the behavior of ethanol added by the EAT has to be clarified by systematically measuring the binary adsorption equilibria of water and ethanol for the packaging materials. In addition, the adsorption equilibria for the filter as well as the activated carbon need to be determined in the desorption step for considering the application of the EAT. Further, to investigate the mechanism of the decrease in the flavor migration, the ternary adsorption equilibria of water-ethanol-flavor vapor should be measured for typical materials contained in the tobacco product.

The objectives of this study were to (a) measure the adsorption isotherms of pure ethanol and binary water-ethanol vapors for typical packaging materials used for the tobacco product and (b) evaluate the effect of the EAT on decreasing the water and flavor migration during storage, based on these adsorption equilibria.

Experimental

Materials Typical materials contained in the box of a tobacco product are listed in Table 1 and used in the measurements of binary adsorption equilibria. The values of micropore volume (v_p) as well as the specific surface area (s)

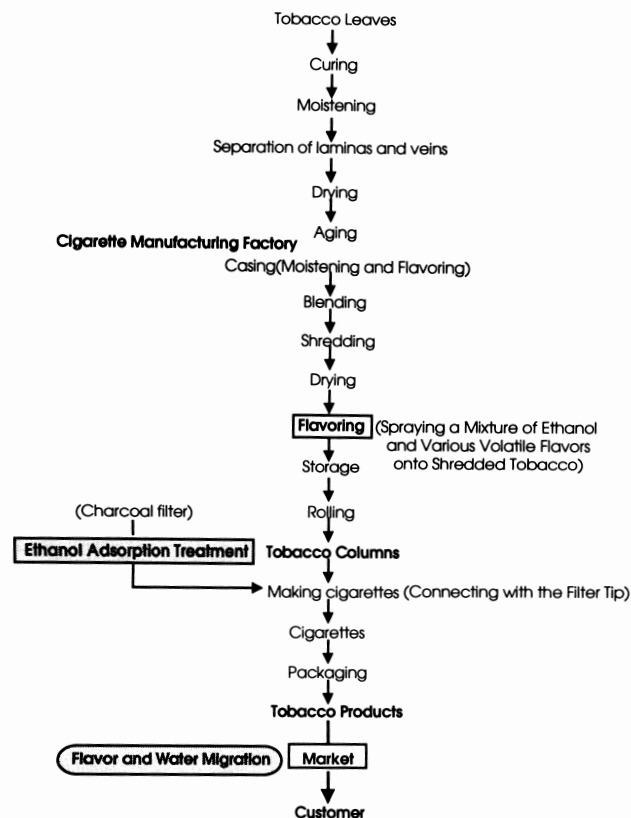


Fig. 1. Manufacturing and marketing process diagram of tobacco product.

Table 1. Experimental materials.

Paper	Filter	Activated carbon
Cigarette paper	Tow	Coconut
Tipping paper	Filter	Oil palm
Wrapping paper		Coconut in filter
Aluminum foil		
Label (soft pack)		
Label (hard pack)		
Stamp		

Table 2. Physical properties of activated carbon.

Activated carbon	Micropore volume, v_p $\times 10^{-3}$ [m ³ /kg]	Specific surface area, s $\times 10^3$ [m ² /kg]
Coconut	0.49	1255
Oil palm	0.39	1039
Coconut in filter	0.22	529

for activated carbon were determined from the measurement of nitrogen adsorption isotherms and are presented in Table 2.

For investigating the effect of the EAT, the following cigarettes were used; the tobacco columns were made from a variety named “bright yellow,” having the dimensions of 70 mm in length, 25 mm in circumference and 1030 to 1070 mg in mass. The filter tips were composed of charcoal and acetate filter with a length of 10 mm and 15 mm. The charcoal filter contained the activated carbon of 40 mg in mass. The characteristics of the cellulose acetate tow, which was used in both charcoal and acetate filters, was 4 denier per filament and 35,000 total denier, having a “Y” shape in cross section. In the

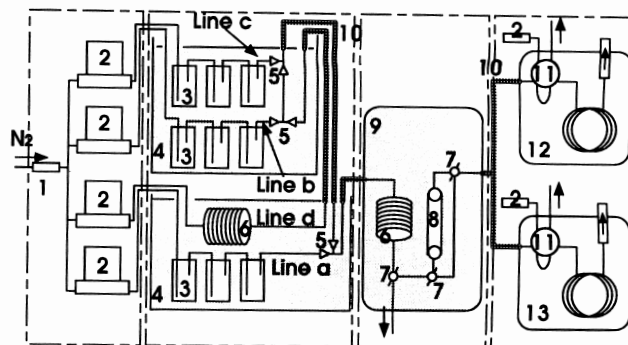


Fig. 2. Flow-type multi-component adsorption system. 1, dryer; 2, flow meter controller; 3, bubbler; 4, water bath; 5, valve; 6, heat exchanger; 7, three-way cock; 8, adsorption cell; 9, air bath; 10, heater; 11, six-way cock; 12, gas chromatograph (TCD); 13, gas chromatograph (FID).

same manner as used in the manufacturing process, both the tobacco column and filter tip were conditioned at a constant temperature of 295 K and relative humidity of 60% before the measurements.

Apparatus The schematic drawing of a flow-type multi-component adsorption system is shown in Fig. 2. The main functions of the system are the control of gas flow-rates, volatile vapor supply, operation of adsorption, and analysis of vapor concentrations.

Nitrogen dehumidified in a dryer (1) flowed through the flow meter controllers (2) at 0–100 cm³/min in each line. A part of it flowed into bubblers (3) of water to obtain gas saturated with water vapor (Line a), and another part flowed into bubblers of ethanol and ethyl acetate to obtain gas saturated with their vapor (Line b, c), in the same arrangements as shown in Fig. 2. Three bubblers used in all lines were immersed in a water bath (4) at a constant temperature of 303 K. The binary or ternary mixtures of water-ethanol or water-ethanol-ethyl acetate vapor were obtained by mixing the saturated vapors (Line a, b, c) and the dehumidified gas (Line d). The connection line (10) between the water and air baths was heated to prevent undesirable condensation. Two coil-tube heat exchangers (6) were used to control the temperature of the gas, and then the gas was injected into the adsorption cell (8). During the adsorption processes, the cell was located in an air bath (9) where the temperature was maintained at 303 K. At the outlet of the adsorption cell, the concentrations of water or ethanol as well as ethyl acetate vapors were analyzed with a thermal conductivity detector (TCD) (12) or a flame ionization detector (FID) (13) of the gas chromatograph by operating a six-way cock (11) and using a sampling tube, respectively.

Measurements of binary adsorption equilibria To remove intrinsic volatile compounds and any adsorbed vapor, the materials were pre-treated by vacuum drying. The drying conditions are listed in Table 3 along with other experimental conditions. The binary adsorption experiments were conducted as follows; In the adsorption step of ethanol, each material was held in equilibrium with a constant vapor pressure of pure water of $p_w = 2.5$ kPa corresponding to $p_w/p_w^0 = 0.6$ where the ethanol vapor pressure was zero, i.e., $p_e/p_e^0 = 0$. On the other hand, in the desorption step the material was held in equilibrium with that of pure ethanol of $p_e = 8.4$

Table 3. Experimental conditions.

Materials	Pre-treatment conditions	Desorption conditions
Tobacco	353 K, 8 H	403 K, 1 H
Paper	373 K, 8 H	403 K, 1 H
Filter	373 K, 8 H	403 K, 1 H
Activated carbon	473 K, 3 H	473 K, 3 H
Gas chromatograph conditions		
	Water	Ethanol
For adsorption		
Column	ID 2 mm×1 m	ID 3 mm×3 m
Liquid phase	Porapak Q	PEG20
Detector	TCD	FID
Flow rate	50 ml/min	80 ml/min
Oven temperature	413 K	343 K
Detector temperature	503 K	503 K
For analysis		
Column	ID 0.53 mm×30 m	ID 0.25 mm×30 m
Liquid phase	GS-Q (J&W)	DB-WAX (J&W)
Detector	TCD	FID
Flow rate	3 ml/min	1 ml/min
Oven temperature initial rate	393 K	308 K (Hold 5 min)
final	10 K/min	5 K/min
Injection temperature	473 K	473 K
Detector temperature	503 K	523 K

kPa corresponding to $p_e/p_e^0=0.8$ on condition that the water vapor pressure was zero. After these pure equilibria, the binary equilibria of the desired mixtures of water and ethanol vapor, i.e., $p_w=2.5$ kPa and $p_e=0.1$ – 2.1 kPa, were attained. After the water and ethanol were desorbed by vacuum drying, they were collected with a liquid nitrogen trap, and the liquid in the trap was diluted with *n*-propanol to analyze each adsorbed amount of water or ethanol with the TCD or FID of the gas chromatograph, respectively. The adsorbed amounts of the pure component were obtained based on the weighing method (Miyachi *et al.*, 1995a).

EAT The adsorption of both pure ethanol and binary water-ethanol vapors on the charcoal filter tip were carried out until their equilibria were attained. The pure adsorption during the EAT was performed under the conditions of a constant temperature of 303 K and a vapor pressure range of 0.1 to 8.4 kPa, while the binary adsorption at the same temperature was carried out under the vapor pressure conditions of 2.1 kPa for water and 0.1–2.1 kPa for ethanol to prevent condensation.

Storage tests Ethyl acetate and isopentyl alcohol were used as model flavors and applied to the tobacco column as ethanolic solutions. The loading amount was 2.5 mg for each flavor and 8 mg for ethanol. The cigarettes were prepared by connecting the flavored tobacco column to the filter tip with tipping paper after the EAT. They were stored in a sealed glass jar at a constant temperature of 295 K for over 2 weeks until an equilibrium was attained within the jar (Samejima, 1983). The amounts of water, ethanol and flavor among each material were measured by means of their desorptions as mentioned above.

Measurements of ternary adsorption equilibria Using ethyl acetate as a model flavor, the ternary adsorption equilibria of water-ethanol-ethyl acetate vapor was measured

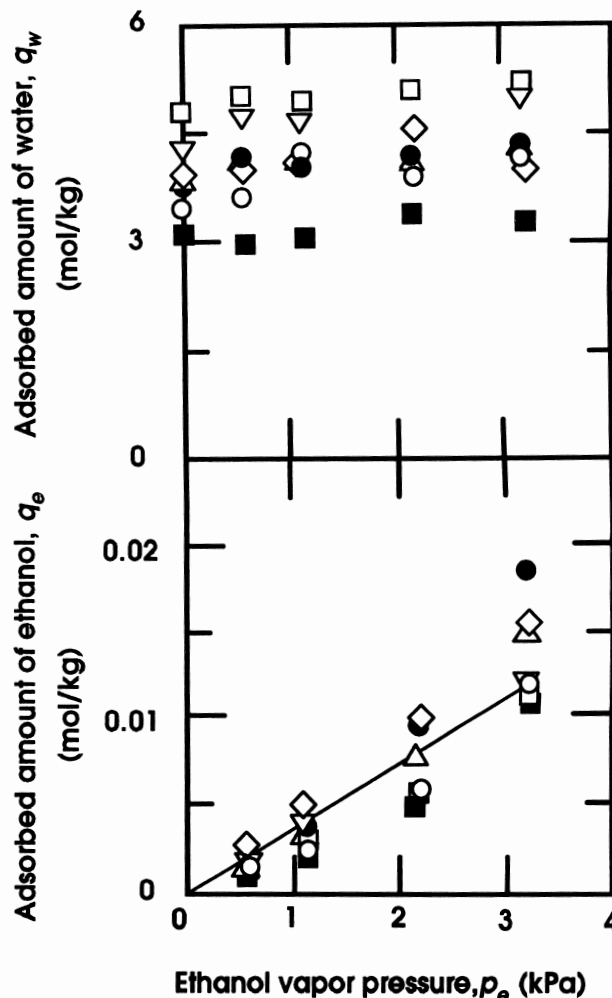


Fig. 3. Adsorbed amounts of water and ethanol for papers. ○, cigarette paper; ●, tipping paper; □, wrapping paper; ■, aluminum foil; ◇, label (hard pack); △, label (soft pack); ▽, stamp.

at a temperature of 303 K. As mentioned above, each material was pre-treated by vacuum drying and conditioned at a temperature of 303 K and $p_w=2.5$ kPa. It was then held in equilibrium with the ternary mixtures of water-ethanol-ethyl acetate vapor, i.e., $p_w=2.5$ kPa, $p_e=0.5$ or 2.1 kPa, and $p_t=0.2$ – 3.4 kPa. The adsorbed amounts were measured by means of their desorptions as mentioned above.

Results and Discussion

1. Adsorption isotherms

Paper The binary adsorption isotherms for various kind of papers are shown in Fig. 3. In the figure, the amounts of adsorbed water (q_w) and ethanol (q_e) were plotted against the various vapor pressures of ethanol (p_e). The results showed that q_w was nearly equal to the amount of pure water-vapor adsorbed at $p_w=2.5$ kPa, while q_e increased proportionally with an increase in p_e as shown as an example of a linear regression curve for cigarette paper that was expressed as follows;

$$q_e = k \times p_e \quad (1)$$

where k is an empirical constant and its obtained values for papers are listed in Table 4.

Filter The adsorption isotherms of pure ethanol or ethyl

Table 4. Empirical constants of equilibrium adsorption isotherm.

Material	Empirical constant	
Linear equation	k [mol/kg/kPa]	
Tobacco ^{a)}		
bright yellow	0.20	
Burley	0.13	
Paper		
Cigarette paper	0.04	
Tipping paper	0.05	
Wrapping paper	0.04	
Aluminum foil	0.03	
Label (soft pack)	0.04	
Label (hard pack)	0.05	
Stamp	0.04	
Filter		
Tow	0.49	
Filter	0.51	
DA equation	w_e^0 $\times 10^{-3}$ [m ³ /kg]	E [kJ/kg]
Activated carbon		
Coconut	0.42	11.4
Oil palm	0.32	11.7
Coconut in filter	0.22	11.5

^{a)} Values were obtained from the adsorption equilibrium reported in the previous paper (Nakanishi & Kobari, 1989).

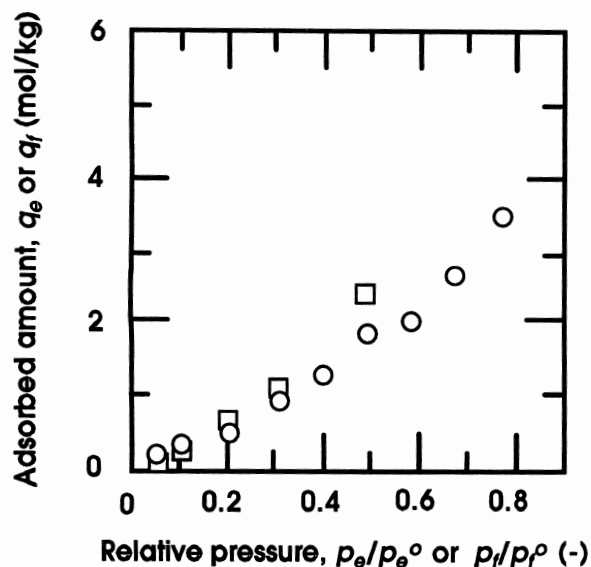


Fig. 4. Adsorbed amounts of pure component for tow. \circ , ethanol; \square , ethyl acetate.

acetate vapor for the tows are shown in Fig. 4. The adsorbed amount of ethanol was observed to be equal to that of ethyl acetate. This result can be explained in terms of ethanol adsorption on the hydrophobic sites of the acetyl group of the tow, the same as the ethyl acetate (Miyauchi *et al.*, 1995b).

The binary adsorption isotherms for the tow and filter are presented in Fig. 5 and compared with the isotherm obtained from pure ethanol vapor. The filter is molded out of the tow by adding triacetin as a plastic agent at a concentration of about 6 wt%. Because there are no marked differences between the adsorption isotherms for the tow and filter, it was found that triacetin did not affect the binary adsorption of water and ethanol vapor. The values of q_w were almost equal to that of pure water vapor adsorbed at $p_w = 2.5$ kPa, while q_e increased

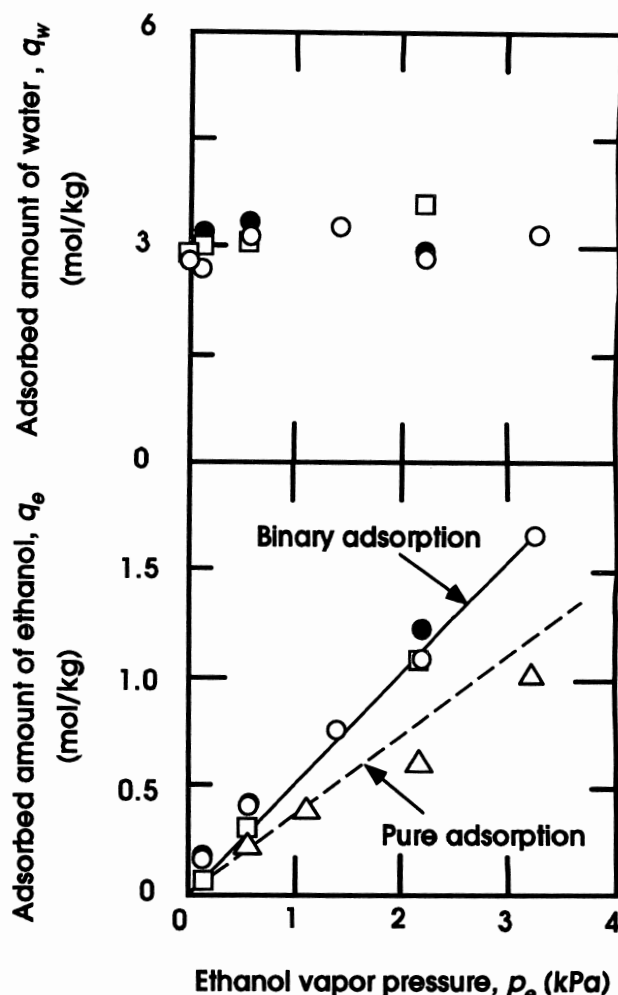


Fig. 5. Adsorbed amounts of water and ethanol for tow and filter. tow: Δ , pure adsorption; binary adsorption \circ , adsorption step; \bullet , desorption step. filter: binary adsorption \square , adsorption step.

proportionally with an increase in p_e . For the binary adsorption isotherm, the linear regression curves are presented in Fig. 5, and the values of k for empirical constants are listed in Table 4. The values of q_e from the binary vapor were always found to be greater than those from the pure ethanol vapor. This was explained by considering the adsorbed water as an agent for the structural swelling of the material, leading to its larger surface area to adsorb a relatively greater flavor component from the binary vapor. The same phenomena was also reported for the water-ethyl acetate system (Miyauchi *et al.*, 1995b). There is good agreement between the adsorption and desorption steps for ethanol; therefore, the adsorption of water and ethanol appears to be caused mainly by physical adsorption rather than by chemical reaction.

Activated carbon The adsorption isotherms of pure ethanol vapor for the activated carbons obtained from the coconut, oil palm and coconut in filters are shown in Fig. 6. Compared with the adsorption isotherms of any other adsorbate in the same figure, the pure adsorption isotherms of methanol and water vapor were also presented as solid and broken curves, respectively. The curves were obtained by applying the Dubinin-Astakhov (DA) equations to the experimental data for both adsorbates. Methanol adsorption

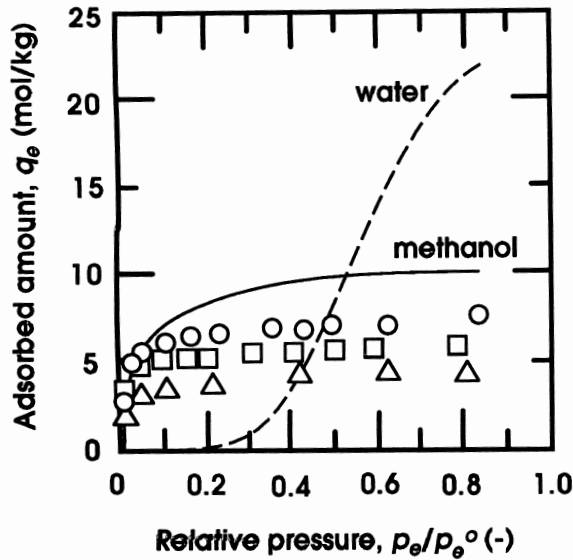


Fig. 6. Adsorbed amounts of ethanol for activated carbons. ○, coconut; □, oil palm; △, coconut in filter.

for the activated carbon, which is commonly used to measure its pore size distribution, is recognized to be caused by the existence of both the adsorption multilayer and capillary condensation phase (Urano *et al.*, 1970). As shown in Fig. 6, because the amount of adsorbed ethanol in the lower pressure range is greater than that of any adsorbate such as water which does not form the adsorption multilayer, ethanol appeared to show the same methanol adsorption behavior. This consideration demonstrated that ethanol adsorption could be explained by the same mechanism as methanol adsorption.

The binary adsorption isotherms for the activated carbons are shown in Fig. 7, compared with those obtained from adsorption of pure ethanol vapor as shown by the broken curve. As p_e increased, q_w decreased, and oppositely, q_e increased. These results were explained in terms of substitution for some adsorbed water as well as adsorption of ethanol into the micropores. The following DA equation was known to correlate with the adsorption data of organic solvents on activated carbon (Dubinin, 1960, 1967) i.e.;

$$w_e = w_e^0 \exp\left(-\left(\frac{A}{E}\right)^n\right) \quad (2)$$

$$w_e = \frac{q_e \times M}{\rho} \quad (3)$$

where w_e^0 is the micropore volume and A is the adsorption potential $[= RT \ln(p_e/p_e^0)]$. The recommended value of n is 2 for the organic solvent adsorption on activated carbon. The curves calculated from the DA equation are presented as solid curves in Fig. 7, and the empirical constants for w_e^0 and E are listed in Table 4. They appeared to correlate well with the experimental data in spite of the existence of the adsorbed water. Because the constants of w_e^0 coincided with the micropore volume (v_p) measured by the nitrogen adsorption method, the obtained DA equations were appraised to provide the verified values. The value of w_e^0 of coconut in the filter was half that of new coconut because some of the micropores in the former activated carbon were filled with

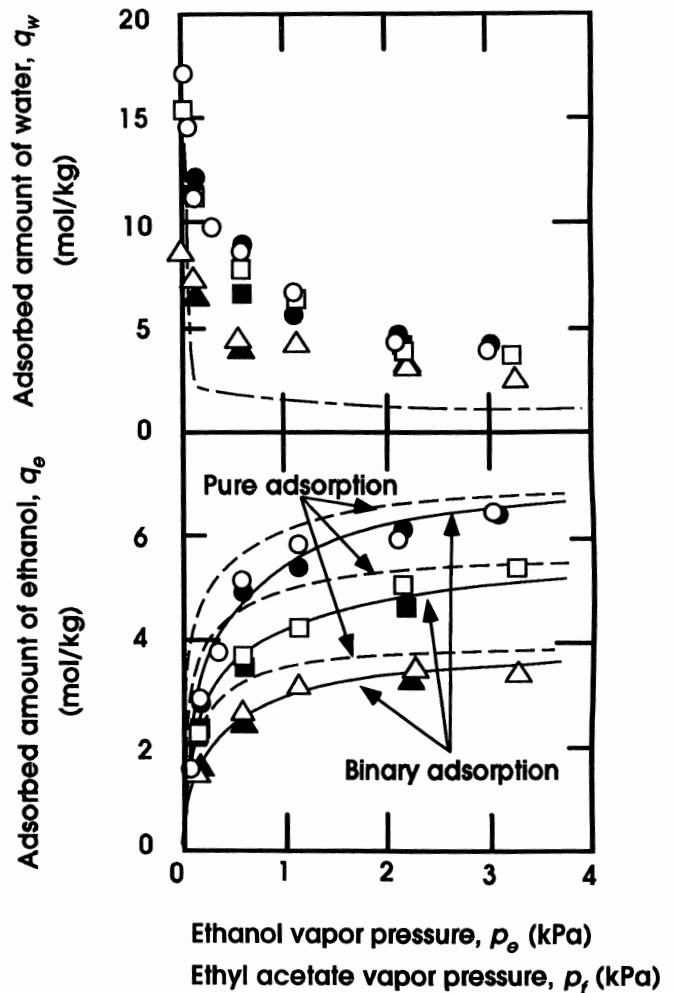


Fig. 7. Adsorbed amounts of water and ethanol for activated carbons. coconut: ○, adsorption step; ●, desorption step. oil palm: □, adsorption step; ■, desorption step. coconut in filter: △, adsorption step; ▲, desorption step. ·····, calculated for pure adsorption; —, calculated for binary adsorption; —··—, values (q_w) of water-ethyl acetate system (Miyachi *et al.*, 1995b).

triacetin added in the process of molding the filter. The value of q_e for the binary vapor was less than that for the pure one. Further, q_w for the water-ethanol system was found to be greater than that for the water-ethyl acetate system, which is shown as a dotted curve in Fig. 7 (Miyachi *et al.*, 1995b). Therefore, the effect of adsorbed water on the ethanol adsorption was confirmed to be impossible to disregard.

Because there was no significant difference in q_w as well as q_e between adsorption and desorption steps, the physical adsorption process seemed to proceed in both steps.

2. Concepts of the EAT

Selection of agent and treatment method The amount of adsorbed water and ethanol on the filter tip after the EAT is shown in Fig. 8. In the figure, the amounts of adsorbed water (WW_{fil}) and ethanol (WE_{fil}) within the filter tip are plotted against the various vapor pressures of ethanol (p_e). Existing data on WW_{fil} within the filter tip before and after connecting it with the tobacco column are also presented. If pure ethanol was selected as an agent of the EAT, it would easily cause the desorption of water within the filter tip, and

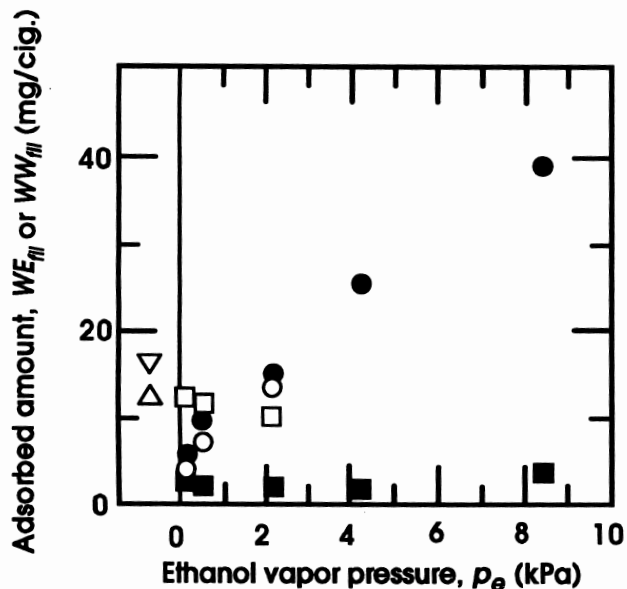


Fig. 8. Adsorbed amounts of the charcoal filter after the Ethanol Adsorption Treatment. ethanol: ●, pure adsorption; ○, binary adsorption. water: ■, pure adsorption; □, binary adsorption value (q_w) within the filter tip used in commercial process. ▽, before connecting the filter tip with the tobacco column. △, after connecting the filter tip with the tobacco column. EAT conditions: Temperature: 303 K. pure adsorption: ethanol vapor pressure, 0.1, 0.5, 2.1, 4.2, 8.4 kPa. binary adsorption: water vapor pressure, 2.1 kPa ethanol vapor pressure, 0.1, 0.5, 2.1 kPa.

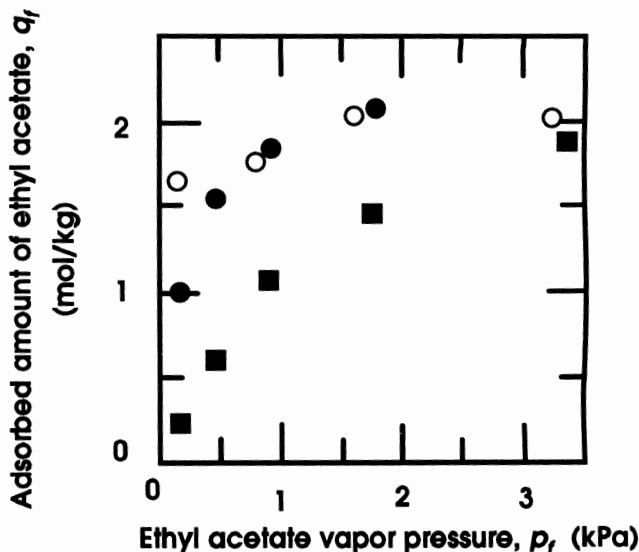


Fig. 10. Adsorbed amounts of ethyl acetate from water-ethanol-ethyl acetate mixtures for coconut activated carbon in filter. ethanol vapor pressure: ○, 0 kPa; ●, 0.5 kPa; ■, 2.1 kPa.

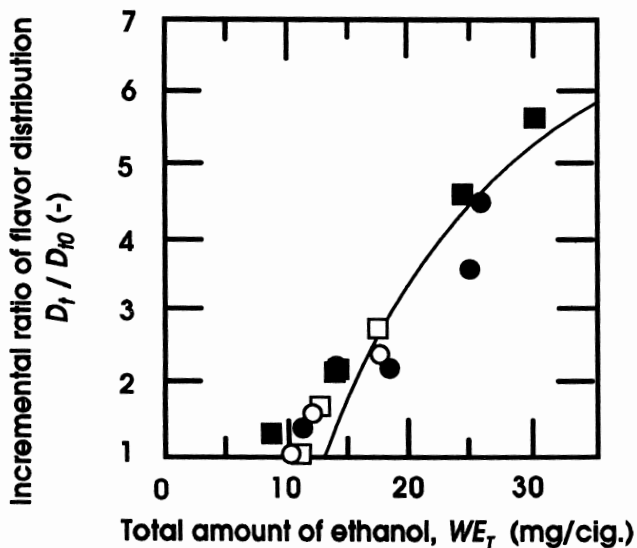


Fig. 9. Effect of total amount of ethanol on ratio of flavor distribution. pure adsorption treatment: ●, ethyl acetate; ■, isopentyl alcohol. binary adsorption treatment: ○, ethyl acetate; □, isopentyl alcohol. —, calculated for ethyl acetate.

then WW_{fil} would approach a value of nearly zero. This result indicated that undesirable migration of water from the tobacco column to the filter tip takes place during storage and marketing. In contrast, if the binary adsorption treatment of water and ethanol was selected, the amount of water was controlled at its desirable level, corresponding to the water content within the practical filter tip after its connection. Furthermore, both the amounts of adsorbed ethanol within

the filters as well as its vapor existing in the spaces between the cigarettes increased. These results lead to the fundamental concept of the EAT, and the application of binary adsorption of water and ethanol vapor was found to be desirable for the EAT.

Effect of ethanol concentration on flavor distribution ratio
The ratio of flavor distribution within the tobacco column (D_t) is defined by the following technical term.

$$D_t = \frac{WF_t}{WF_t + WF_{fil}} \quad (4)$$

where WF_t and WF_{fil} are the amounts of flavor within the tobacco column and the filter tip, respectively. The ratio (D_t/D_{t0}) was represented in Fig. 9 as a function of the total amount of ethanol (WE_T) which was the sum of the ethanol amounts added in the flavoring and the EAT process. The value of D_{t0} means the ratio of flavor distribution when ethanol does not exist in the product. As shown in Fig. 9, the ratio (D_t/D_{t0}) increased with an increasing amount of ethanol added to the filter tip, indicating the decreased flavor migration due to the EAT. Based on these results, the amount of flavor within the tobacco column for the condition of the EAT of $WE_T = 16$ mg, which corresponds to a p_e of 0.6 kPa, was found to be twice that without the EAT.

Evaluation of the EAT From the measurements of the ternary adsorption isotherms of the water-ethanol-ethyl acetate system, it was found that ethanol exerted a great influence on the adsorption of ethyl acetate as a model flavor onto activated carbons, demonstrating a negligibly small effect on the tobacco as well as the filters. The ternary adsorption isotherms of ethyl acetate for the activated carbon are shown in Fig. 10. As shown in the figure, the value of q_f decreased by both decreasing the vapor pressure of ethyl acetate (p_f) and increasing that of ethanol (p_e). Here, Lewis *et al.* (1950) presented the following equation to correlate the quantities of a hydrocarbon gas mixture adsorbed on silica gel and activated carbon;

$$\sum_1^n \left(\frac{q_i}{q_i^0} \right) = 1 \quad (5)$$

where q_i^0 is the adsorbed amount of the pure component corresponding to the total pressure ($\sum p_i$). This correlation was valid for our experimental concentration range.

Further, the ratio of flavor distribution (D_t/D_{t0}) was estimated based on the following assumptions and using the adsorption isotherms obtained above and Eq. (5). The assumptions are as follows; (a) the effect of adsorbed water on the adsorption of ethyl acetate for activated carbon was negligible (Miyachi *et al.*, 1995b), (b) the distribution of ethanol could be evaluated by the binary adsorption isotherms of ethanol and water vapor, and (c) ethanol exerts no influence on the adsorption of ethyl acetate on tobacco and the filter as mentioned above. Thus the calculation proceeded as follows;

(1) According to assumption (b), q_e for each material at the specific p_e was calculated utilizing the binary adsorption isotherm of water and ethanol vapor, and the values of WE_T were given by the sum of q_e ($WE_T = \sum q_e$). The values of p_e and q_e among each material were then obtained relative to the various WE_T .

(2) The value of D_t where p_w and p_e were specified was estimated as follows. The total vapor pressure ($\sum p_i = p_w + p_e + p_f$) was calculated by assuming the p_f of ethyl acetate. The quantities of q_i^0 for the activated carbon corresponding to the total vapor pressure were obtained by DA equations for pure water, ethanol and ethyl acetate. However, the DA equation for ethyl acetate obtained from the binary adsorption isotherm was used according to assumption (a). Because the value of (q_w/q_w^0) was assessed to be 0.1 from the ternary adsorption equilibria, q_f of ethyl acetate for the activated carbon was then calculated using Eq. (5). Ignoring the ethanol vapor according to assumption (c), the q_f of ethyl acetate for the tobacco, papers and filter was given from the p_f of ethyl acetate by utilizing the binary adsorption isotherm of water and ethyl acetate vapor (Miyachi *et al.*, 1995b). WE_T was then estimated from the sum of q_f of ethyl acetate for the tobacco, papers, filter and activated carbon. The assumption of p_f of ethyl acetate was repeated until WE_T was equal to the initial values (2.5 mg). By satisfying this limitation, the value of D_t at the specific p_e was obtained by Eq. (4).

(3) This procedure (2) was carried out within the whole range of our experimental ethanol vapor pressure.

The calculated values of (D_t/D_{t0}) are shown as a solid curve in Fig. 9. The calculated values show good agreement with the experimental ones; therefore, the validity of the assumptions was certified. These results lead to the conclusion that the depression of flavor migration in the activated carbon was caused by increasing the ethanol vapor pressure.

Ethanol migration In Fig. 11 the amounts of ethanol within the tobacco column (WE_t) as well as the filter tip (WE_{ft}) for the cigarettes after their storage tests are plotted against the total amount of ethanol (WE_T). In the calculation, the amount of ethanol for each material was determined against p_e , using the binary adsorption isotherms obtained, and these values were then grouped and summed to estimate the amounts of ethanol contained in the tobacco columns (WE_t), the filter tips (WE_{ft}) and the whole products or

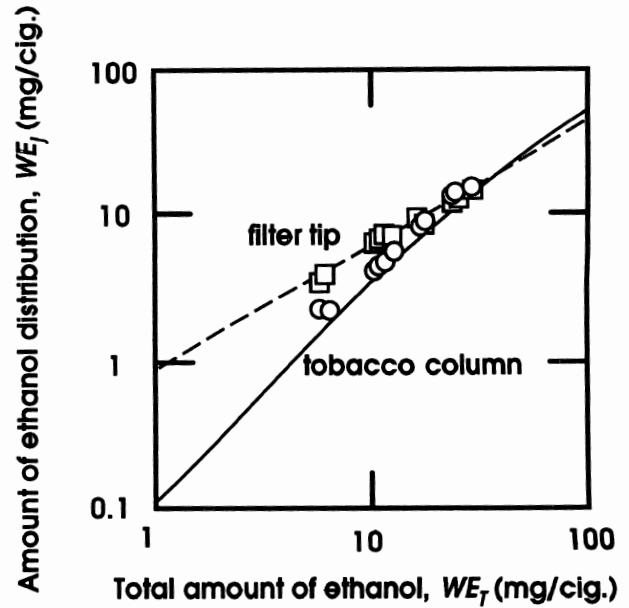


Fig. 11. Ethanol distribution between tobacco column and filter tip as a function of total amount of ethanol. \circ , tobacco column; \square , filter tip. —, calculated for tobacco column; ·····, calculated for filter tip.

cigarettes (WE_T). The calculated values of WE_t or WE_{ft} were plotted against WE_T as a solid or broken curve, respectively.

Because no marked differences were found between the experimental and calculated values, the equilibrium distribution of ethanol was recognized to be capable of being evaluated by the binary adsorption isotherms of water and ethanol. The calculation of flavor distribution indicated that p_f ranged in the lower level of one-tenth that of p_e ; therefore, the ethanol distribution was almost independent of the flavor vapor. Therefore, this condition between p_f and p_e in the spaces between the cigarettes led to the effective depression of flavor migration.

Nomenclature

A	=free energy of adsorption [J/mol]
D	=ratio of flavor distribution [—]
E	=characteristic free energy of adsorption [J/mol]
k	=constant in Eq. (1) [mol/kg/kPa]
M	=molecular weight [kg/mol]
n	=constant in Eq. (2) [—]
p	=vapor pressure [Pa]
p^0	=saturated vapor pressure [Pa]
q	=amount of adsorbed component [mol/kg]
q^0	=amount of adsorbed component, as in pure isotherm [mol/kg]
R	=gas constant [J/K/mol]
s	=specific surface area [m ² /kg]
T	=temperature [K]
v_p	=micropore volume [m ³ /kg]
WE	=amount of ethanol [mg/cigarette]
WF	=amount of flavor [mg/cigarette]
WW	=amount of water [mg/cigarette]
w	=volume of adsorbed component [m ³ /kg]
w_e^0	=constant in Eq. (2) (=micropore volume) [m ³ /kg]

ρ =density [kg/m³]

<Subscripts>

e =ethanol
 f =ethyl acetate
 fil =filter tip
 i =species in binary or ternary component system
 j =part of cigarette (tobacco column or filter tip)
 T =total (=the sum of tobacco column and filter tip)
 t =tobacco column
 w =water

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