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Chen-Chia Huang[ab]; Yann Ching Lin[b]; Fang Cheng Lu[b]
[a] Department of Chemical Engineering, National Yunlin University of Science & Technology, Touliu, Yunlin, Taiwan, REPUBLIC OF CHINA [b] DEPARTMENT OF APPLIED CHEMISTRY, CHUNG CHENG INSTITUTE OF TECHNOLOGY, TAIWAN, REPUBLIC OF CHINA

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Dynamic Adsorption of Organic Solvent Vapors onto a Packed Bed of Activated Carbon Cloth

CHEN-CHIA HUANG,* YANN CHING LIN, and FANG CHENG LU
DEPARTMENT OF APPLIED CHEMISTRY
CHUNG CHENG INSTITUTE OF TECHNOLOGY
TASHI, TAOYUAN, 335, TAIWAN, REPUBLIC OF CHINA

ABSTRACT

The adsorption behavior of organic compound vapors onto a packed bed of activated carbon cloth (ACC) has been investigated. Three types of ACCs have been employed: KF1500, FT200-20, and E-ACC. The volatile organic compounds (VOCs) used in this study are acetone, dichloromethane, acrylonitrile, and *n*-hexane. The operating parameters studied are temperature of adsorber, weight of ACC, relative humidity of fluid, inlet concentration of VOCs, and total volumetric flow rate of gas stream. A simple theoretical model, originally introduced by Yoon and Nelson, has been utilized to simulate the breakthrough curve of VOC vapor on an adsorption column packed with activated carbon cloth. A modified model is proposed to predict the adsorption behavior of an adsorber at different temperatures.

Key Words. Activated carbon cloth; Volatile organic compound; Packed bed; Adsorption; Breakthrough curve

INTRODUCTION

Volatile organic compounds (VOCs), commonly existing in indoor and outdoor air, may cause human health risks. The treatment methods presently taken for controlling VOCs emission are incineration, condensation, absorption, and adsorption (1). The adsorption method is frequently used for treatment of VOCs, particular for the low concentration range. Commonly used

*To whom correspondence should be sent at his present address: Department of Chemical Engineering, National Yunlin University of Science & Technology, Touliu, Yunlin, Taiwan, Republic of China.

porous adsorbents in industrial applications are activated carbon, silica gel, activated alumina, zeolites, and molecular sieves. Activated carbon has several specific characteristics compared to other types of adsorbent, such as a pore size that is distributed over a wild range. Activated carbon has been applied to many fields for a long time. There are four types of activated carbons in commercial use: powdered activated carbon (PAC), granular activated carbon (GAC), carbon molecular sieve (CMS), and activated carbon cloth (ACC) (2). ACCs have been on the market since the beginning of the 1960s. ACCs have unique characteristics compared with granular and powder adsorbents. According to literature (3, 4), ACCs have the advantages of fast adsorption rate, low resistance, ease of handing, and ability to conduct an electrical current. ACCs have recently become the most interesting adsorbents, and they are widely used in industry. Suzuki (4) reported that ACCs are now commercially marketed by five different corporate groups in Japan. The total amount of production was more than 200 tons in 1992. Commercial activated charcoal cloths are prepared from carbon-containing materials such as rayon fiber, polyacrylonitrile fiber, phenolic fiber, and coal tar pitch.

Although ACCs have been widely used in many fields, few papers have discussed the dynamic adsorption behavior of an adsorber packed with ACCs. The goal of this paper is to discuss the dynamic adsorption behavior of organic solvent vapors onto a packed bed of activated carbon cloth. The theoretical model introduced by Yoon and Nelson was used to simulate the experimental breakthrough data. A modified model is proposed to predict breakthrough behavior of the adsorption column at different operating temperatures.

MATHEMATICAL MODEL

Yoon and Nelson (5, 6) postulated that when a gas stream flows through a fixed bed of activated carbon, some of gas molecules are adsorbed on activated sites and others pass through the activated carbon. Because activated sites are finite, activated carbon can not adsorb again after becoming saturated by gas molecules. Therefore, Yoon and Nelson further postulated that the rate of decrease in the probability of adsorption for each molecule is proportional to the probability of adsorption, Q, and the probability of breakthrough of contaminant, P. That is

$$-\frac{dQ}{dt} \propto QP \tag{1}$$

By experimental evidence, the rate of decrease in the probability of adsorption is directly proportional to the contaminant concentration (C) and the flow rate of the gas stream (F), and inversely proportional to the weight of the ac-

tivated carbon (W_c). Thus,

$$-\frac{dQ}{dt} \propto \frac{CF}{W_c} QP \tag{2}$$

In addition, the number of activated sites is proportional to the weight of the activated carbon (W_c) and also the adsorption capacity of activated carbon (W_e). Equation (2) is rewritten as

$$-\frac{dQ}{dt} = k\frac{CF}{W_e} QP \tag{3}$$

where k is a constant of proportionality.

Since $Q = 1 - P$, Eq. (3) may be rewritten as

$$\frac{dP}{dt} = k'(1 - P)P \tag{4}$$

where $k' = kCF/W_e$. Equation (4) may be integrated to give

$$\ln \frac{P}{1 - P} = k'(t - \tau) \tag{5}$$

where τ is the time required to obtain 50% breakthrough. Equation (5) may be rearranged as a form of breakthrough time

$$t = \tau + \frac{1}{k'} \ln \frac{P}{1 - P} \tag{6}$$

The probability of breakthrough of contaminant, P, is defined as Y_{out}/Y_{in}, in which Y_{out} expresses the concentration of effluent and Y_{in} denotes the concentration of contaminant in the inlet stream. Both Y_{out} and Y_{in} are obtained from experiments. The constants k' and τ are determined by the slope (inverse) and ordinate intercept, respectively, from the plot of t vs $\ln[P/(1 - P)]$.

If k' or C is not large, it can be assumed that the shape of breakthrough curve is symmetric. The saturation adsorption capacity, W_e, is equal to the product of $CF\tau$. Therefore,

$$k' = k/\tau \tag{7}$$

where k is a constant. For a given adsorbate and specified type of activated carbon, k is independent of contaminant concentration and flow rate.

Yoon and Nelson (6) postulated that the adsorptive reaction is an nth-order reaction. They chose an nth-order reaction rating based on chemical kinetic principles. In addition, the half-life of the reactant corresponds to the 50%

breakthrough time, τ. Finally, Yoon and Nelson obtained

$$\tau = \frac{k_w}{C^{n-1}F} \tag{8}$$

where k_w is a proportionality constant. Equations (6), (7), and (8) can be used to quickly model and predict the breakthrough curve of a system with various inlet concentrations and volumetric flow rates of the gas stream. Only two sets of experimental breakthrough data at various inlet concentrations are required.

Yoon and Nelson (7) considered the effect of relatively humidity and then modified the previous theoretical model. The breakthrough percentage is calculated by

$$P = \frac{1}{1 + C^{A - k'' \ln(W_a + t)}} \tag{9}$$

where A, k'', and W_a are all constants and may be obtained by nonlinear regression of the Gauss–Newton method (8). In this study the least-squares method (9) was used to determine k'', A, and W_a.

The theoretical models proposed by Yoon and Nelson were all derived at a constant temperature. Dynamic gas adsorption, however, is usually operated under various temperatures. In order to widen the use of the theoretical model, the Arrhenius' equation is introduced to substitute for the rate constant k_w:

$$k_w = k_0 e^{-E/RT} \tag{10}$$

where k_0 is a constant, E is the activation energy, R is the gas constant, and T is the absolute temperature. Substitution of this expression for k_w into Eq. (8) gives

$$\tau = \frac{k_0 e^{-E/RT}}{C^{n-1}F} \tag{11}$$

Equations (6), (7), and (11) can be used to quickly simulate and predict the breakthrough curves for various operating temperatures. Therefore, only three different sets of experimental breakthrough data are required.

EXPERIMENTAL

Three types of adsorbents were used: KF1500 ACC manufactured by the Toyobo Chemical Co., Japan; FT200-20 ACC by the Kurray Chemical Co., Japan; and E-ACC ACC by the Charcoal Cloth Chemical Co., England. The characteristics and physical properties of the ACCs are listed in Table 1. Four organic compounds, acetone, dichloromethane, acrylonitrile, and *n*-hexane,

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TABLE 1
Physical Properties of Activated Carbon Cloth

	Activated carbon cloth		
	KF1500	E-ACC	FT200-20
Raw material	Viscose rayon	—	Phenolic resin
Shape	Felt	Woven form	Felt
Surface area (BET) (m^2/g)	1499	1103	2431
Pore volume (cm^3/g)	0.630	0.547	0.880
Mean pore radius (nm)	0.840	0.990	0.724

were investigated as adsorbates. All were more than 99.5% pure (tested grade).

The adsorption vessel was fabricated from an 18.2-cm long, 1.7-cm inside diameter stainless steel pipe. The schematic experimental flow diagram is shown in Fig. 1. In order to remove impurities, the ACCs were put in a vacuum oven and degassed by heating at 383 K for 24 hours. Layers of ACC were packed in the adsorption column at a fixed packed density. The number of layers and the packed weight of ACCs are listed in Table 2. The temperature of the thermostat into which the adsorption column was placed was controlled at 303 K. A multiple-channels mass flow controller (MKS 247C) was employed to regulate the flow rate of nitrogen at each channel. The VOC and water vapors were obtained by branches of nitrogen streams which flowed through a series of bottles of VOC and water in a thermostat. The inlet concentration of VOC and the relative humidity of gas streams were then set before the start of the experiment. A Shimadzu 14-A Gas Chromatograph with a flame ionization detector (FID) was used to analyze the effluents of the adsorber.

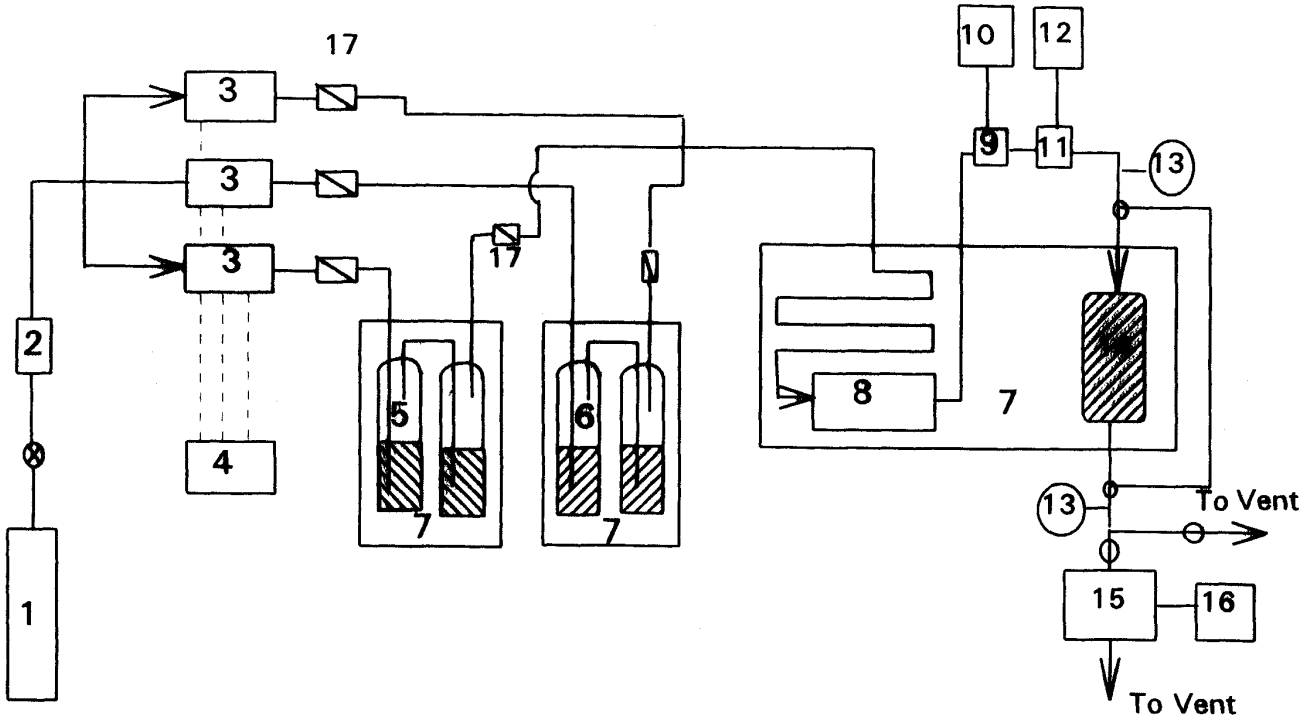
RESULTS AND DISCUSSION

Total adsorption capacity W_e is first defined as

$$W_e = \frac{FY_{in}}{24.9} \int_0^{t_e} \left(1 - \frac{Y_{out}}{Y_{in}}\right) dt \tag{12}$$

where t_e is the complete breakthrough time ($Y_{out}/Y_{in} = 1$), Y_{out} is the concentration of the effluent, Y_{in} is the inlet concentration, and F is the volumetric flow rate. The breakthrough time (t_b) is defined the time when $Y_{out}/Y_{in} = 1\%$.

The operating conditions were set as follows except for the ones specified as independent variables. The temperature of the adsorption column (T) was controlled at 303 K, the relative humidity (RH) of the fluid was controlled to



1. Carrier gas(N_2)
2. Dehumidifier
3. Flow meter
4. Flow rate controller
5. Water saturator
6. VOC saturator
7. Thermostat
8. Mixer
9. Thermocouple
10. Thermometer
11. Humidity Probe
12. Humidity indicator
13. Pressure gauge
14. Adsorber
15. G.C.
16. G.C. recorder
17. Check valve

FIG. 1 Schematic diagram of dynamic experiment apparatus.

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TABLE 2
Physical Properties of Adsorption Column Packed with Activated Carbon Cloth

	Activated carbon cloth						
	KF1500			E-ACC			FT200-20
Number of layers	15	30	45	70	100	130	30
Packed density (g/cm^3)	0.1	0.1	0.1	0.35	0.35	0.35	0.07
Height (cm)	3.6	6.8	10.0	2.4	3.2	4.5	4.0
Weight (g)	1.36	2.41	3.82	3.18	4.31	6.03	1.05

be less than 1%, the volumetric flow rate (F) was set at 1 dm^3/min, the inlet concentrations of VOCs (Y_{in}) were set to be 1.71% (acetone), 2.15% (dichloromethane), 1.67% (acrylonitrile), and 1.37% (*n*-hexane), respectively.

Experimental data and simulation of the first and second runs (after regeneration) of the adsorption of acetone onto FT200-20 ACC are presented in Fig. 2. After saturation by VOC, the packed bed of ACC was regenerated by nitrogen at 333 K. It is apparent from Fig. 2 that the two breakthrough curves are almost identical. This implies that the FT200-20 ACC adsorbed by acetone can be regenerated by using a hot nitrogen purge. Huang et al. (10) reported that the activated carbon adsorbed by acetone vapor can be sufficiently re-

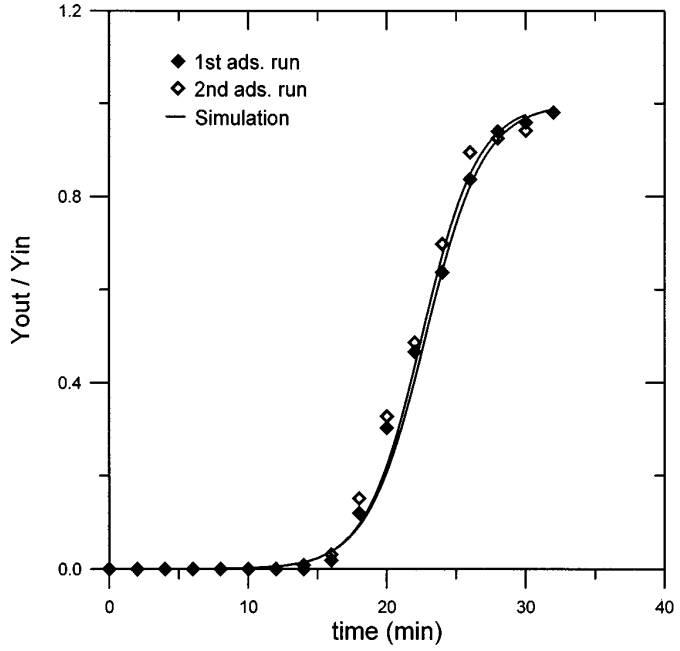

FIG. 2 Comparison of breakthrough data of first and second adsorption runs of acetone on FT200-20 ACC adsorber. $Y_{in} = 1.71\%$, $T = 303$ K, RH $< 1\%$, 30 layers.

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generated by using a hot nitrogen purge (423 K). The above-mentioned model (Eq. 6) was utilized to simulate the breakthrough curves of the adsorption column. The simulation result was very good.

The temperature effect on the breakthrough curve of the ACC adsorber is shown in Fig. 3. The operating temperatures (adsorption column temperature) were set at 298, 318, and 333 K. The breakthrough times read from Fig. 3 are 15, 10, and 6 minutes, respectively. Apparently the breakthrough time decreases with increasing bed temperature. The adsorption behavior of VOCs onto ACC could be considered to be physical adsorption. The capacity of an adsorbent by physical adsorption decreases with increasing temperature. The same result was obtained by when VOC vapors were adsorbed onto an activated carbon bed (10). As shown in Fig. 3, Eq. (6) performed very well when applied to simulate breakthrough curves at the three different temperatures.

Experimental breakthrough data of acrylonitrile adsorbed onto a fixed-bed packed with different weights of E-ACC ACC are shown in Fig. 4. It is observed that the breakthrough time is longer for heavier ACC packings. The shapes of three breakthrough curves seem to be similar, and the breakthrough time seems to be proportional to the weight of ACC. This indicates that the mass transfer zone of VOC onto an ACC column is a constant pattern (11). Figure 5 shows a plot of the weight of ACC (W_c) versus the breakthrough time.

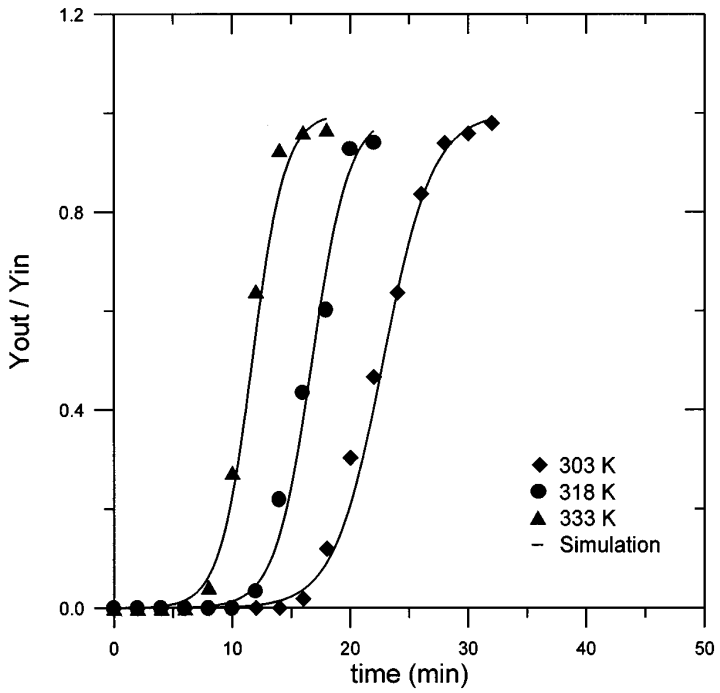

FIG. 3 Effect of temperature on breakthrough curve of acetone adsorbed onto FT200-20 adsorber. $Y_{in} = 1.71\%$, RH $< 1\%$, 30 layers.

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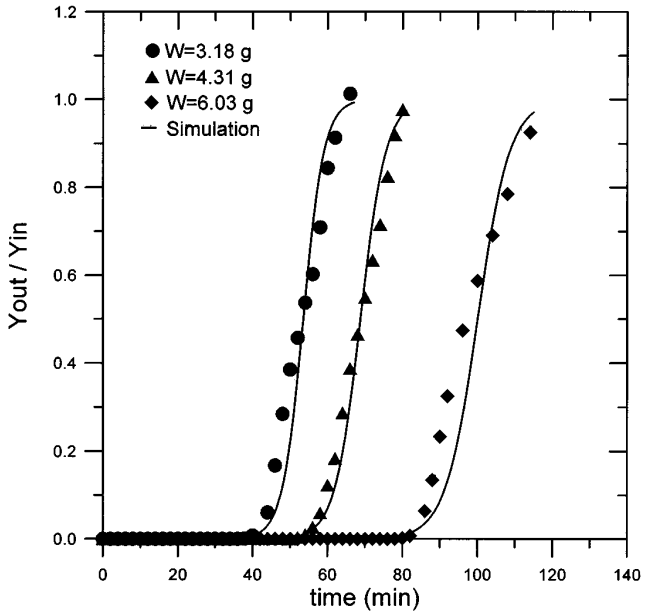

FIG. 4 Comparison of breakthrough curve of acrylonitrile onto adsorber packed with different weights of E-ACC ACC. $Y_{in} = 1.67\%$, $T = 303$ K, RH $< 1\%$.

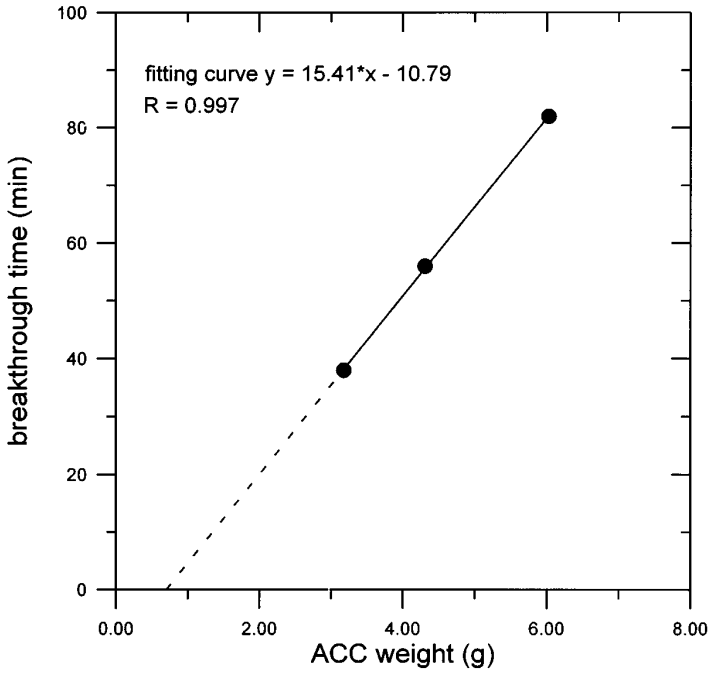

FIG. 5 Relationship between breakthrough time and the packed weight of E-ACC ACC.

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A straight line is obtained by linear regression. The intercept of the straight line on the abscissa is 0.7 ACC. In other words, acrylonitrile (1.67%) will immediately pass through if a 1.7-cm diameter column is packed with fewer than 17 layers of E-ACC ACC.

In order to investigate the effect of relative humidity on the ACC adsorption process, the relative humidity of the gas stream was designated to be 1, 60, and 90%. Figure 6 shows experimental breakthrough data of acrylonitrile adsorbed onto the E-ACC ACC bed at three different relative humidity values. Because the active sites of ACC may be occupied by water molecules, the breakthrough time decreases with an increase in the relative humidity of the fluid. The result observed in Fig. 6 is similar to that found by using activated carbon (11). Equation (9) gave good results when it was used to simulate the breakthrough curve of E-ACC ACC-adsorbed acrylonitrile at three different relative humidity values.

Figure 7 illustrates experimental breakthrough data of acetone adsorbed onto KF1500 ACC at different inlet concentrations of the gas stream. It is observed that the breakthrough times are 28 and 49 minutes when the inlet concentrations are 1.71 and 0.96%, respectively. The breakthrough time decreases with increasing inlet concentration of fluid at the same flow rate. Because more VOC molecules per unit time pass through the packed bed at a higher feed concentration, the ACC becomes saturated sooner.

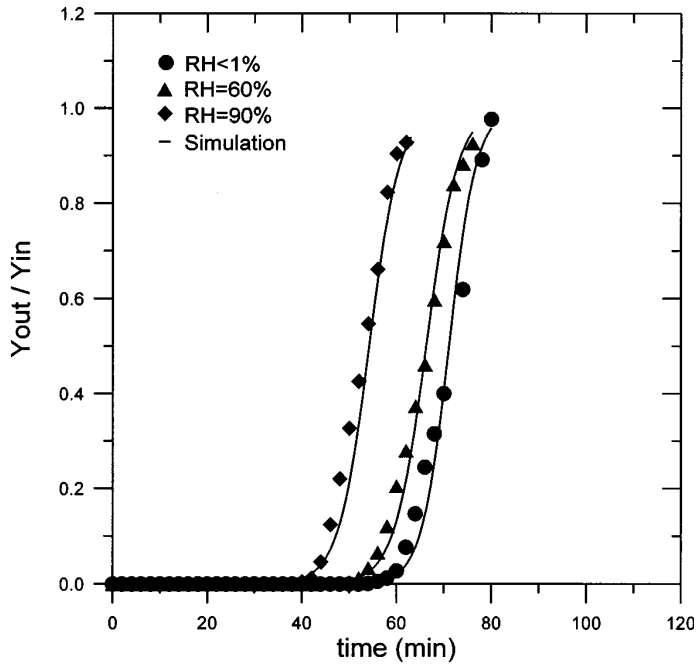

FIG. 6 Effect of relative humidity on breakthrough curve of acrylonitrile on an adsorber packed with E-ACC ACC. $Y_{in} = 1.67\%$, $T = 303$ K, 100 layers.

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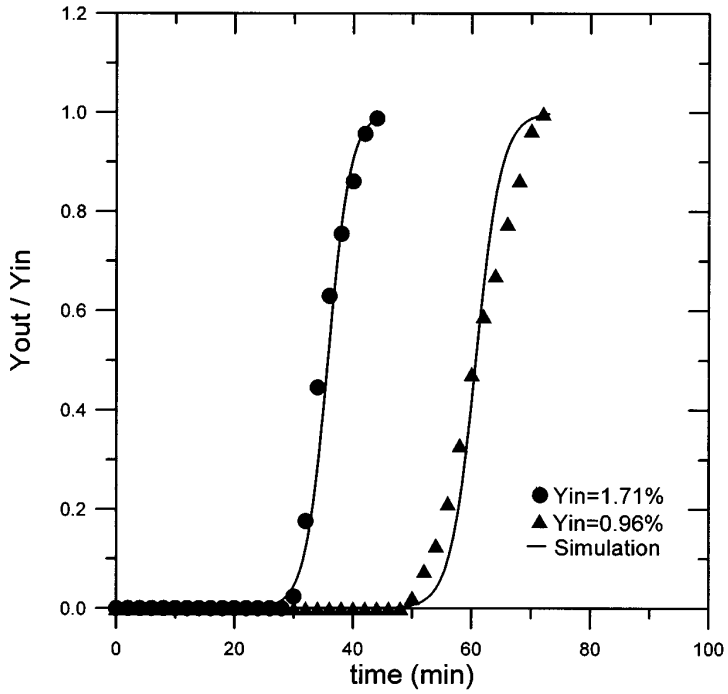

FIG. 7 Effect of inlet concentration on breakthrough curve of acetone on an adsorber packed with KF-1500 ACC. $T = 303$ K, RH $< 1\%$, 30 layers.

The effect of volumetric flow rate on the breakthrough curve of acrylonitrile adsorbed onto the packed bed of KF1500 ACC is shown in Fig. 8. The volumetric flow rates studied were 1 and 2 dm^3/min. The linear flow rates are 4.4 and 8.8 cm/s, respectively, which are in the range used for industrial operations. As expected, the breakthrough time is inversely proportional to the flow rate. According to theory, this does not affect the adsorption capacity because the fluid remains in the ACC bed longer than the time of reaction. Stern (12) reported that the flow rate does not affect adsorption performance at a flow rate of 1 dm^3/min. From Fig. 8 it is found that the breakthrough times are 42 and 19 minutes for flow rates of 1 and 2 dm^3/min, respectively. In addition, the adsorption capacities of these two runs are obtained by Eq. (12) to be 1.125 and 1.191 g, respectively. The difference in adsorption capacities between the two flow rate runs is only 5%.

The experimental breakthrough data of acetone adsorbed onto different types of ACCs are compared in Fig. 9. The adsorption capacities of acetone onto a unit mass of the three ACC adsorbents are 0.285 g/g (E-ACC), 0.377 g/g (KF1500), and 0.493 g/g (FT200-20). As shown in Table 1, the BET surface area of the three ACC adsorbents is in the order FT200-20(2431 m^2/g) > KF1500 (1499 m^2/g) > E-ACC (1103 m^2/g). It was found that the adsorption capacity per unit weight of adsorbent directly depends on the BET surface area of ACC.

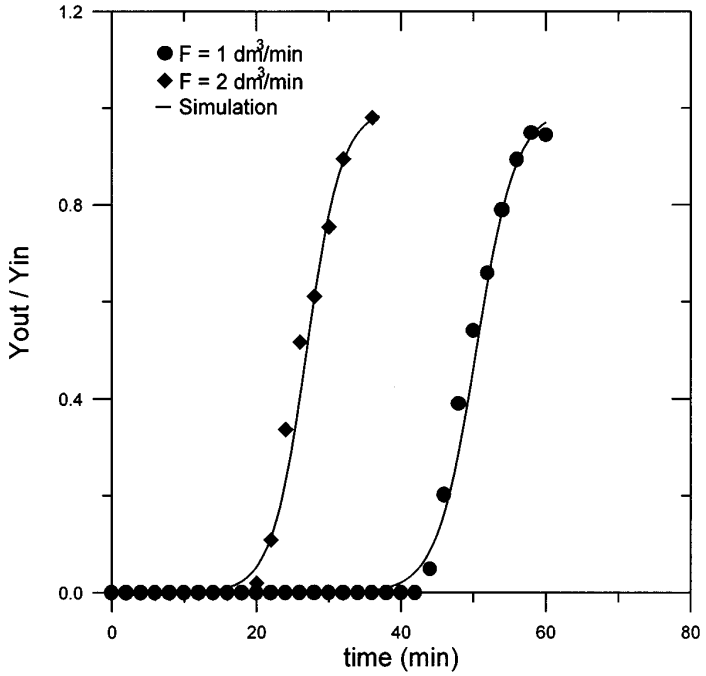

FIG. 8 Effect of flow rate on breakthrough curve of acrylonitrile on an adsorber packed with KF-1500 ACC. $Y_{in} = 1.67\%$, $T = 303$ K, RH $< 1\%$, 30 layers.

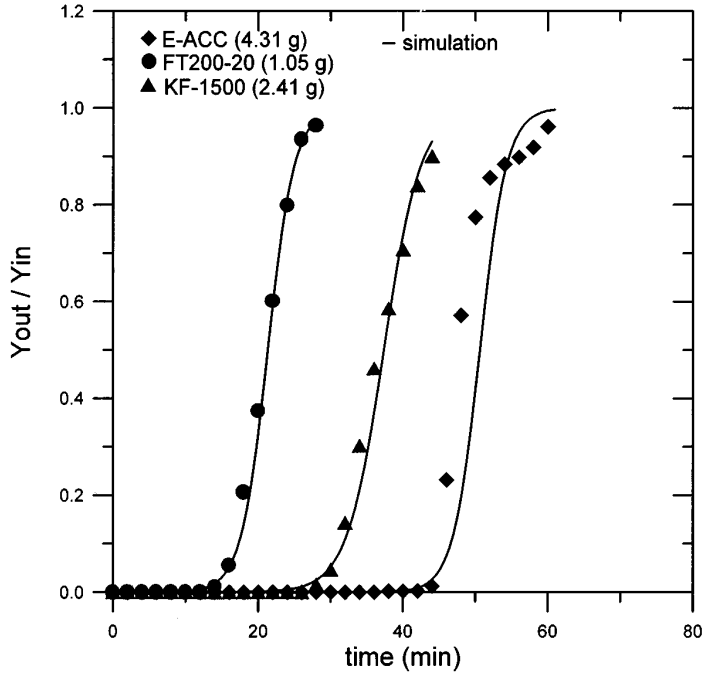

FIG. 9 Comparison of breakthrough curve of acetone on an adsorber packed with different ACCs. $Y_{in} = 1.71\%$, $T = 303$ K, RH $< 1\%$.

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TABLE 3
Values of Simulation Parameters for Adsorption of VOCs onto KF1500 ACC Adsorbents

	n	k_w	k
Acetone	2.08	0.32	17.02
Dichloromethane	1.72	1.66	14.56
n-Hexane	2.08	0.32	16.75
Acrylonitrile	—	—	15.71

As shown in the aforementioned figures, the theoretical model introduced by Yoon and Nelson simulates the experimental breakthrough data very well. The parameters obtained for the three different VOCs are listed in Table 3. The given parameters can be used to predict the breakthrough curves of an ACC adsorber under different inlet concentrations and flow rates. Figure 10 presents the experimental data and simulation results of *n*-hexane adsorbed onto KF1500 ACC at different inlet concentrations (Runs 44 and 48). Obviously, simulation results agree well with the experimental data. The parameters k, k_w, and n, obtained by the simulation of Runs 44 and 48, were substi-

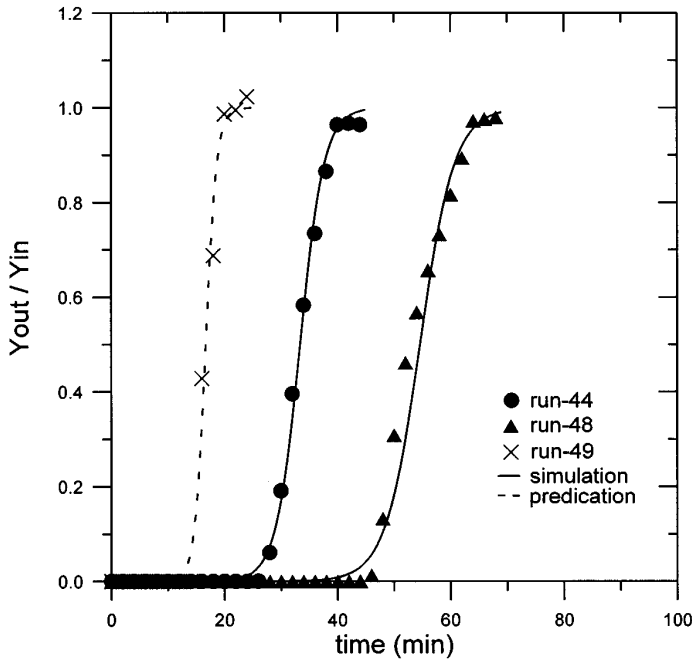

FIG. 10 Simulation and prediction of breakthrough curve of n-hexane on an adsorber packed with KF-1500 ACC at different inlet concentrations and flow rates. Run 44: $Y_{in} = 1.37\%$, $F = 1$ dm^3/min. Run 48: $Y_{in} = 0.87\%$, $F = 1$ dm^3/min. Run 49: $Y_{in} = 1.37\%$, $F = 2$ dm^3/min.

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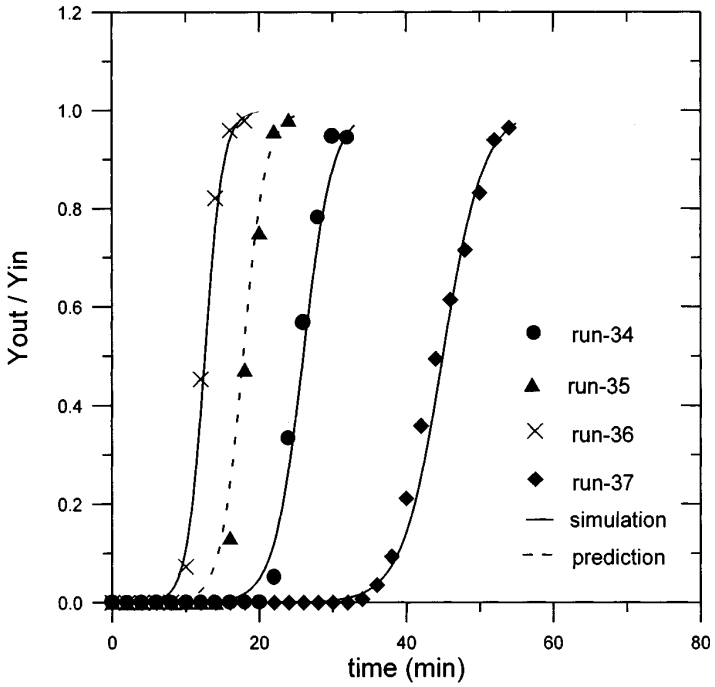

FIG. 11 Simulation and prediction of breakthrough curve of dichloromethane on an adsorber packed with KF-1500 ACC at different temperatures. Run 34: $T = 303$ K, $Y_{in} = 2.15\%$, $F = 1$ dm^3/min. Run 35: $T = 318$ K, $Y_{in} = 2.15\%$, $F = 1$ dm^3/min. Run 36: $T = 333$ K, $Y_{in} = 2.15\%$, $F = 1$ dm^3/min. Run 37: $T = 303$ K, $Y_{in} = 1.0\%$, $F = 1$ dm^3/min.

tuted into Eqs. (7) and (8) to predict the breakthrough curve of Run 49 at different flow rates. As shown in Figure 10, the theoretically calculated values are in very good agreement with the experimental data.

Experimental breakthrough data of dichloromethane adsorbed onto KF1500 ACC at different operating temperatures are shown as Fig. 11. The simulation results of Runs 34, 36, and 37 are well matched with the experimental data. The obtained parameters (k_0, E_1, and n) were then applied to Eqs. (6), (7), and (11) to predict the breakthrough curve of Run 35 at different operating temperatures. It is found from Fig. 11 that the theoretical model proposed by this study can precisely predict the breakthrough curve of VOC adsorbed onto a fixed-bed adsorber packed with ACC.

CONCLUSIONS

The ACCs adsorbed by VOCs can be regenerated by using a hot nitrogen purge. The breakthrough time of an ACC adsorber decreases with increasing operating temperature. The adsorption capacity of VOCs onto ACCs increases with increasing weight of packed ACCs. The adsorption capacity of VOCs

onto ACCs decreases with increasing relative humidity of the environment. The breakthrough time of VOCs onto an adsorber packed with ACCs decreases with increasing inlet concentration of gas stream and the flow rate. For acetone, the adsorption capacity per unit weight of three types of ACC adsorbents follows the order FT200-20 (0.493 g/g) > KF1500 (0.377 g/g) > E-ACC (0.285 g/g). This indicates that the higher the BET surface area, the higher will be the adsorption capacity. The theoretical model introduced by Yoon and Nelson can be applied to simulate and to predict the breakthrough curve of VOCs adsorbed onto a packed bed of ACC. The modified model can be used to predict the breakthrough curve of an ACC adsorber operated at various temperatures, inlet concentrations, and flow rates.

NOMENCLATURE

A	constant $[k'' \ln(W_a + \tau)]$
C	gas stream concentration of VOC (mol/dm^3)
E	activated energy (J/mol)
F	flow rate (dm^3/min)
k	proportionally constant
k'	rate constant (min^{-1})
k''	constant
k_0	constant
k_w	work constant
n	order of reaction
P	probability of contaminant breakthrough
Q	probability of contaminant adsorption on the adsorbent
R	gas constant (8.314 J/g·mol·K)
t	operating time (min)
t_b	breakthrough time (min)
t_e	complete breakthrough time (min)
T	temperature of adsorption column (K)
W_a	constant (min)
W_c	weight of adsorbent (g)
W_e	total adsorption capacity (g)
Y_{in}	inlet concentration of gas stream (mol)
Y_{out}	outlet concentration of gas stream (mol)
τ	time require for 50% contaminant breakthrough (min)

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