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Gas phase tricholoethylene removal at low concentration using activated carbon fiber

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Abstract: The breakthrough adsorption behaviors of gas phase trichloroethylene in a packed bed of activated carbon fibers (ACF) were investigated. The specific surface area of the ACF was 600 m^2/g, 1400 m^2/g and 1600 m^2/g, respectively, and the concentration of trichloroethylene ranged from 270 mg/m^3 to 2700 mg/m^3. Results showed that the capacity of adsorption increased with increasing specific surface area, the relationship between the logarithms of 10% breakthrough time and concentration was approximately linear over the experimental range, the breakthrough time decreased with increasing temperature and humidity. The breakthrough curves at different inlet concentration or different temperature can be predicted by several simple theoretical models with good agreements.

Keywords: trichloroethylene; activated carbon fiber; adsorption; breakthrough curve

Introduction

Tricholoethylene (TCE), as an organic solvent, has been widely used in many fields with large quantity, and a large amount of TCE has been found to be emitted from the users, such makes serious pollution, especially for groundwater (Yu, 2000). It is well recognized that conventional drinking water treatment processes have low efficiency in removing tricholoethylene from water; new process must be used to ensure the health of the population. Research has shown that human exposure to volatile organic compounds (VOCs) in water can occur from pathways other than ingestion, inhalation and dermal absorption are also import pathways (Mckone, 1987), and the exposure dose through the three pathways are the same order of magnitude and the major exposure pathway is inhalation (Yoshida, 1993). It is necessary to remove the VOCs both in drinking water and the volatiles from drinking water.

Granular activated carbon (GAC) has been applied for many years to control VOCs due to its high micropore volume and low price. Recently activated carbon fiber (ACF) has been an interesting topic of the removal of VOCs because ACF has uniform micropore structure, faster adsorption kinetics, and a lower pressure drop comparing with GAC. Based on the superior characteristics of ACF, it is considered favorable for the treatment of VOCs. There are several reports (Yu, 2000; Li, 1997; Suzuki, 1991) about ACF absorbing VOCs in drinking water, however, few researches has been done to identify the feasibility of ACF removing gas phase TCE at low concentration. In this work, the effect of the specific surface area, the inlet TCE concentration, the adsorption temperature and the relatively humidity on the breakthrough curves was studied.

A simple model, originally introduced by Yoon and Nelson (Yoon, 1984a) has been used to simulate breakthrough curve.

$$C = C_0 \frac{1}{1 + \exp[k'(\tau - t)]}. \qquad (1)$$

Where C is the exit concentration at breakthrough; C_0 is the inlet concentration; k' is a rate constant; τ is 50% breakthrough time (ratio of exit to inlet gas concentration of 0.5); and t is the operate time. k' can be express as Eq.(2):

$$k' = k/\tau. \qquad (2)$$

Where k is the proportionality constant, it is independent on concentration and flow rate.

If the adsorptive reaction is a nth order reaction, the half life of reactant is inversely proportional to the initial concentration raised to the $(n-1)$th power, then Eq.(3) can be obtained (Yoon, 1984 b):

$$\tau = \frac{k_w}{C^{n-1} F}. \qquad (3)$$

Where k_w is proportionality constant; F is the flow rate. Eq.(1), Eq.(2) and Eq.(3) can be used to simulate and predict the breakthrough curves for various inlet concentrations.

If the adsorption reaction operated under various temperatures, the Arrhenius' equation is introduced to substitute for the constant k_w,

$$k_w = k_0 e^{-E/RT}. \qquad (4)$$

Where k_0 is a constant; E is the activation energy; R is the gas constant, and T is the absolute temperature. Substitution of Eq.(4) to Eq.(3) gives Eq.(5) (Huang, 1999):

$$\tau = \frac{k_0 e^{-E/RT}}{C^{n-1} F}. \qquad (5)$$

Eq.(1), Eq.(2) and Eq.(5) can be used to simulate and predict breakthrough curves under various temperatures.

1 Materials and methods

The experiment system included a small ACF holder, a humidifier, a tricholoethylene generator section, flow meters, and temperature controller. The schematic experimental flow diagram is shown in Fig.1. High-purity nitrogen was used as the carrier gas. The gas phase tricholoethylene was generated using temperature-controlled bubbler, and tricholoethylene used was AR grade. The flow rate through the bubble was adjusted to obtain the desired tricholoethylene concentration. The humid gas was produced by bubbling the high-purity nitrogen through a vessel filled with water and the humid gas was mixed with dry high-purity nitrogen to get designed relative humidity. The ACF holder was a 15 cm long, 0.5 cm inside diameter glass pipe. The total flow rate and the operate temperature was kept at 200 cm^3/min and 25℃. 200 mg ACF was used each time and the ACF bed was 6 cm thick and operated with a residence time of 0.35 s. A GC-9790 gas chromatograph (GC) with ECD

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was used to analyze the TCE concentration of inlet and effluent of the adsorber. The adsorbents used in the study were manufactured by the Ansan Activated Carbon Fiber Factory, China. They are denoted as ACF-600, ACF-1400 and ACF-1600, respectively, referring to their specific surface areas, and their physical characteristics of the ACFs are listed in Table 1.

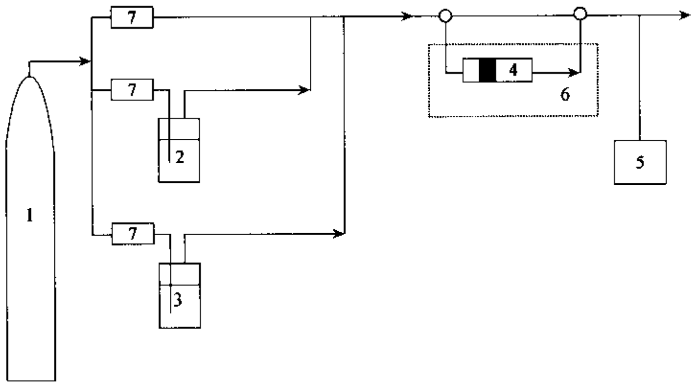

Fig. 1 Schematic diagram of experiment apparatus

1. high-purity nitrogen; 2. tricholoethylene; 3. high-purity water; 4. ACF; 5. gas chromatograph; 6. thermostat; 7. flow meter

Table 1 Physical properties of activated carbon fiber

Characteristics	ACF-600	ACF-1400	ACF-1600
BET surface area, m^2/g	641	1457	1678
Total pore volume, cm^3/g	0.220	0.549	0.759
Micropore volume, cm^3/g	0.172	0.320	0.432
Mescopore volume, cm^3/g	0.007	0.087	0.090

2 Results and discussion

2.1 ACF properties effect

The breakthrough curves of 2700 mg/m^3 TCE in the bed packed with different ACFs are shown in Fig. 2. It is obvious that the breakthrough time increased with the increasing specific surface area, this indicates that the higher the specific area of ACF, the higher the adsorption capacity will be. It should be noted that a beginning of partial breakthrough was observed soon after the beginning and the breakthrough curve then exhibited steps in the case of ACF-600. In the previous work (Kang, 2001) the diffusion time constants (i.e. D/r^2) of MEK and benzene for ACF-600 were much less than the value for ACF-1400, this indicated that ACF-600 was not suitable for using as adsorbent of VOCs due to its lower adsorption kinetics.

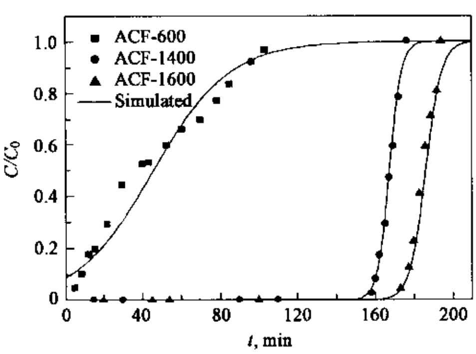

Fig. 2 Effect of specific surface area on breakthrough curve

$C_0 = 2700$ mg/m^3; $RH = 0$; $T = 25$℃

2.2 Concentration effect

The effect of concentration on 10% breakthrough time (ratio of exit to inlet gas concentration of 0.1) is shown in Fig. 3. The inlet concentrations are 2700 mg/m^3, 1900 mg/m^3, 1360 mg/m^3, 540 mg/m^3 and 270 mg/m^3, respectively. The 10% breakthrough time is 150 min, 205 min, 235 min, 408 min and 583 min, accordingly. The breakthrough time decreased with increasing inlet concentration of tricholoethylene at the same flow rate. Fig. 3 shows that the linear relationship provided a reasonable fit to the data. The relation between the 10% breakthrough time and the inlet concentration is:

$$\log(t_b) = -0.576 \times \log(C_0) + 4.176.$$

And the correlation coefficient is $r = 0.99$, where t_b is 10 breakthrough time. There is a same result for GAC adsorbing VOCs (Vanosdell, 1996), their result suggested that the breakthrough time might be cautiously extrapolated to indoor concentration for TCE.

In order to predict the breakthrough curves at different inlet concentration, Eq.(1) and Eq.(2) were used to determine k, k' and τ from the breakthrough curve data both at 2700 mg/m^3 and 270 mg/m^3. Then, the values of τ at 2700 mg/m^3 and 270 mg/m^3 were substituted to Eq.(3) to get the parameter n, then n is substituted to Eq.(3) to get the values of τ for different inlet concentrations. Similarly, the values of k' for different inlet concentrations were obtained using Eq.(2). Finally, k' and τ were substituted to Eq.(1) to predict various inlet concentration breakthrough curves. The simulated and predicted breakthrough curves are shown in Fig. 4. It can be seen that the predicted breakthrough curves fit the experiment data with relative good agreements.

2.3 Temperature effect

The temperature effect on the breakthrough curve of the ACF is shown in Fig.5. The operating temperatures were 298 K, 323 K and 343 K. It is clearly seen that the breakthrough time decreased with increasing operating temperature. The adsorption behavior of VOCs onto ACF could be considered to be physical process, the capacity of an adsorbent by physical adsorption decreases with increasing temperature.

In order to predict the breakthrough curve of tricholoethylene at different temperatures, for example at 323 K, breakthrough curve data of tricholoethylene at 2700 mg/m^3 and 298 K, 270 mg/m^3 and 298 K were substituted to Eq.(3) to get the parameter n, then breakthrough curve data of tricholoethylene at 2700 mg/m^3 and 298 K, 2700 mg/m^3 and 343 K were substituted Eq.(5) to get the parameters E and k_o, the obtained

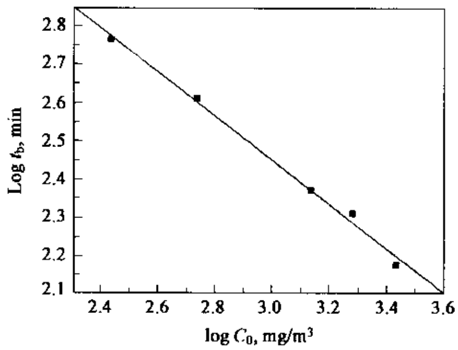

Fig. 3 Effect of inlet concentration on 10% breakthrough time for ACF-1400
RH = 0; T = 25℃

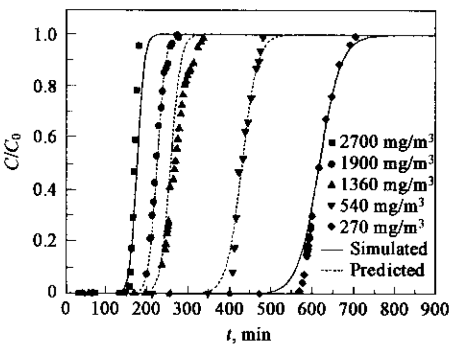

Fig.4 Simulated and predicted breakthrough curves for ACF-1400 at various inlet concentrations
RH = 0; T = 25℃

parameters (n , k_o and E) were applied to Eq.(1), Eq.(2) and Eq.(5) to predict the breakthrough curve at 2700 mg/m³, 323 K. The simulated and predicted breakthrough curves are shown in Fig.5. It is seen that the predicted breakthrough curve fit the experiment data with good agreement.

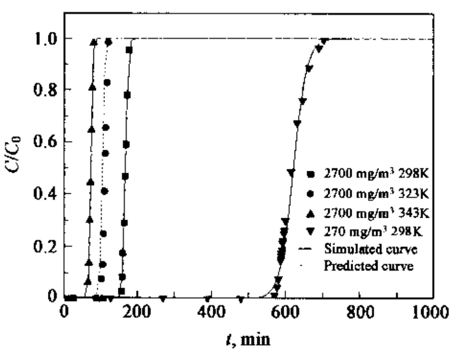

Fig. 5 Effect of temperature on breakthrough curve for ACF-1400

2.4 Relative humidity effect

Adsorption of vapors from humid air is a complex process. In order to investigate the effect of relative humidity on the ACF adsorption process, the relative humidity of the gas stream was designed to be 0, 30% and 80%. Fig.6 shows the breakthrough curves of TCE at different relative humidity, apparently the breakthrough time decreased with increasing relative humidity. Adsorbed water in the pores of ACF would be expected to reduce the volume available to tricholoethylene, thereby reducing the adsorption capacity of the ACF bed. Therefore, the breakthrough time decreased with the increasing humidity. Someone (Cal, 1996) found that water soluble organic(acetone) showed little decrease in its adsorption capacity on ACF up to about 90% RH, while water vapor had an effect on water insoluble organic(benzene) adsorption starting around 65% RH.

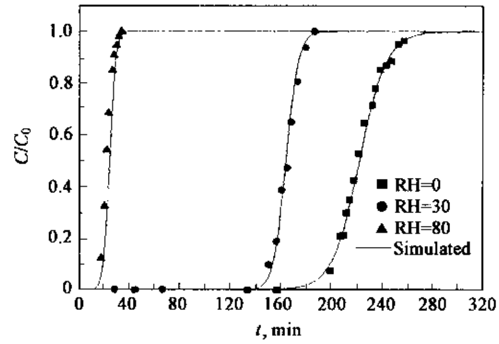

Fig.6 Effect of relative humidity on breakthrough curve for ACF-1400
C_0 = 2700 mg/m³, T = 25℃

3 Conclusions

ACF can be used with packed bed for removal of gas phase TCE at low concentration. The breakthrough characteristics depended on both the ACF's properties and the operation conditions. It was found that the breakthrough time increased with the increasing specific surface area, decreased with increasing operating temperature or humidity. It was important that the breakthrough curves at various inlet concentrations or temperatures can be predicted by several simple models with good agreements. It suggested that we could obtain the breakthrough behaviors under certain condition.

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