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Zheng-Hong Huang^a, Feiyu Kang^a, Jun-Bing Yang^a,
Kai-Ming Liang^a & Aiping Huang^b

^a Department of Materials Science and Engineering,
Tsinghua University, Beijing, 100084, P. R. China

^b Institute of Materials, Zhongshan University,
Guangzhou, 510275, P. R. China

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ADSORPTION OF VOLATILE ORGANIC COMPOUNDS ON ACTIVATED CARBON FIBER PREPARED BY CARBON DIOXIDE

Zheng-Hong Huang, Feiyu Kang, Jun-Bing Yang, and
Kai-Ming Liang

Department of Materials Science and Engineering,
Tsinghua University, Beijing 100084, P. R. China

Aiping Huang

Institute of Materials, Zhongshan University Guangzhou,
510275, P. R. China

Viscose rayon fabric impregnated with $(\text{NH}_4)_2\text{HPO}_4$ were carbonized in $200\text{ cm}^3/\text{min}$ N_2 at $5^\circ\text{C}/\text{min}$ with a residence time of 1 h at 850°C and then activated with $200\text{ cm}^3/\text{min}$ CO_2 at 850°C for different burnoffs. The pore structure of all samples was characterized by N_2 adsorption at 77 K. Adsorption of methy-ethyl-ketone (MEK) and benzene vapor on the adsorbents was measured by gravimetric method. The results show that viscose rayon-based activated carbon fiber (ACF) exhibited narrower microporosity and higher micropore volume as compared with that activated by steam. ACFs prepared in this way have higher adsorption capacity for low concentration VOCs.

Keywords: activated carbon fiber; adsorption; volatile organic compound

INTRODUCTION

Due to the threat they pose to human health, volatile organic compounds (VOCs) are of great concern. Microporous carbons are frequently used for controlling VOCs. Porosity in porous carbon materials is determined by the method of preparation and the precursor material used. Physical activation with steam or carbon dioxide is the conventional method. There are some differences in the molecular size and reactivity of H_2O and CO_2 , resulting in different porosity development in porous carbon. Carbon dioxide produces an opening, followed later of a widening of narrow microporosity, whereas steam widens the microporosity from the early stages of the activation process [1]. Thus, steam activation produces a larger development of

meso- and macroporosity than carbon dioxide activation, and the resulting porous carbon exhibits a wider pore size distribution [2]. The present work prepared viscose rayon based activated carbon fibers (ACF) with carbon dioxide and studied the adsorption characteristics of polar methy-ethyl-ketone (MEK) and non-polar benzene vapors on the ACFs. In addition, adsorption of VOC on ACF with steam activation was performed for comparison.

EXPERIMENTAL

Strips of viscose rayon were first soaked in aqueous solutions containing 5% $(\text{NH}_4)_2\text{HPO}_4$ for 10 minutes and then dried at 110°C. The treated fabrics were carbonized in 200 cm³/min N₂ at 5°C /min with a residence time of 1 h at 850°C and then activated with 200 cm³/min CO₂ at 850°C for different times. The samples obtained are denoted as A-14, A-28 and A-60 according to activation time. The adsorbent used in present study also included the commercially prepared ACF with steam activation, ACF14. The pore structure of all samples was characterized by N₂ adsorption at 77 K using Micromeritics (Norcross, Georgia) ASAP 2010. Gas adsorption of the samples was measured by gravimetric method in a thermo-gravimetric analyzer (TA Instruments, TGA 51) interfaced with a TA Instruments 2000 system. The test details have been described previously [3].

RESULTS AND DISCUSSION

Nitrogen Adsorption and Desorption

The nitrogen adsorption and desorption isotherms at 77 K of ACFs are shown in Figure 1. BET equation was used to calculate the specific surface area. The micropore volume was determined from t-plot analysis. The mesopore volume was obtained by subtraction of the volume adsorbed at $P/P_0=0.1$ from the total pore volume corresponding to the volume adsorbed at $P/P_0=0.95$. The density value of the used liquid N₂ was 0.808 g/cm³. Nitrogen adsorption isotherms indicate that all ACF samples exhibit type I isotherm, which are the typical adsorption characteristics of microporous materials. Table I lists the results obtained by N₂ adsorption. For the samples prepared by CO₂ activation, the BET surface area and the micropore volume increased with increasing burn-off, corresponding to increasing HK median micropore size. In contrast, ACF14 prepared by steam has a smaller micropore volume and a larger micropore size, though it has similar BET surface area in comparison with A-28.

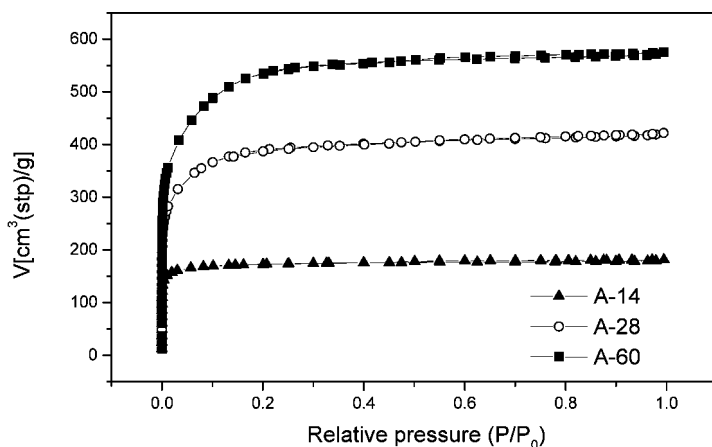


FIGURE 1 Nitrogen adsorption isotherms at 77 K for ACFs.

The surface fractal dimension, $D_{s,N}$, can be calculated from the nitrogen adsorption isotherms according to the following fractal Frenkel-Halsey-Hill (FHH) equation [4,5].

$$\ln V = \text{constant} + \frac{D_{s,N} - 3}{3} [\ln(P_0/P)] \quad (1)$$

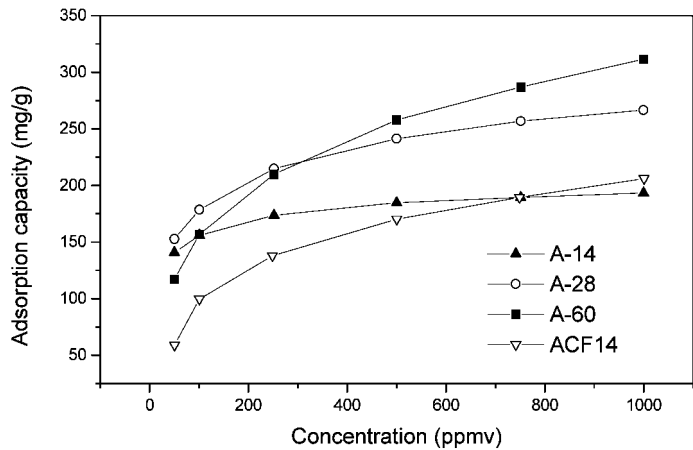
where V is the gas volume, at STP, adsorbed at equilibrium pressure P and P_0 is the saturation pressure of the adsorbate. The equation holds well at the lower end of the isotherm, representing the early stages of the multi-layer buildup, where the van der Waals attraction forces, i.e. gas/solid interaction dominate. The previous fractal analysis [6] showed that the fractal exponent $(D - 3)/3$ provides more realistic results than $(D - 3)$,

TABLE I Nitrogen Adsorption Results of Samples

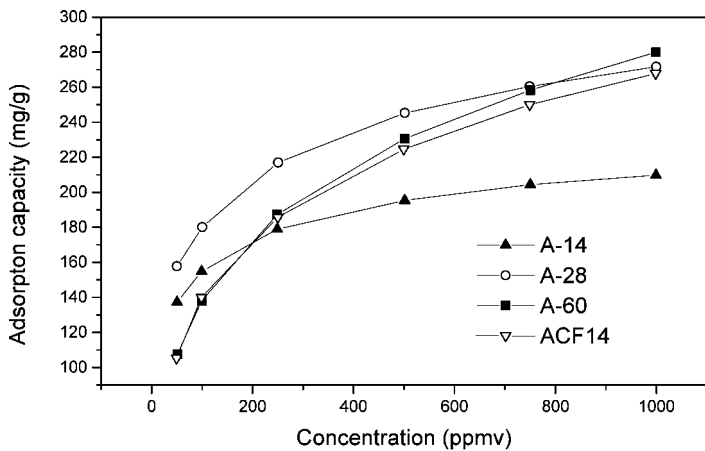
	S_{BET} (m^2g^{-1})	V_{mi} (cm^3g^{-1})	median pore diameter (\AA)	V_{t} (cm^3g^{-1})	V_{me} (cm^3g^{-1})	$D_{s,N}$
A-14	577	0.235	4.9	0.277	0.015	2.80
A-28	1326	0.420	6.1	0.646	0.079	2.32
A-60	1847	0.422	6.6	0.881	0.125	2.03
ACF14	1460	0.320	9.6	0.549	0.087	2.26

S_{BET} : Surface area calculated by BET equation.

V_{mi} : Micropore volume; V_{t} : Total volume; V_{me} : Mesopore volume.



(a)



(b)

FIGURE 2 Adsorption isotherms of ACF samples at 30°C for (a) MEK; (b) benzene.

thus being used in present study. The fractal dimension obtained by nitrogen adsorption can be seen in Table I. The lower the specific surface area is, corresponding to narrower median micropore size, the higher the fractal dimension is, corresponding to rougher surface.

Equilibrium Adsorption of VOCs

Figure 2 shows the adsorption isotherms for VOCs on ACFs. For A-14 and A-28, higher specific area resulted in higher adsorption capacity for VOCs in all the experimental range. However, A-60 had a lower adsorption capacity at lower concentration region and a higher adsorption capacity at higher concentration region as compared with A-14 and A-28. It is noted that ACF14 had a lower adsorption capacity for VOCs as compared with A-28 because of its wider pore size distribution.

In order to compare the limiting adsorption capacity and characteristic adsorption energy, Dubinin-Radushkevich (DR) equation based on Polanyi's potential theory is used to fit the experimental adsorption isotherms, the mass of adsorbate adsorbed per gram of adsorbent, W , can be expressed as [7]

$$W = W_0 \exp[-(A/\beta E_0)^2] \quad (2)$$

where W_0 is the limiting adsorption capacity (mg/g), A is the differential molar work equal to the negative of the differential Gibbs free energy (ΔG), β is the affinity coefficient, E_0 is the characteristic adsorption energy for a standard vapor (usually benzene). A can also be expressed as

$$A = -\Delta G = RT \ln(P_0/P) \quad (3)$$

where T is the absolute temperature, P is the partial pressure of the adsorbate, P_0 is the saturation vapor pressure of the adsorbate. P/P_0 represents the relative pressure. Using the equation above (benzene is adopted as the reference vapor), the parameters W_0 and E_0 are obtained and the results are listed in Table II. The DR equation gave good fits to the experimental data for MEK and benzene adsorption. It shows that the E_0 values obtained from both MEK and benzene adsorption decreased with increasing surface area. The results also show that the E_0 from MEK was much higher than that from benzene for A-14.

TABLE II The Parameters of ACF Samples Obtained by DR Equation

Sample No.	MEK			Benzene		
	W_0 (mg/g)	E_0 (kJ/mol)	C	W_0 (mg/g)	E_0 (kJ/mol)	C
A-14	263.8	33.1	0.996	317.2	27.4	0.999
A-28	457.4	24.9	1.000	459.1	24.1	1.000
A-60	779.9	19.0	0.999	693.7	18.3	1.000
ACF14	663.9	17.1	0.987	643.0	18.7	0.999

C: Correlation coefficient.

CONCLUSIONS

Viscose rayon-based ACF prepared with carbon dioxide activation exhibited narrower microporosity and higher micropore volume as compared with that activated by steam, resulting in higher adsorption capacity for low concentration VOCs. It was the narrow porosity that determined the adsorption capacity at low concentration range.

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