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Adsorption Equilibria of Acetate Compounds on Activated Carbon, Silica Gel, and 13X Zeolite

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The adsorption of volatile organic compounds (VOCs) on activated carbon, silica gel, and type 13X synthetic zeolite was measured by the method of gravimetric adsorption. The selected VOCs were oxygenated hydrocarbons including methyl acetate, ethyl acetate, propyl acetate, butyl acetate, isopropyl acetate, and isobutyl acetate. The equilibrium isotherm data of this study were fitted by the Freundlich and Langmuir equations. The average values of R^2 when using the Freundlich and Langmuir equations to fit the equilibrium isotherms are 0.99 and 0.96, respectively. The results demonstrate that both the Freundlich and Langmuir equations are adequate for fitting the adsorption data of oxygenated hydrocarbons onto the activated carbon, silica gel, and 13X zeolite.

Introduction

Low-molecular-weight organic compounds are either liquids or solids at room temperature, but their vapor pressures are high enough that there will be a substantial amount of the vapor phase. Volatile organic compounds (VOCs) can be defined as those having a vapor pressure greater than 133.3 Pa at room temperature. A substantial number of VOCs can be emitted in processes such as drying, gluing, and coating using solvents, perfume, and fabrics. The VOCs of oxygenated hydrocarbons are emitted considerably in the printing, painting, and solvent industries. The performance of porous sorbents is determined by their intrinsic characteristics and porous structure. The amount of VOCs that can be adsorbed on the external surface of the sorbent is small. Thus, a porous sorbent with a large internal surface area is generally selected to deal with the VOCs. To the best of our knowledge, few studies have been reported on the adsorption of oxygenated hydrocarbons onto porous sorbents. Therefore, methyl acetate, ethyl acetate, propyl acetate, butyl acetate, isopropyl acetate, and isobutyl acetate were selected as sorbates to be adorbed onto activated carbon, silica gel, and 13X zeolite in this study.

The adsorption process provides a method for removing VOCs from the air, but discussions about the adsorption of oxygenated hydrocarbons are rare in the open literature. The adsorption process is one of the nondestructive processes that can recycle adsorbed VOCs. Activated carbon, silica gel, and zeolite (molecular sieves) are widespread adsorbents used to achieve VOC reduction in industry. In general, activated carbon has a greater affinity for volatile organic compounds because of the nonpolar nature of its surface compared to other solid sorbents. Silica gel is usually prepared from a sol−gel process.[1] Because of the polar surface of silica gel, it adsorbs molecules from the gas phase and keeps them on the surface. A systematic evaluation of the adsorption capacities of organic vapors on silica gel was started in the late 1980s. The first family of synthetic zeolites was known as type A. Later, a type X zeolite was developed with a larger pore size than that of type A.[2]

A number of mathematical models have been developed to describe situations in which the sorbate occurs either locally on specific sites or is mobile over the surface of the sorbent. The Freundlich and Langmuir equations were the oldest and most frequently used isotherm models.[4] The semiempirical Freundlich equation has been applied in the low-to-intermediate concentration range.[3] Adsorption capacity can also be expressed by the Langmuir isotherm equation on the uniform adsorption surface and by the Fruendlich isotherm equation on the nonuniform adsorption surface.[5] With the goal of achieving a better understanding of the adsorption behavior of oxygenated hydrocarbons, activated carbon, silica gel, and 13X zeolite were chosen, and the regression results were analyzed by the Langmuir and Freundlich equations in this study. The operating pressures (from 1 to 62 mmHg) controlled in this study can be regarded as the range of low-to-intermediate pressure. Generally speaking, the Freundlich equation was usually well fit in this pressure range. However, both the Langmuir and Freundlich equations were used to fit these equilibrium adsorption data in the acceptable deviation.

Experimental System

Sorbents. The commercial sorbents were activated carbon (China Activated Carbon Industries Co.), silica gel (Kanto Chemical Co.), and 13X zeolite (Lancaster Synthesis Co.). Their surface properties were measured with a BET sorptometer (Micromeritics ASAP 2000) and are shown in Table 1. The sorbent was degassed at 473.15 ± 1 K under vacuum before the adsorption measurements were carried out at 77 ± 1 K. The specific surface area, pore volume, and average pore diameter were calculated by the BET method.[3]

Chemicals. Methyl acetate (99%, Lancaster), ethyl acetate (99.5%, Pharmco), propyl acetate (98%, Acros),

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Table 1. Surface Properties of the Sorbents

sorbent	a/m^2·g^{-1} [a]	v/cm^3·g^{-1} [b]	d/nm [c]
activated carbon	990	0.094	2.7
silica gel	535	0.653	3.6
13X zeolite	440	0.117	9.9

[a] BET surface area. [b] Average pore volume. [c] Average pore diameter.

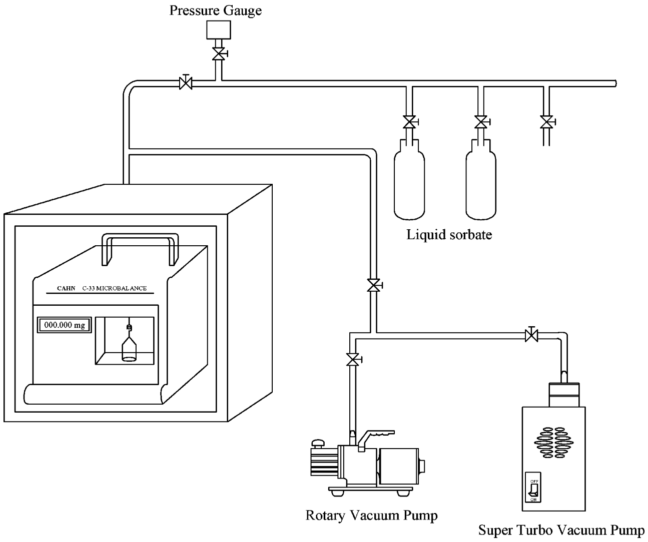

Figure 1. Gravimetric adsorption apparatus used in this study (static gravimetric method).

butyl acetate (99.46%, TEDIA), isopropyl acetate (99%, Lancaster) and isobutyl acetate (98%, Acros) were used.

Apparatus and Procedure.[6] Figure 1 shows the equipment used in this study. The mass change during adsorption was measured by an electronic microbalance (Cahn C-33). The uncertainty of the microbalance is ±0.1 μg. The pressure was controlled from 1.333×10^2 Pa to 8.265×10^3 Pa and measured with a pressure gauge (Cole Parmer U-68700; uncertainty ±0.1333 Pa).

The temperature of this experimental system was maintained at 298.15 K, and the adsorption isotherms were depicted from the adsorption capacity (mg/g adsorbent) and relative pressure, P/P^0, where P is the operating pressure (mmHg) and P^0 is the vapor pressure of the liquid solvent (mmHg). The step-by-step operating procedure for measuring the adsorption capacity (uptake) of VOCs on the sorbent using the static adsorption system is as follows.

After regeneration in a vacuum dryer (373.15 K, 24 h), 50 mg of the granular sorbent was placed on a pan of the electric microbalance (Cahn C-33). The liquid sorbate was placed into the glass bottles shown in Figure 1. We repeated the degassing procedure at least three times (thawing and then freezing). The vapor of the liquid sorbate went into the adsorption system. After equilibrium, we recorded the total mass of the sorbent and sorbate and the pressure of the adsorption system. The vapor of the liquid sorbate was allowed into the adsorption system again to change the pressure of the VOCs. After equilibrium, we again recorded the total mass of the sorbent and sorbate and the pressure of the adsorption system. We repeated the above procedure until the saturated pressure was attained.

Results and Discussion

Because the method of static gravimetric adsorption is well known and the adsorption capacity of aromatic compounds onto activated carbon, silica gel, and 13X zeolite was tested and measured by the same apparatus in the authors' previous study,[6] the adsorption apparatus used in this study is considered to be adequate.

The selected VOCs in this study were methyl acetate, ethyl acetate, propyl acetate, butyl acetate, isopropyl acetate, and isobutyl acetate. The adsorption capacity of these VOCs on the activated carbon, silica gel, and 13X zeolite are listed in Table 2. The adsorption experiments were measured at 298.15 K. The adsorption capacity of the VOCs on activated carbon versus the relative pressure, P/P^0, was recorded as the adsorption isotherm and shown in Figure 2a. P^0 represents the vapor pressure of the liquid solvent at 298.15 K. The nonpolar surface of activated carbon was induced to produce polarity by polar oxygenated hydrocarbons, and the polar-induced force was formed between activated carbon and the oxygenated hydrocarbons. Therefore, the oxygenated hydrocarbons were adsorbed onto the surface of activated carbon. In general, the numbers of electrons in a molecule increases with its molecular weight, and the polarity-induced force is proportional to its molecular weight. As shown in Figure 2a, the adsorption capacity on activated carbon is increased as the molecular weights of the oxygenated hydrocarbons increase, except for ethyl acetates. The structure of ethyl acetate is more symmetric than others. Its polarity is expected to be weaker, which results in a stronger attractive force between activated carbon and ethyl acetate. For the isomers of propyl acetate and butyl acetate, the branching structures of isopropyl acetate and isobutyl acetate cause a geometric barrier effect and decrease the contact area for isomers of the acetate esters adsorbed by activated carbon. Therefore, the adsorption capacity of isopropyl acetate and isobutyl acetate was less than that of propyl acetate and butyl acetate in this study.

It is generally accepted that carbonyl-containing compounds such as esters are adsorbed on silica gel through the formation of hydrogen bonds involving the lone-pair electrons of the carbonyl oxygen per ester and hydrogen atom of hydroxyl groups on silica. The adsorption of oxygenated hydrocarbons on silica gel belongs to the polarity−polarity phenomenon. Because the interaction of polarity−polarity and hydrogen bonds exist in the system of esters adsorbed by silica gel, the adsorption behavior is more complex than that of other sorbents. The effect of the geometric barrier might be the result of repulsion between polar materials, especially for the longer c-chain. As mentioned earlier, there are many parameters leading the isotherm curves to be crossed in the relative pressure from 0.3 to 0.5 (as shown in Figure 2b). From Figure 2c, the adsorption capacity was increased by the increased polarity and the decreased effect of the geometric barrier. Therefore, the order of the adsorption capacity was found to be isobutyl acetate, ethyl acetate, butyl acetate, propyl acetate, isopropyl acetate, and methyl acetate. However, the effect of the geometric barrier is increased by the increased relative pressure. Therefore, the adsorption capacity of larger molecules was increased slowly by the increased relative pressure, and the order of the adsorption capacity changs to ethyl acetate, isobutyl acetate, methyl acetate, isopropyl acetate, and n-butyl acetate at higher relative pressure in Figure 2d. Figure 2e depicts the adsorption isotherms of oxygenated hydrocarbons on the 13X zeolite.

Comparing part e of Figure 2 with parts a and b of Figure 2, we find that the adsorption capacity of oxygenated hydrocarbon onto 13X zeolite is much lower than those of activated carbon and silica gel. The affinities between

Table 2. Experimental Data from This Work

sorbent: activated carbon

methyl acetate		ethyl acetate		*n*-propyl acetate		*n*-butyl acetate		isopropyl acetate		isobutyl acetate	
P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}
0.006	45.697	0.027	244.197	0.0210	160.320	0.1065	191.200	0.022	105.062	0.0556	127.887
0.069	124.555	0.178	375.488	0.0842	185.615	0.2308	253.433	0.111	126.862	0.1111	135.664
0.156	148.287	0.370	408.268	0.2105	193.723	0.3846	260.065	0.289	136.708	0.2222	141.603
0.243	156.548	0.534	416.210	0.3158	197.574	0.6154	260.796	0.400	140.589	0.3333	144.405
0.358	161.370	0.671	419.856	0.3790	199.460			0.533	143.531	0.3889	146.753
0.497	165.537	0.740	420.918	0.5053	199.644			0.600	144.793	0.5555	149.415
								0.667	145.766	0.6667	150.914
								0.711	146.496	0.7222	151.705
								0.756	147.017		
								0.800	147.272		
								0.822	147.446		

sorbent: silica gel

methyl acetate		ethyl acetate		*n*-propyl acetate		*n*-butyl acetate		isopropyl acetate		isobutyl acetate	
P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}
0.012	9.513	0.041	43.607	0.021	37.402	0.077	65.564	0.089	48.759	0.056	63.641
0.081	27.635	0.137	69.214	0.105	56.760	0.231	74.621	0.200	63.275	0.111	74.465
0.156	43.900	0.219	88.664	0.147	65.028	0.308	79.609	0.280	74.719	0.167	86.606
0.243	58.134	0.329	105.663	0.232	75.942	0.385	82.190	0.378	81.943	0.333	98.009
0.318	69.695	0.466	121.875	0.253	78.633	0.462	84.181	0.489	88.174	0.444	103.017
0.393	80.058	0.616	129.158			0.538	84.575	0.556	90.184	0.500	105.407
0.457	87.175	0.685	130.160			0.615	84.881	0.622	90.979	0.556	106.320
0.503	91.862	0.753	130.447							0.611	106.865
0.549	95.456										

sorbent: 13X zeolite

methyl acetate		ethyl acetate		*n*-propyl acetate		*n*-butyl acetate		isopropyl acetate		isobutyl acetate	
P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}	P/P^0	q/mg·g^{-1}
0.012	6.675	0.055	9.839	0.021	9.638	0.077	5.623	0.067	7.643	0.056	9.729
0.040	9.445	0.151	12.194	0.084	11.153	0.231	7.049	0.178	8.957	0.167	11.318
0.116	12.594	0.219	12.909	0.147	12.121	0.308	7.511	0.244	9.492	0.278	12.840
0.179	13.937	0.288	13.456	0.211	12.710	0.385	8.014	0.311	9.736	0.389	13.535
0.272	14.777	0.370	13.792	0.274	12.879	0.462	8.434	0.400	10.174	0.500	13.932
0.387	14.861	0.438	13.834			0.538	8.602	0.533	10.417	0.556	14.098

Table 3. Partial Molar Volumes for Some Oxygenated Hydrocarbons[9]

compound	$\overline{V}_S$/m^3 mol^{-1}
methyl acetate	7.17×10^{-5}
ethyl acetate	8.81×10^{-5}
n-propyl acetate	1.05×10^{-4}
	9.80×10^{-5}

13X zeolite and esters are lower than those of activated carbon and silica gel, and the surface area of 13X zeolite is smaller. Except for two isomers, the molecular volumes of esters (Table 3) are increased by their molecular weight. Methyl acetate, ethyl acetate, and *n*-propyl acetate filled in the channel structure of 13X zeolite more easily than did *n*-butyl acetate.

The adsorption behaviors of oxygenated hydrocarbons adsorbed onto the activated carbon, silica gel, and 13X zeolite are shown in Figure 3a–f. The phenomena of oxygenated hydrocarbons adsorbed onto activated carbon could be attributed to the intermolecular force, surface area, geometric contour, and so forth. Because of the intrinsic difference in the adsorption apparatus, it is hard to demonstrate which parameter is the more important factor to influence the adsorption capacity in theory. Therefore, Figure 3a–f and Table 1 demonstrate that the surface area of activated carbon is large enough and that the adsorption capacity of activated carbon is larger than that of silica gel.

Adsorption Isotherm. Both the Frendlich and Langmuir equations were applied to fit the data of adsorption of oxygenated hydrocarbons onto activated carbon, silica gel, and 13X zeolite. The results of the fitted data were also analyzed in this study.

A. Freundlich Isotherm.[8] The gas pressure was controlled from low to intermediate pressure in this study. The Freundlich isotherm equation can be applied to fit these equilibrium data and is expressed as

$$q = Kp^n$$

where q is the adsorption capacity (mg sorbate/g sorbent), K is the Freudlich constant, p is the gas pressure of the oxygenated hydrocarbons (mmHg), and n is an exponent. The regression constants and exponents for all of the oxygenated hydrocarbons adsorbed by different sorbents at 298.15 K are shown in Table 4. The values of R^2 for the predicted adsorption isotherms in the activated carbon bed are from 0.885 to 0.997, from 0.991 to 0.999 in the silica gel bed, and from 0.986 to 0.998 in the 13X zeolite bed.

B. Langmuir Isotherm.[3] The Langmuir isotherm was developed to describe the adsorption equilibrium for the gas phase. Under the assumption that the adsorption rate and desorption rate are equal, the Langmuir equation can be expressed as

$$q = A\frac{BC}{1 + BC}$$

where A and B are adsorption constants and C is the equilibrium concentration (mg/L). The regression constants for oxygenated hydrocarbons adsorbed by different sorbents

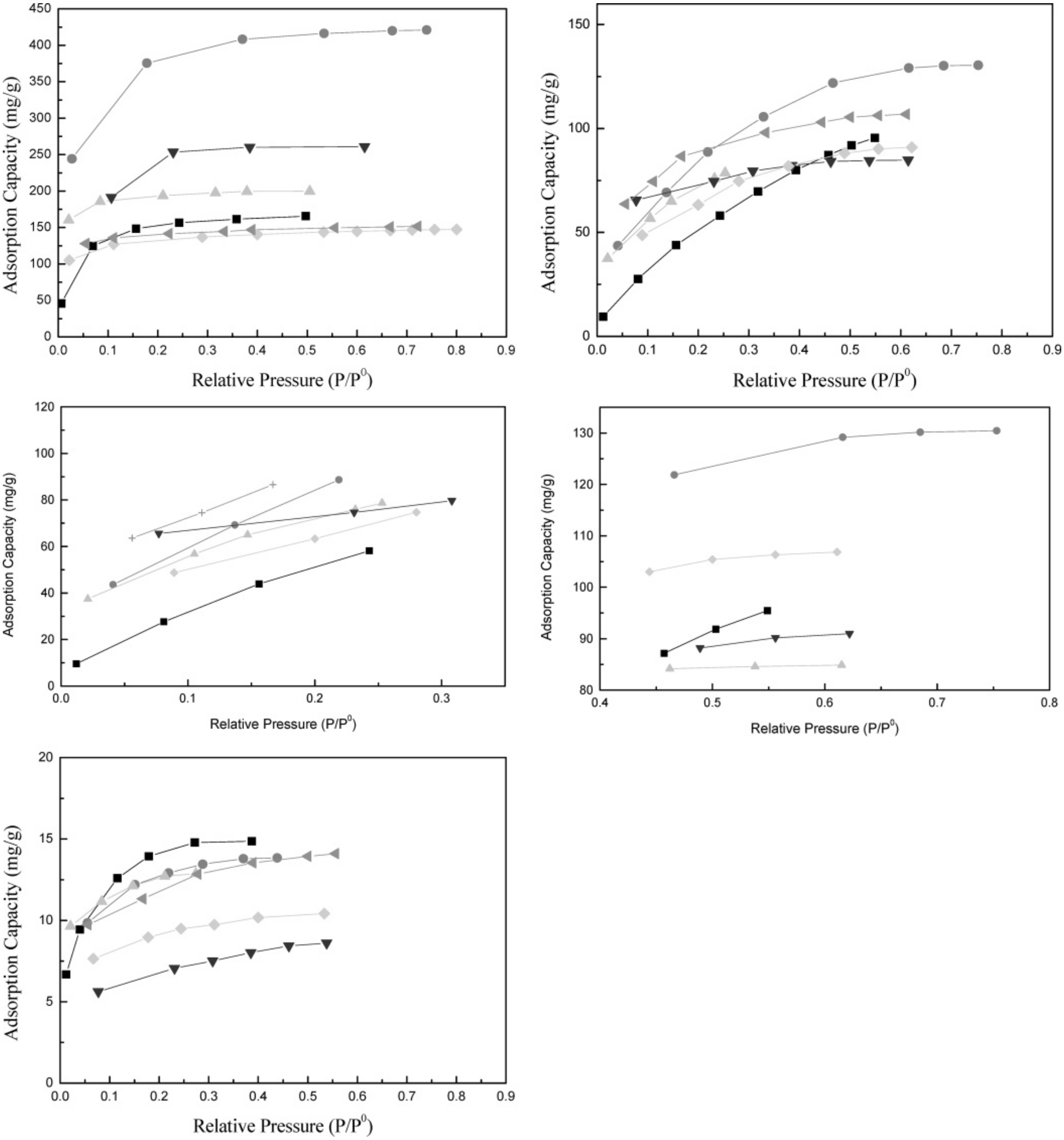

Figure 2. (a) Adsorption capacities for acetate esters adsorbed by activated carbon at different pressures: ■, methyl acetate; ●, ethyl acetate; ▲, propyl acetate; ▼, butyl acetate; ♦, isopropyl acetate; left-facing triangle, isobutyl acetate. (b) Adsorption capacities for acetate esters adsorbed by silica gel at different pressures: ■, methyl acetate; ●, ethyl acetate; ▲, propyl acetate; ▼, butyl acetate; ♦, isopropyl acetate; left-facing triangle, isobutyl acetate. (c) Adsorption capacities for acetate esters adsorbed by silica gel at 0−0.3 relative pressures: ■, methyl acetate; ●, ethyl acetate; ▲, propyl acetate; ▼, butyl acetate; ♦, isopropyl acetate; +, isobutyl acetate. (d) Adsorption capacities for acetate esters adsorbed by silica gel at 0.4−0.8 relative pressures: ■, methyl acetate; ●, ethyl acetate; ▲, butyl acetate; ♦, isobutyl acetate; ▼, isopropyl acetate. (e) Adsorption capacities for acetate esters adsorbed by 13X molecular sieves at different pressures: ■, methyl acetate; ●, ethyl acetate; ▲, propyl acetate; ▼, butyl acetate; ♦, isopropyl acetate; left-facing triangle, isobutyl acetate.

at 298.15 K are shown in Table 5. The values of R^2 for the predicted adsorption isotherms in the activated carbon bed are from 0.966 to 0.999, from 0.970 to 0.989 in the silica gel bed, and from 0.959 to 0.997 in the 13X zeolite bed.

The average values of R^2 obtained by regression of the Freundlich and Langmuir equations are 0.985 and 0.959, respectively. Beyond 94% data points, the values of R^2 are larger than 0.970 for the Freundlich equation; however, there are only 83% data points larger than 0.970 for the Langmuir equation. The characteristic curves are different for these two equations. If the adsorption isotherm presents the parabolic curve, then results regressed by the Fruendlich equation would be excellent. On the contrary, if the adsorption capacity levels off in the higher relative pressure, then the adsorption data are suitable for regression by the Langmuir equation. Most of the adsorption capaci-

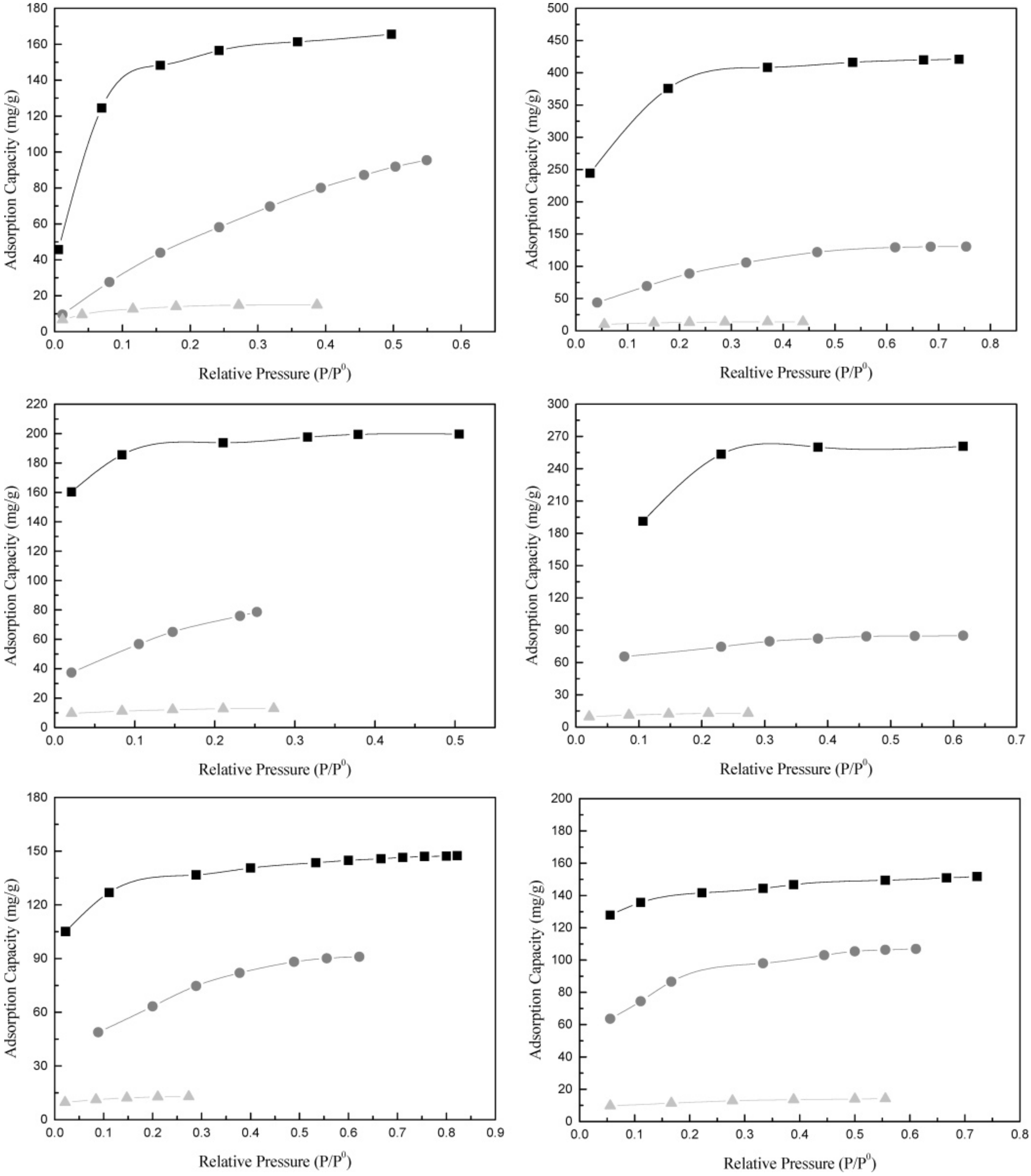

Figure 3. (a) Adsorption capacities for methyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieves. (b) Adsorption capacities for ethyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieves. (c) Adsorption capacities for propyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieves. (d) Adsorption capacities for butyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieve. (e) Adsorption capacities for isopropyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieves. (f) Adsorption capacities for isobutyl acetate adsorbed by different adsorbents against relative pressures: ■ activated carbon; ●, silica gel; ▲, 13X molecular sieves.

ties of this study do not completely level off at the higher relative pressure. The experimental results demonstrate that the regressions of adsorption capacities of this study are better described by the Fruendlich equation than the Langmuir equation, and the Freundlich isotherm has been shown to be adequate for the adsorption of oxygenated hydrocarbons onto the activated carbon, silica gel, and 13X zeolite.

Table 4. Regression Values of *K* and *n* in the Freundlich Equation

sorbent	sorbate	K	n	R^2
activated carbon	methyl acetate	50.48	0.30	0.971
	ethyl acetate	226.74	0.17	0.973
	n-propyl acetate	163.57	0.07	0.974
	n-butyl acetate	190.88	0.18	0.885
	isopropyl acetate	106.93	0.09	0.994
	isobutyl acetate	128.83	0.06	0.997
silica gel	methyl acetate	5.99	0.61	0.999
	ethyl acetate	28.90	0.39	0.994
	n-propyl acetate	36.78	0.30	0.996
	n-butyl acetate	65.59	0.13	0.991
	isopropyl acetate	30.90	0.33	0.995
	isobutyl acetate	64.94	0.22	0.992
13X zeolite	methyl acetate	5.86	0.24	0.989
	ethyl acetate	7.95	0.17	0.986
	n-propyl acetate	9.60	0.12	0.997
	n-butyl acetate	5.59	0.22	0.998
	isopropyl acetate	6.51	0.15	0.997
	isobutyl acetate	9.66	0.17	0.995

Table 5. Regression Values of *A* and *B* in the Langmuir Equation

sorbent	sorbate	A	B	standard deviation
activated carbon	methyl acetate	0.00631	0.0158	0.999
	ethyl acetate	0.00233	0.00355	0.999
	n-propyl acetate	0.00499	0.00126	0.994
	n-butyl acetate	0.00336	0.00247	0.966
	isopropyl acetate	0.00686	0.00275	0.971
	isobutyl acetate	0.00661	0.00129	0.971
silica gel	methyl acetate	0.01207	0.18906	0.989
	ethyl acetate	0.00735	0.04875	0.980
	n-propyl acetate	0.01282	0.0142	0.974
	n-butyl acetate	0.01144	0.00396	0.970
	isopropyl acetate	0.00962	0.04527	0.987
	isobutyl acetate	0.0089	0.00727	0.984
13X zeolite	methyl acetate	0.06891	0.1686	0.978
	ethyl acetate	0.06838	0.13467	0.997
	n-propyl acetate	0.07811	0.02656	0.960
	n-butyl acetate	0.11156	0.06837	0.976
	isopropyl acetate	0.09371	0.11504	0.984
	isobutyl acetate	0.0701	0.03446	0.959

Conclusions

The static gravimetric apparatus, which was built for our previous study,[6] was applied to measure the adsorption equilibrium data of the adsorption of oxygenated hydrocarbons onto the activated carbon, silica gel, and 13X zeolite. As noted, the effects of the specific surface area of the sorbent and the molecular weight and structure of the sorbate on the adsorption capacity of each system were significant. The results showed that the great adsorption performance for the same oxygenated hydrocarbons adsorbed by different sorbents was found in the order of activated carbon, silica gel, and 13X zeolite. Nonpolar absorbents were induced by polar molecules to get the polarity-induced force to attract each other. Because the characteristic curve of the adsorption capacities present a parabolic curve in this study, the values of R^2 for the Fruendlich equation were greater than those of the Langmuir equation, and the average value of R^2 is 0.985 for the Fruendlich isotherm.

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