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Adsorption of *n*-Butane and *n*-Heptane on 5A Zeolite

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ABSTRACT

Adsorption thermodynamics of *n*-butane and *n*-heptane on 5A zeolite was studied by the flow gravimetric-chromatograph technique at temperatures ranging from 533 K to 653 K and over a range of hydrocarbon partial pressures. It is shown that the equilibrium amount of *n*-heptane adsorbed is much larger than that of *n*-butane. Adsorption isotherms are modeled satisfactorily by using a modified virial equation and the Dubinin–Astakhov (D–A) equation. It is concluded that the influence of *n*-butane is very small while the influence of *n*-heptane is significant in the separation stage of the total isomerization process.

Modified virial equation: $P = a^n \exp(C_1 + C_2 a + C_3 a^2)$

D–A equation: $a = a_m \exp\{B[\ln[(T/T_c)^2 P_c/P]]^n\}$

The adsorption isosteric heat and entropy were calculated, and the results show that the adsorption isosteric heat of *n*-heptane is larger than that of butane at low loadings.

INTRODUCTION

The total isomerization process (TIP) developed by Union Carbide is an efficient technique to increase the iso-fraction of hydrocarbons, thus improving the quality of gasoline. In this method, 5A zeolite is used to separate *n*/*i*-

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paraffins (1). The molecular critical diameters of *n*-paraffins are around 4.9A but *i*-paraffins are all larger than 5A. Therefore, 5A zeolite can adsorb linear *n*-C_5 and *n*-C_6 hydrocarbons but exclude their branched counterparts. The appropriate development of sorption and desorption cycles plays the crucial role in the successful application of zeolites as industrial sorbents (2). In the past two decade, much interest has arisen in the adsorption thermodynamics and kinetics of 5A zeolite.

There are a limited number of studies of the adsorption properties of 5A zeolite. Peterson and Redlich investigated the sorption of normal paraffins on Linde's 5A molecular sieve pellets at less than 573 K (3). 4–8 mesh 5A molecular sieve beads have been used to study the thermodynamics of sorption of long-chain normal paraffins by the McBain spring balance method, and it was observed that the isosteric heat of adsorption did not rise at a constant rate with increasing carbon number (4). The behavior of *n*-heptane and *n*-pentane in 5A zeolite has been studied separately by Doetsch et al. and Silva et al. and these authors have been able to explain the equilibrium isotherms and adsorption kinetics of these hydrocarbons (5, 6). The dual-site Langmuir model (DLS) has been used to correlate nitrogen and oxygen adsorption on 5A zeolite (7), but adsorbent heterogeneity plays a very significant and complex role in the adsorption of pure gas and multicomponent gas mixtures (8). Although several adsorption theories and models have been developed, the conditions of their application are usually very limited and conflicting conclusions have been drawn about the adsorption of medium-chain hydrocarbons in 5A zeolite (5).

This work deals with the very specific conditions used in TIP and gives a simple explanation about the adsorption of *n*-butane and *n*-heptane on 5A zeolite.

EXPERIMENTAL

The adsorbent is 20–40 mesh 5A zeolite binderless particles provided by Jinling Petrochemical Co. of the People's Republic of China. Its physical properties as measured by an ASAP2400 Nitrogen Auto-Adsorber (Micrometrics, USA) are shown in Table 1.

The experimental systems are shown in Fig. 1 for *n*-butane and in Fig. 2 for *n*-heptane.

TABLE 1

Physical Properties of 5A Zeolite

Surface area	405.1 m^2/g
Pore volume	0.1248 cm^3/g
Apparent density	1.423 g/cm^3
Solid density	2.841 g/cm^3

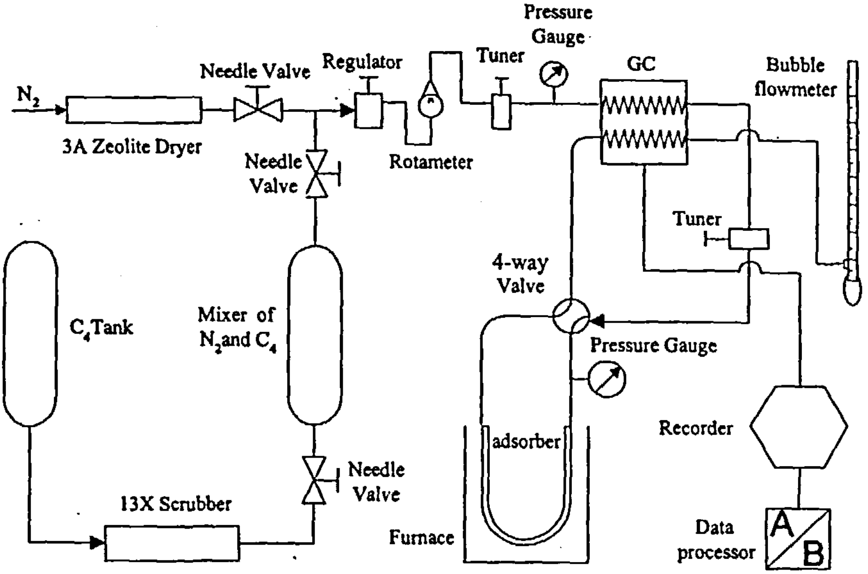

FIG. 1 Schematic drawing of an experimental apparatus for adsorption of *n*-butane on 5A zeolite.

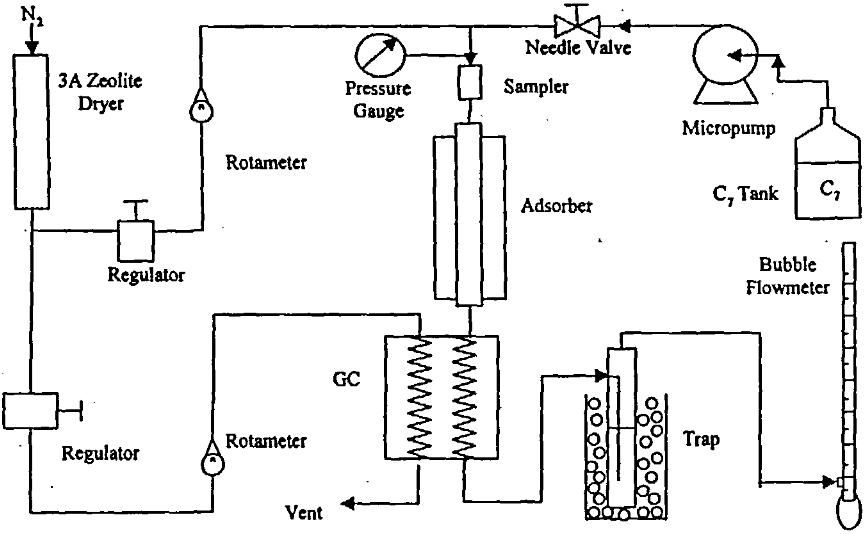

FIG. 2 Schematic drawing of an experimental apparatus for adsorption of *n*-heptane on 5A zeolite.

For the *n*-butane system, the gases of *n*-butane (99.0%) and nitrogen (99.999%) were first mixed according to the desired ratio of *n*-butane to nitrogen. The resulting gas mixture was further analyzed with a gas chromatograph (GC-103) in order to determine the actual gas ratio. The mixture flow rate was controlled with a needle valve and a rotameter. The gas mixture was then introduced into the reference column of a gas chromatograph (GC), and further directed into the analytical column of the GC after passing through a 4-way valve. After the GC was stabilized for about 20 minutes, the 4-way valve was switched and the mixture was introduced into a U-tube filled with 5A zeolite, and then directed into the analytical column of the GC. The gas mixture was shut off when the recorder showed a stabilized line, which indicated that the 5A zeolite was saturated. The 5A zeolite was activated at 400°C in a nitrogen atmosphere for 1 hour before being used as an adsorbent. The U-tube was sealed by two rubber septa. The U-tube with 5A zeolite was weighed before and after the adsorption by using an electronic balance.

For the *n*-heptane system, after the gas chromatograph reached the stable state, liquid *n*-heptane (analytical grade) was pumped into the adsorber by a SY-02A micropump, and then collected by a trap held in liquid nitrogen to minimize evaporation. Once the trace of the recorder reached a maximum, indicating adsorption had reached saturation, the micropump was stopped. The amount of adsorbed *n*-heptane was determined by mass balance.

The entire system was operated at ambient atmosphere. At each temperature the ratios of *n*-butane to nitrogen and *n*-heptane to nitrogen were altered, and thus the partial pressure of the adsorbate was also changed.

RESULTS AND DISCUSSION

Equilibrium Isotherms

The equilibrium amount of *n*-butane and *n*-heptane on 5A zeolite has been measured in the 533–653 K temperature range, and the results are shown in Figs. 3 and 4. It is clear that the amount of hydrocarbon adsorbed increases more rapidly at low partial pressures compared to higher pressures, and the amount of *n*-heptane is much larger than that of *n*-butane at a given partial pressure. Comparing the present results with those of the adsorption of *n*-pentane and *n*-hexane on 5A zeolite (9), we found that the adsorption ability of hydrocarbons on 5A zeolite at a given temperature and pressure is in following order:

n-heptane > *n*-hexane > *n*-pentane > *n*-butane

In TIP, about 20% of *n*-butane and *n*-heptane is present in the products mixture. In the separation process, the existence of *n*-pentane, *n*-hexane, and *n*-heptane would suppress the adsorption of *n*-butane and thus reduce the

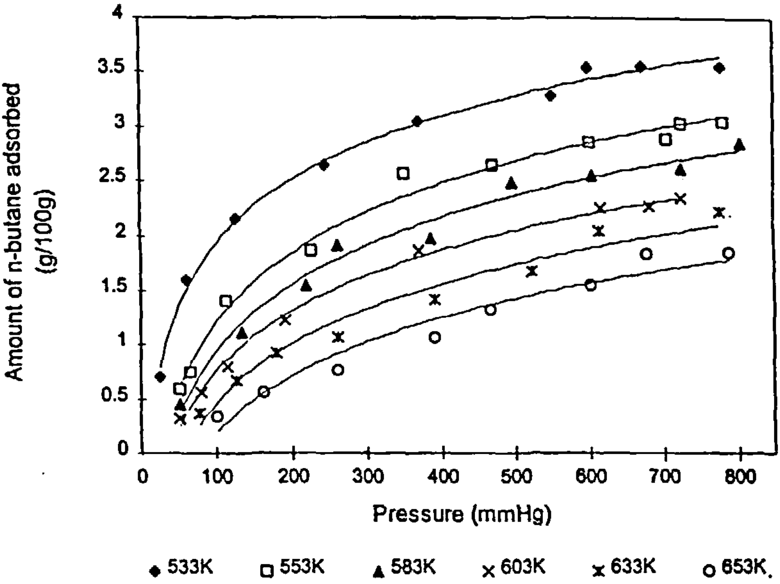

FIG. 3 Adsorption equilibrium isotherms of *n*-butane on 5A zeolite.

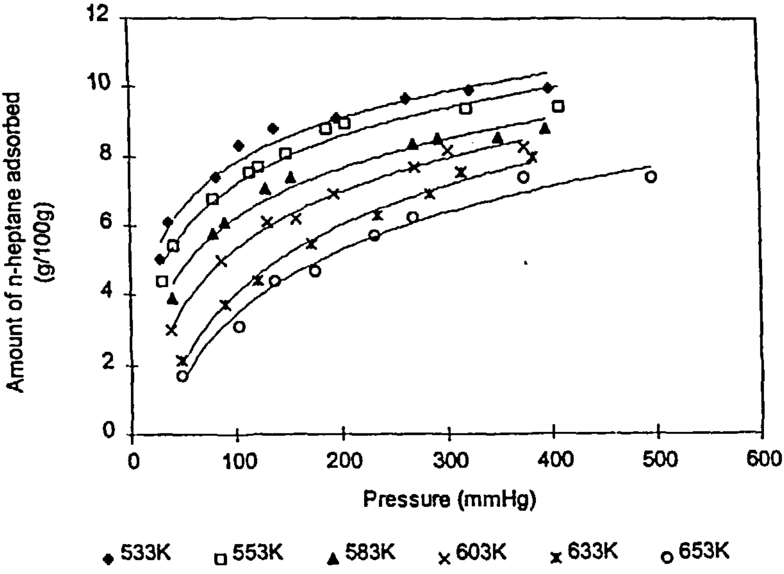

FIG. 4 Adsorption equilibrium isotherms of *n*-heptane on 5A zeolite.

amount of n-butane adsorbed to an even lower level. Therefore, in TIP the influence of the adsorption of n-butane on the adsorption of n-pentane and n-hexane is quite small, even negligible. However, the influence of n-heptane is significant because the adsorption of n-pentane or n-hexane would be suppressed by the n-heptane, and therefore the amount of n-heptane should then be minimized as much as possible.

Modeling of Equilibrium Isotherms

Significant effort has been made to understand the adsorption phenomena, and there are constantly new ways to interpret adsorption data and to perform adsorption experiments. Some publications provide excellent reviews of these theories (8). Among these theories, nine commonly used isotherms were chosen to model the experimental data as follows.

1. Langmuir equation

$$a = \frac{a_{\mathrm{m}} bP}{1 + bP}$$

where a = amount adsorbed of adsorbate, g/100 g
a_{m} = maximum amount adsorbed of adsorbate, g/100 g
P = partial pressure of adsorbate, mmHg
b = constant

2. Freundlich equation

$$a = K_{\mathrm{f}} P^{1/n}$$

where K_{f} = Freundlich constant, dependent on type of adsorbent and its density
n = constant

3. Langmuir–Freundlich equation

$$a = \frac{a_{\mathrm{m}} bP}{1 + bP^n}$$

4. Freundlich–Langmuir equation

$$a = \frac{a_{\mathrm{m}} bP^n}{1 + bP}$$

5. Dubinin–Astakhov (D–A) equation

$$a = a_{\mathrm{m}} \exp\{B[\ln[(T/T_{\mathrm{c}})^2 P_{\mathrm{c}}/P]]^n\}$$

where B = D–A parameter
T = absolute temperature, K
T_c = critical absolute temperature, K
P_c = critical pressure of adsorbate, mmHg

6. 2-Langmuir equation

$$a = \frac{a_{m1}b_1P}{1 + b_1P} + \frac{a_{m2}b_2P}{1 + b_2P}$$

where $a_m = a_{m1} + a_{m2}$
a_{m1} = maximum amount adsorbed of adsorbate on one kind of active site in 2-Langmuir equation, g/100 g
a_{m2} = maximum amount adsorbed of adsorbate on another kind of active site in 2-Langmuir equation, g/100 g
b_1, b_2 = constants

7. Virial equation

$$P = a \exp(C_1 + C_2a + C_3a^2 + \cdots)$$

where C_1, C_2, C_3 = constants

8. Modified virial equation

$$P = a^n \exp(C_1 + C_2a + C_3a^2)$$

9. Temkin equation

$$a = k_t \ln bP$$

where k_t = constant

Generally, 10% of average relative error (γ) is allowable in these types of experiments. According to the average relative error, two out of the nine isotherms can be used satisfactorily to predict the behavior of the adsorption of *n*-butane and *n*-heptane on 5A zeolite. These two equations are the modified virial equation (Eq. 1) and the Dubinin-Astakhov (D–A) equation (Eq. 2). The results are presented in Table 2 and Table 3.

$$\text{Modified virial equation: } P = a^n \exp(C_1 + C_2a + C_3a^2) \tag{1}$$

$$\text{D–A equation: } a = a_m \exp\{B[\ln[(T/T_c)^2P_c/P]]^n\} \tag{2}$$

From these results we can see all average relative errors are less than 6%, which is typical in data fitting. Fitting of the experimental data favors the modified virial equation slightly over the D–A equation, but the D–A equation parameters have more physical significance. For *n*-butane, the value of

TABLE 2[a]

Modeling Results of *n*-Butane with Correlation Coefficient $\rho \geq 0.99$

Temperature (K)	Modified virial equation					D–A equation			
	Parameters				γ	Parameters			γ
	C_1	C_2	C_3	n	(%)	a_m	B	n	(%)
533	1.28	2.32	−0.10	−1.03	2.87	4.29	-1.62×10^{-3}	3.51	3.83
553	4.32	−0.11	0.20	0.68	2.85	4.04	-2.22×10^{-3}	3.54	5.25
583	4.51	0.15	0.12	0.79	3.21	3.76	-1.67×10^{-3}	3.76	3.41
603	4.96	−0.25	0.27	0.85	1.65	3.39	-1.84×10^{-3}	3.71	2.99
633	2.79	3.18	−0.58	−0.48	3.36	7.53	-7.36×10^{-2}	1.96	5.76
653	5.35	0.64	−0.14	0.88	3.99	6.88	-5.37×10^{-2}	2.21	4.90

[a] Average relative error:

$$\gamma = \frac{1}{N}\sum_i \left|\frac{a_i - \bar{a}_i}{a_i}\right| \times 100\%$$

where a_i = experimental value
$\bar{a}_i$ = calculated value from isotherms
N = number of experiments

TABLE 3[a]

Modeling Results of *n*-Heptane with Correlation Coefficient $\rho \geq 0.99$

Temperature (K)	Modified virial equation					D–A equation			
	Parameters				γ	Parameters			γ
	C_1	C_2	$C_3 \times 10^3$	n	(%)	a_m	$B \times 10^5$	n	(%)
533	1.58	−1.43	8.97×10^{-2}	4.16	1.60	10.83	-1.81×10^{-4}	4.43	2.06
553	−0.96	−2.99	1.42×10^{-1}	9.94	1.94	10.53	-1.94×10^{-4}	4.46	1.70
583	0.73	−3.37	1.72×10^{-1}	9.91	0.57	9.60	-5.37×10^{-5}	5.25	0.65
603	2.45	−3.37	4.09×10^{-3}	0.13	1.52	11.03	-1.46×10^{-3}	3.63	1.54
633	2.81	0.74	-2.71×10^{-3}	−0.52	1.30	14.30	-7.83×10^{-3}	2.95	1.84
653	3.75	−0.75	5.96×10^{-3}	2.29	3.09	10.08	-6.58×10^{-4}	4.27	4.7

[a] Average relative error:

$$\gamma = \frac{1}{N}\sum_i \left|\frac{a_i - \bar{a}_i}{a_i}\right| \times 100\%$$

where a_i = experimental value
$\bar{a}_i$ = calculated value from isotherms
N = number of experiments

n in the D–A equation is close to 4.0 at temperatures lower than 603 K. When the temperature is raised above 603 K, *n* is approximately equal to 2.0 and the D–A equation transforms into the Dubinin–Radushkevich (D–R) equation which applies to micropore adsorption, in good agreement with the theory developed by Barrer (10). For *n*-heptane, the *n* value of 4.0 roughly holds for all temperatures. From the D–A equation the maximum quantity of *n*-butane adsorbed at saturation of the adsorbent (a_m) is about 4.0 g/100 g at low temperatures and about 7.0 g/100 g at higher temperatures, whereas the a_m of *n*-heptane is around 11.0 g/100 g for all temperatures. Peterson estimated the maximum number of *n*-butane molecules which can fit into one cavity is 5 while this number for *n*-heptane is 3.5 (3). Based on the effective volume of 760 Å^3 for an α cavity (which is the major cavity in type A zeolite) (11), the a_m of *n*-butane is 7.9 g/100 g and the a_m of *n*-heptane is 9.5 g/100 g. However, Doetsch et al. suggested that only two molecules of *n*-C_7 can go into one cavity in 5A zeolite (5), while Ruthven suggests that five molecules of *n*-C_3 can be accommodated (12). From our experimental data, each cavity can hold 2.5 *n*-C_4 molecules at low temperature and 4.5 molecules at high temperature. For *n*-C_7 each cavity can hold 4.0 molecules over all temperatures, consistent with the model proposed by Peterson (3).

One possible explanation for a_m increasing with increasing temperature for *n*-C_4 adsorption on 5A zeolite can be the formation of a small amount of coke at high temperatures, which was supported through measurement of the surface area and pore volume of the zeolite. The results are presented in Table 4. Coke probably exists in the porous state, or in layers, and thus the surface area of coked zeolite becomes larger than that of fresh zeolite, but the pore volume decreases. Another evidence of coke formation is that the zeolite was dotted with grey spots after adsorption at high temperatures.

Chemisorption (chemical adsorption) probably contributed to the increase of a_m of *n*-C_4 with increasing temperature as well. There is no sharp dividing line between physical adsorption and chemisorption, though the adsorption of hydrocarbons on 5A zeolite is generally classified as physical adsorption at mild conditions. However, at high temperatures, chemisorption may happen

TABLE 4
Comparison of Fresh and Coked 5A Zeolite. The Zeolite Was Used to Adsorb *n*-C_4 and *n*-C_7 for 1 Hour at 633 K

	Fresh 5A	*n*-C_4 coked 5A	*n*-C_7 coked 5A
Surface area (m^2/g)	405.1	426.9	429.5
Pore volume (cm^3/g)	0.1248	0.1228	0.1207

TABLE 5[a]
Modeling Results of *n*-Butane and *n*-Heptane by Temkin Equation with Correlation Coefficient $\rho \geq 0.99$

Temperature (K)	*n*-Butane Parameters k_t	*n*-Butane Parameters b	*n*-Butane γ (%)	*n*-Heptane Parameters k_t	*n*-Heptane Parameters b	*n*-Heptane γ (%)
533	0.74	0.12	10.15	1.87	0.65	8.58
553	0.91	0.04	9.58	2.03	0.35	8.68
583	0.88	0.03	9.87	2.11	0.19	6.86
603	0.80	0.03	6.19	2.39	0.09	5.42
633	0.79	0.02	5.98	2.84	0.04	5.45
653	0.77	0.01	4.48	2.66	0.03	5.12

[a] Average relative error:

$$\gamma = \frac{1}{N}\sum_i \left|\frac{a_i - \bar{a}_i}{a_i}\right| \times 100\%$$

where a_i = experimental value
$\bar{a}_i$ = calculated value from isotherms
N = number of experiments

in the hydrocarbons–5A zeolite system. The Temkin equation can be used to qualitatively describe chemisorption of medium coverage over a heterogeneous surface. Hence, the Temkin equation was employed to fit the data of the adsorption of *n*-butane on 5A zeolite. The results showed improved data fitting at temperatures higher than 600 K (Table 5), indicating chemisorption probably occurred at high temperature. More work is required to investigate whether the a_m increase of *n*-C4 with increasing temperature is caused by coke formation, chemisorption, or both of them.

One reason why n-C_7 has no such phenomenon is that *n*-heptane probably cokes even at low temperatures. As shown in Table 5, although the Temkin equation did not fit the experimental data as well as the modified virial equation and the D–A equation, it might as well be proposed that chemisorption of some *n*-heptane on 5A zeolite occurred at all temperatures in our system, which is probably another reason why the a_m of n-C_7 did not increase with increasing temperature.

Isosteric Heat of Adsorption

The isosteric heat, q, can be calculated from the adsorption isotherm equation or can be measured experimentally (13). The former method is more widely employed because the latter requires specialized analytical equipment. In addition to describing phase equilibrium, an adsorption model should reflect the energetics of adsorption processes. The temperature dependency of adsorption equilibrium is linked theoretically to various heats of adsorption (14). Temperature dependency is often neglected, since the sensible heat of the adsorbed phase is small compared to the much higher energy necessary to remove a molecule from the surface (15). In thermodynamics, the Clausius–Clapeyron equation is used to derive the isosteric heat of adsorption, q.

$$q = RT^2\left(\frac{\partial \ln P}{\partial T}\right)_a = -R\left(\frac{\partial \ln P}{\partial(1/T)}\right)_a \tag{3}$$

$$\int \partial \ln P = -\frac{q}{R}\int \partial(1/T) + C \tag{4}$$

where C is an integral coefficient. So

$$\ln P = -\frac{q}{R}\frac{1}{T} + C \tag{5}$$

The isosteric heat was obtained through the modified virial equation (because of improved data fitting), and is shown in Fig. 5.

At adsorption equilibrium:

$$\Delta G = \Delta H - T\Delta S = 0 \tag{6}$$

so

$$\Delta S = \frac{\Delta H}{T} = \frac{-q}{T} \tag{7}$$

where ΔG is the change of Gibbs free energy, ΔH is the change of enthalpy, and ΔS is the change of entropy.

Obviously, $-\Delta S$ varies with temperature, and similar trends of $-\Delta S$ against the amount of adsorption (a) are observed, as in Fig. 5.

Overall, at low loading the isosteric heat of *n*-heptane is larger than that of *n*-butane, in agreement with the proposed stronger interaction between molecules of *n*-heptane and the zeolite than that between *n*-butane molecules and the zeolite structure. With the increase in loading, each zeolite cavity holds more *n*-butane molecules than *n*-heptane molecules, so the interaction between *n*-butane molecules is stronger than that between *n*-heptane molecules, and correspondingly there is much more heat released for *n*-butane than for *n*-heptane. For both molecules the isosteric heat initially decreases with increasing loading, and then increases after the loading increases to a certain extent. This observation is quite similar to those of Burgess (4) and

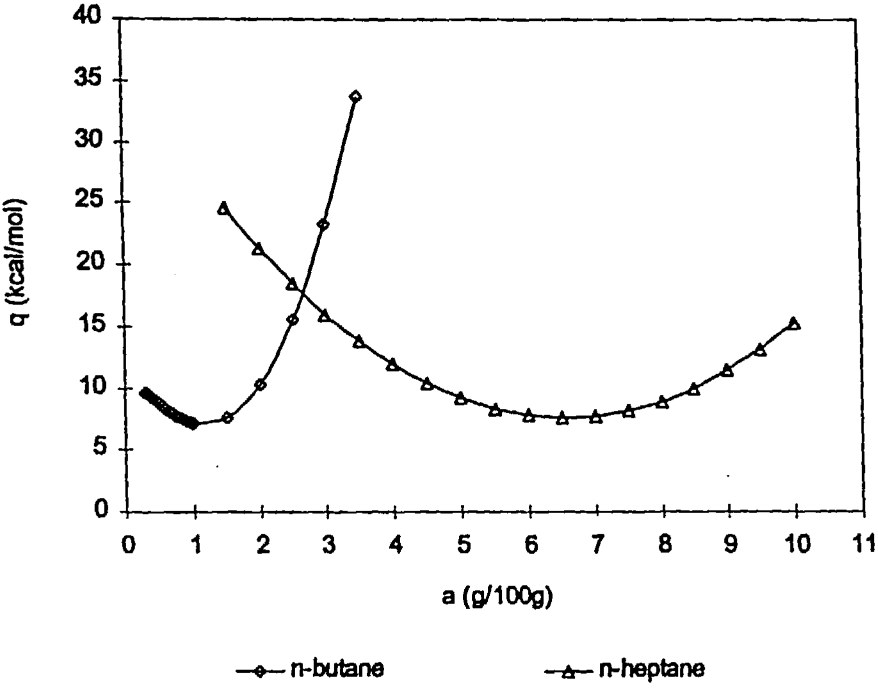

FIG. 5 Variation of heat of adsorption (q) with amount of adsorption (a) for adsorption of *n*-butane and *n*-heptane on 5A zeolite

Zhou (9), and further suggests that the interaction between adsorbate molecules and adsorbent molecules is dominant at low loading while the interaction among adsorbate molecules is more important at high loading.

One interesting feature of the adsorption of *n*-butane on 5A zeolite is that at high loading the isosteric heat of adsorption rises dramatically with increasing loading under our experimental conditions. As discussed above, the formation of coke or the chemisorption, or both of them, may contribute to this phenomenon. Whatever happens, only a small amount of *n*-butane is involved in the reaction or chemical adsorption. Otherwise, the amount of *n*-butane adsorbed would rise with increasing temperature in the isotherms. However, this small magnitude of reaction or chemisorption has a tremendous influence on the heat of adsorption.

CONCLUSION

The thermodynamics of adsorption of *n*-butane and *n*-heptane on 5A zeolite has been studied by the flow gravimetric-chromatograph technique in the temperature range applicable to the TIP process. The modified virial equation and D–A equation were chosen to correlate the experimental data, and the corresponding parameters were calculated. The modified virial equation fits the data slightly better than does the D–A equation, but the D–A equation has more physical significance. The amount of *n*-heptane adsorbed is much larger than the amount of *n*-butane under the same conditions, and the isosteric heat and entropy of *n*-heptane are higher at low loading and lower at high loading compared to the isosteric heat and entropy of *n*-butane on 5A zeolite. Coking, chemisorption, or both of them very likely occur, especially at temperatures higher than 603 K for *n*-butane adsorption, while they may be present over the entire temperature range for *n*-heptane.

NOMENCLATURE

a	amount adsorbed of adsorbate (g/100 g)
a_m	maximum amount adsorbed of adsorbate (g/100 g): $a_m = a_{m1} + a_{m2}$
a_{m1}	maximum amount adsorbed of adsorbate on one kind of active site in 2-Langmuir equation (g/100 g)
a_{m2}	maximum amount adsorbed of adsorbate on another kind of active site in 2-Langmuir equation (g/100 g)
b	constant
b_1, b_2	constants
B	D–A equation parameter
C	integral coefficient
C_1	virial parameter
C_2	virial parameter
C_3	virial parameter
G	Gibbs free energy (kcal/mol)
H	enthalpy (kcal/mol)
K_f	Freundlich constant
k_t	constant
n	constant
n	parameter of virial and D–A equations
N	number of experiments
P	partial pressure of adsorbate (mmHg)
P_c	critical pressure of adsorbate (mmHg)
q	isosteric heat of adsorption (kcal/mol)
R	ideal gas law constant (1.987 cal·K^{-1}·mol^{-1})
S	entropy (cal/mol)
T	absolute temperature (K)
T_c	critical absolute temperature (K)

Greek Symbols

γ	average relative error
ρ	correlation coefficient

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