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Adsorption of Propane and *n*-Butane on Polystyrene Adsorbents

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Summary. The adsorption isotherms of propane and *n*-butane adsorbed on two polystyrene adsorbents with high and low specific surface areas were measured in the concentration range from 500 ppm to 10 800 ppm in a helium carrier gas at 273, 298, 303, and 313 K. The isotherms can be represented by either a Freundlich or a Langmuir-Freundlich type equation. The energetic heterogeneities of the two adsorbents were characterized in terms of the dispersion of the adsorption energy. Also, the isosteric heats were calculated from the dimensionless adsorption capacities.

Keywords. Adsorption on polystyrene adsorbents; Gas adsorption; Energetic heterogeneity of solids; Hydrocarbons adsorption.

Adsorption von Propan und *n*-Butan auf Polystyrol-Adsorbentien

Zusammenfassung. Es wurden die Adsorptionsisothermen von Propan und *n*-Butan an zwei Polystyrol-Adsorbentien mit hohen und niederen spezifischen Oberflächen im Konzentrationsbereich von 500 bis 10 800 ppm in einem Helium-Trägergas bei 273, 298, 303 und 313 K gemessen. Die Isothermen können entweder mittels Gleichungen vom Freundlich- oder Langmuir-Freundlich-Typ dargestellt werden. Die energetische Heterogenität der zwei Adsorbentien wurde in Termen der Dispersion der Adsorptionsenergie charakterisiert. Es wurden auch die isosterischen Adsorptionswärmen aus den dimensionslosen Adsorptionskapazitäten berechnet.

Introduction

One of the most popular equations for describing the adsorption isotherm is the Chakravarti-Dhar or Langmuir-Freundlich equation. This equation can be simplified to the Langmuir equation or to the Freundlich equation. A discussion of the mathematical basis for deriving the Langmuir-Freundlich equation is given in the literature [1]. It can be obtained by integrating a quasi-Gaussian energy distribution [1, 2] on the solid surface with each energy patch obeying a Langmuir isotherm. With this simple picture, the exponent ν in the Langmuir-Freundlich equation represents the heterogeneities of the adsorbent. Different materials are expected to have different heterogeneities.

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Experimental adsorption isotherms are used often for characterizing sorption properties especially the energetic heterogeneity of solid adsorbents. It follows from review [2] that this characterization frequently is incomplete because of the lack of the experimental isotherms measured at various temperatures. In this study, we measured adsorption isotherms of propane and *n*-butane on two types of cross-linked polystyrene with similar chemical structures but different specific surface areas. These two polystyrene adsorbents are used often in adsorption and chromatographic studies [3, 4]. Quantities obtained from an analysis of these isotherms are the surface phase capacity, the dispersion of the adsorption energy, and the isosteric heat of adsorption. The last two quantities will be used to compare energetic heterogeneities of the samples studied. The result of this study will disclose the extent of the surface heterogeneity that originates from the pore-size distribution in these two polystyrene adsorbents.

Experimental

Adsorbents

Two cross-linked polystyrene adsorbents are Chromosorb 101 and Chromosorb 106. Adsorbent specifications supplied by the manufacturer [5] and adsorber bed parameters used in this study are listed in Table 1. These porous polymers are produced by copolymerizing monofunctional monomers with small amount of difunctional monomers. Although they have the same chemical composition (viz., cross-linked styrene-divinylbenzene type resin), their different degrees of cross-linking causes their pore sizes and surface areas to differ. The average pore diameters for Chromosorb 101 and Chromosorb 106 are 3 500 Å and 50 Å, respectively; and the surface area for Chromosorb 101 and Chromosorb 106 are 35 m^2/g and 780 m^2/g, respectively. The high surface area of Chromosorb 106 indicates that the adsorbent possesses a well-developed porous structure.

Method

The adsorption isotherms are obtained by measuring the transmission (or breakthrough) curves in a flow system. Because the apparatus used in this study was described elsewhere [6], only a brief description is given here. The flow system was constructed of stainless-steel tubings and valves with teflon seats and gaskets. The composition of the inlet gases were controlled by two valves which separately adjusted the flow rates of the pure helium and the calibrated hydrocarbon-helium mixtures. A flowmeter was used to measure the gas flow rates with an accuracy of $\pm$ 0.5%. The calibrated propane-helium and *n*-butane-helium mixtures have concentrations of 1.03% and 1.08%, respectively

Table 1. Specifications of the cross-linked polystyrene adsorbents and the parameters of the adsorber beds

	Manufacturer's specifications			Bed parameters		
Adsorbents	True specific gravity (g/cm^3)	Total pore volume (cm^3/g)	Specific surface area (m^2/g)	Length (cm)	Mass of polymer (g)	Void fraction ε
Chromosorb 101	1.095	1.03	35	24.4	8.24	0.313
Chromosorb 106	1.115	0.98	780	10.0	3.23	0.351

(He from Matheson Gas Co., East Rutherford, New Jersey, 07073). The outlet gas composition was measured at regular time intervals by a Varian 3700 gas chromatograph. A cylindrical stainless-steel adsorber bed (see Table 1 for parameters) was immersed in a constant temperature water bath. A temperature of zero degrees was maintained to within ± 0.02°C by a water-ice mixture. Other temperatures (viz., 25°C, 30°C, 40°C) were controlled within ± 0.05°C by a circulator. Before measurement, the adsorber bed was desorbed at 120°C with pure helium for 24 h. Also, between each run, the bed was desorbed for 6 h for *n*-butane and 4 h for propane. Repeated experiments verified the reproducibility of the results.

Results and Discussion

Adsorption Isotherms

The time-dependent transmission $T(t)$ is defined as the ratio of the adsorbate concentration $C(t)$ at the outlet of the adsorber bed to the inlet concentration C_0:

$$T(t) = C(t)/C_0. \tag{1}$$

The equilibrium solid-phase concentration q can be calculated from the mass-balance equation [7] when the time-dependent transmission $T(t)$ is known:

$$q(1-\varepsilon)L + C_0\varepsilon L = uC_0\varepsilon \int_0^\infty [1 - T(t)]\,\mathrm{d}t. \tag{2}$$

Here the adsorbent fraction ε of the adsorber bed indicates the voids between the polystyrene beads, L is the length of the adsorber bed, and u is the interstitial flow velocity. Equation (2) can be simplified as:

$$q = KC_0, \tag{3}$$

where

$$K = C_0\{u\varepsilon \int_0^\infty [1 - T(t)]\,\mathrm{d}t - \varepsilon L\}/[(1-\varepsilon)L] \tag{4}$$

is a dimensionless quantity called the dimensionless adsorption capacity. From a transmission curve measured at an inlet concentration C_0, the corresponding solid phase concentration q can be calculated from Eq. (2). The adsorption isotherms are generated by measuring several transmission curves at different values of the inlet concentration C_0.

The propane and *n*-butane adsorption isotherms on two polystyrene adsorbents (Chromosorb 101 and Chromosorb 106) were obtained at different temperatures (273 K, 298 K, 313 K for Chromosorb 101, and 273 K, 298 K, 303 K for Chromosorb 106). Figure 1 presents the propane isotherms at 273 K and 298 K plotted in log-log scale, while Fig. 2 shows the *n*-butane isotherms. The circles on the figures refer to the adsorption on Chromosorb 106 which has a higher specific area, whereas the triangles refer to the adsorption on Chromosorb 101 which has a lower specific area.

The adsorption isotherms measured on Chromosorb 106 at 273 K and 298 K may be described by the classical Freundlich equation [8] because they are linear on a log-log scale:

$$q = A_F C^{\nu_F} \qquad \text{for} \quad 1 \geqslant \nu_F > 0. \tag{5}$$

Here q is the solid-phase concentration that is in an equilibrium with the gas-phase concentration C, and A_F and ν_F are the two Freundlich parameters.

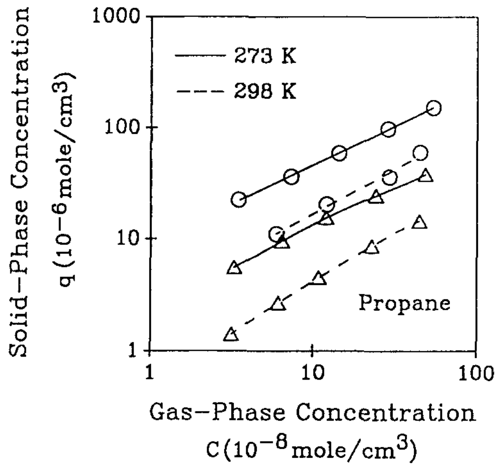

Fig. 1. Adsorption isotherms for propane on polystyrene adsorbent Chromosorb 101 (triangles) and Chromosorb 106 (circles) at 273 K (solid lines) and 298 K (dash lines)

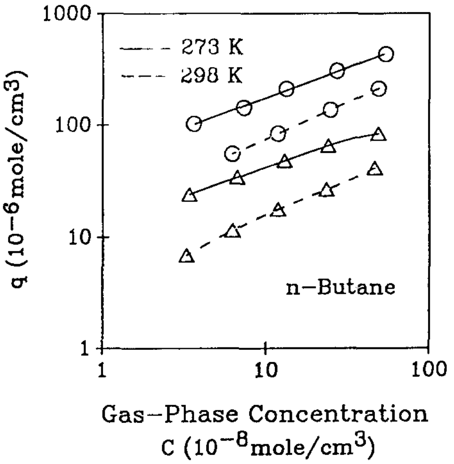

Fig. 2. Adsorption isotherms for *n*-butane on polystyrene adsorbent Chromosorb 101 (triangles) and Chromosorb 106 (circles) at 273 K (solid lines) and 298 K (dash lines)

The adsorption isotherms measured on Chromosorb 101 at 273 K and 298 K show curvatures on the log-log scale as seen in Figs. 1 and 2; thus, they can not be represented by the Freundlich power-law Eq. (5). The Langmuir-Freundlich equation [9], which was proposed first by Chakravarti-Dhar [10], can be used to fit these isotherms:

$$q = q_0(K_m C)^\nu / [1 + (K_m C)^\nu] \qquad \text{for} \quad 0 < \nu \leqslant 1. \tag{6}$$

Here q_0 is the surface phase capacity, and K_m and ν are two constants.

The adsorption isotherms measured at 303 K and 313 K can be represented also by either the Freundlich or the Langmuir-Freundlich isotherm. The best-fit parameters of the isotherms are listed in Tables 2 and 3. For all the Langmuir-Freundlich isotherms, notice that the highest solid-phase concentrations are in the range from 20% to 60% of the corresponding surface phase capacities q_0; hence, all the adsorption measurements were made in the monolayer region.

Table 2. Parameters of the adsorption isotherm of propane and *n*-butane adsorbed on cross-linked polystyrene Chromosorb 106 at 273, 298, and 303 K

		Langmuir-Freundlich parameters			Freundlich parameters	
Gas	Temperature (K)	ν	$10^{-6}K_m$ (cm^3/mol)	$10^3 q_0$ (mol/cm^3)	A_F (mol/cm^3)$^{1-n}$	ν_F
Propane	173				4.47	0.712
	298				14.5	0.846
	303	0.990	0.662	0.213	(18.0)	(0.875)
n-Butane	273				1.10	0.543
	298				2.77	0.652
	303	0.851	0.988	0.499	(4.26)	(0.692)

Table 3. Parameters of the adsorption isotherm of propane and *n*-butane adsorbed on cross-linked polystyrene Chromosorb 101 at 273, 298, and 313 K

		Langmuir-Freundlich parameters			Freundlich parameters	
Gas	Temperature (K)	ν	$10^{-6}K_m$ (cm^3/mol)	$10^3 q_0$ (mol/cm^3)	A_F (mol/cm^3)$^{1-n}$	ν_F
Propane	273	0.860	0.958	0.112		
	298[a]	0.998	0.835	0.054	(6.74)	(0.889)
	313				3.53	0.883
n-Butane	273	0.790	4.236	0.128		
	298	0.844	1.360	0.103		
	313				2.02	0.771

[a] In this case, the isotherm can be fitted either by the Langmuir-Freundlich or the Freundlich isotherm; however, the uncertainty in the energetic dispersion σ is much smaller for the Freundlich parameter ν_F

Energetic Heterogeneity of the Adsorbents

For an energetically-heterogeneous adsorbent with an adsorption energy distribution function F(u), the overall adsorption isotherm can be expressed as [11]:

$$\theta = \int_{\Omega} \theta(C, u)\,\mathrm{F}(u)\,\mathrm{d}u, \tag{7}$$

where $\theta(C, u)$ is the local adsorption isotherm, and Ω is the adsorption energy domain of the adsorbent. The adsorption energy distribution function F(u) characterizes the energetic heterogenity of the adsorbent. Theoretical studies [1, 12] indicate that the classical Freundlich isotherm Eq. (5) is associated with an exponentially-decreasing distribution function of adsorption energy. The Freundlich exponent ν_F determines the shape of the exponential distribution; with increasing values of ν_F, the exponential distribution function becomes sharper which reflects a smaller energetic heterogeneity of the adsorbent; hence, the Freundlich exponent ν_F characterizes the energetic heterogeneity. The dispersion σ of the adsorption energy corresponding to this exponential distribution function is

$$\sigma = RT/\nu_F, \tag{8}$$

where R is the universal gas constant, and T is the absolute temperature. The dispersion σ measures the energetic heterogeneity directly.

For the Langmuir-Freundlich isotherm Eq. (6), the corresponding energy distribution function is the symmetrical quasi-Gaussian distribution [9]. The parameter ν determines the shape of the distribution function. For larger value of ν, the distribution function becomes sharper which reflects a smaller energetic heterogeneity of the adsorbent. When $\nu = 1$, the Langmuir-Freundlich isotherm reduces to the Langmuir isotherm, and the symmetrical quasi-Gaussian distribution function becomes the δ-function, which means that the adsorbent surface is energetically homogeneous. It is consistent with the fact that the Langmuir isotherm equation was derived under the assumption that the adsorbent is energetically homogeneous. The dispersion σ of the adsorption energy corresponding to the symmetrical quasi-Gaussian distribution function is [13]:

$$\sigma = 3^{-1/2}\pi RT[\nu^{-2} - 1]^{1/2}. \tag{9}$$

From the isotherm parameters in Table 2 and Table 3, the energy dispersion σ of propane and n-butane adsorbed on Chromosorb 101 and Chromosorb 106 at 273 and 298 are calculated, and the results are listed in Table 4. Application of the dispersion σ for characterizing the energetic heterogeneity permits a comparison of the heterogeneities of these two polystyrene adsorbents even if the adsorption energy distribution functions have different shapes. Table 4 shows that for both propane and n-butane adsorbed on Chromosorb 106 (with the larger specific surface area), the dispersion values σ are greater than those relating to Chromosorb 101 (with the smaller specific surface area). This result means that the adsorbent Chromosorb 106 is more energetically heterogeneous than the adsorbent Chromosorb 101. Why so? From Table 1 one can see that both adsorbents have nearly the same total pore volume, and both adsorbents have the same chemical nature (i.e., chemical composition, surface heterogeneity), the only significant difference is their specific surface area, 780 m^2/g for Chromosorb 106, and 35 m^2/g for Chromosorb 101. This significant increase in the specific area of Chromosorb 106 in comparison to

Table 4. Energetic dispersion σ of two polystyrene adsorbents measured from propane and n-butane adsorption isotherm at 273 and 298 K

Gas	Temperature (K)	σ(kJ/mol)	
		Chromosorb 101	Chromosorb 106
Propane	273	2.4 ± 0.7	3.2 ± 0.2
	298	2.8 ± 0.1	2.9 ± 0.1
n-Butane	273	3.2 ± 0.7	4.2 ± 0.2
	298	2.8 ± 0.8	3.8 ± 0.2

Chromosorb 101 may be achieved by increasing the porous structure in Chromosorb 106. This porous structure may be the source of an additional heterogeneity, which makes Chromosorb 106 more energetically heterogeneous than Chromosorb 101; accordingly, the σ-values for propane and n-butane adsorbed on Chromosorb 106 are greater than those on Chromosorb 101. The existence of the well-developed porous structure in Chromosorb 106 is confirmed by the average pore diameters: 50 Å for Chromosorb 106 and 3 500 Å for Chromosorb 101. According to the definition of the International Union of Pure and Applied Chemistry (IUPAC), the pore diameter for micropores is less than 20 Å, and is between 20 Å and 500 Å for mesopores. The σ-values at 273 K and 298 K presented in Table 4 are the same within uncertainties, as they should be according to the physical meaning of the dispersion σ. The uncertainty in determining the heterogeneity parameter from the experimental isotherm is propagated through Eq. (9) for the Langmuir-Freundlich isotherm or Eq. (8) for the Freundlich isotherm.

Table 4 shows also that the σ-values for n-butane are larger than those for propane on Chromosorb 106 at 273 K and 298 K; for Chromosorb 101, this effect is obscured by the large uncertainties. Because the chemical nature of n-butane (C_4H_{10}) and propane (C_3H_8) are similar, the difference in the values of the dispersion σ may be attributed to the difference in their molecular sizes. Our results are consistent with the theoretical studies of Marczewski et al. [14], who showed that the dispersion of the adsorption energy increases when the molecular size of the adsorbate increases.

Isosteric Heats

The isosteric heat of adsorption can be extracted from the dimensionless adsorption capacity K [see Eq. (3)]. The Clausius-Clapeyron phase equation relates the enthalpy of adsorption to the vapor pressure:

$$\Delta H/RT^2 = \partial \ln p/\partial T, \tag{10}$$

where ΔH is the adsorption enthalpy, T is the absolute temperature, p is the partial pressure of the adsorbate gas, and R is the universal gas constant. The isosteric heat of adsorption ΔH_q is defined as the adsorption enthalpy at the constant adsorbed phase concentration q; hence

$$\Delta H_q = RT^2(\partial \ln p/\partial T)_q. \tag{11}$$

By assuming that the adsorbate gas behaves as an ideal gas ($p = CRT$) and engaging Eq. (2) at a constant solid-phase concentration q, the adsorbate partial pressure p can be expressed:

$$p = RTq/K. \tag{12}$$

The isosteric heat of adsorption can be expressed as:

$$\Delta H_q = R[\partial \ln (K/T)/\partial (1/T)]_q. \tag{13}$$

Over reasonable temperature intervals, the expression $\partial \ln (K/T)/\partial (1/T)$ can be approximated by $\Delta \ln (K/T)/\Delta (1/T)$, and a plot of $\log(K/T)$ versus $1/T$ should be a straight line with a slope of $\Delta H_q/2.3R$. Because the dimensionless adsorption capacity K is dependent on the solid-phase concentration, the isosteric heat of adsorption will be expected to depend on the value of q.

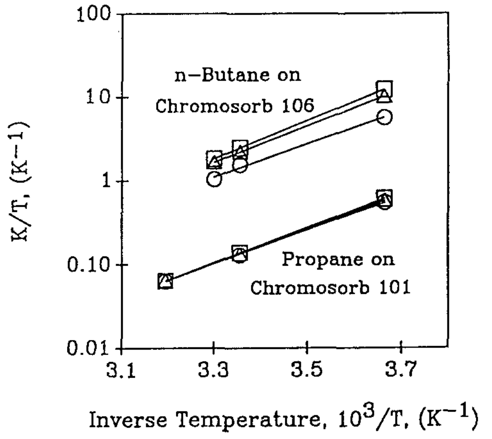

Fig. 3. The product of the adsorption capacity and the reciprocal temperature versus the reciprocal temperature for propane on Chromosorb 101 and *n*-butane on Chromosorb 106. Solid phase concentrations in μmol/cm^3 are as follows: propane 8 (circles), 6.5 (triangles), 5 (squares); *n*-butane 200 (circles), 100 (triangles), 84 (squares)

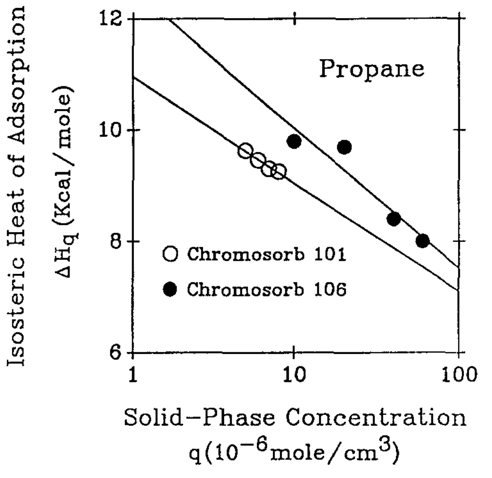

Fig. 4. Isosteric heat of adsorption versus the solid-phase concentration for propane adsorbed on Chromosorb 101 (open circles) and on Chromosorb 106 (solid circles)

The dimensionless adsorption capacities were calculated at fixed solid-phase concentrations for three different temperatures; the results are listed in Table 5. A plot of K/T versus $1/T$ is shown in Fig. 3 for *n*-butane adsorbed on Chromosorb 106 and for propane adsorbed on Chromosorb 101 at three solid phase concentrations. As shown in Fig. 3, the fact that the relations between $\log K/T$ and $1/T$ are linear means the isosteric heats are independent of the temperature in our temperature range. Using the linear regression for each $\log K/T$ versus $1/T$ curves to obtain the straight-line slopes, we calculated the isosteric heat from these slopes. The plot of isosteric heats versus solid-phase concentration for propane adsorbed on two polystyrene adsorbents is shown in Fig. 4; and the plot for *n*-butane is shown in Fig. 5.

From Fig. 4 and Fig. 5, one can see the following features: (i) The adsorption heat as a function of surface coverage differs for the two adsorbents. The fact that the linear fitting curve of the isosteric heats associated with Chromosorb 106 is above that associated with Chromosorb 101 means that the isosteric heats of propane and *n*-butane adsorbed on Chromosorb 106 are larger those of propane and *n*-butane adsorbed on Chromosorb 101. (ii) The isosteric heat decreases gradually with increasing values of the solid-phase concentration. Feature (i) can be explained

Table 5. Dimensionless adsorption capacities for propane and *n*-butane adsorbed on two polystyrene adsorbents at different temperatures

	Chromosorb 101				Chromosorb 106			
	Propane		*n*-Butane		Propane		*n*-Butane	
Temp. (K)	Concentration (ppm)	Capacity *K*	Concentration (ppm)	Capacity *K*	Concentration (ppm)	Capacity *K*	Concentration (ppm)	Capacity *K*
273	10 300	79	10 800	170	10 300	286	10 800	802
	5 133	103	5 168	273	5 140	350	5 396	1 131
	2 547	130	2 766	372	2 580	416	2 650	1 594
	1 282	149	1 344	514	1 330	495	1 322	1 945
	657	168	677	708	610	637	658	2 800
298	10 300	33	10 800	89	10 300	135	10 800	434
	5 227	39	5 415	114	5 200	153	5 515	549
	2 514	42	2 740	151	2 580	170	2 629	709
	1 336	44	1 365	185	1 290	186	1 347	886
	667	45	686	211	—	—	—	—
313[a]	10 300	19	10 800	54	10 300	113	10 800	376
or	5 043	22	5 422	72	5 160	124	5 385	487
303[a]	2 539	23	2 639	81	2 540	135	2 598	602
	1 286	24	1 372	94	—	—	1 465	692
	646	28	682	102	747	144	—	—

[a] 313 K for Chromosorb 101, 303 K for Chromosorb 106

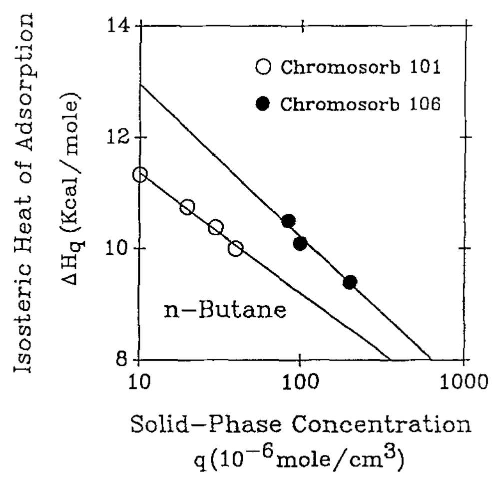

Fig. 5. Isosteric heat of adsorption versus the solid-phase concentration for *n*-butane adsorbed on Chromosorb 101 (open circles) and on Chromosorb 106 (solid circles)

by the existence of a well-developed porous structure in Chromosorb 106. If there were no such porous structure in Chromosorb 106, the adsorption heats would be the same for these two polystyrene adsorbents because their chemical nature is essentially the same. Theoretical [15] and experimental [16, 17] studies indicate that the existence of a well-developed porous structure in an adsorbent will enhance the heat of adsorption because of the overlap of the adsorption field from neighboring pore-walls. The isosteric heat of Chromosorb 106 is expected to be larger than that of Chromosorb 101 at the same surface coverage because Chromosorb 106 possesses a well-developed porous structure. Feature (ii) shows the initial decrease of the isosteric heat. It is generally accepted [18] that this initial decrease is caused by the energetic heterogeneity of the adsorbent, namely, that adsorption occurs first on the sites of the surface with the highest adsorption energy.

The heat of vaporization [19] at 298 K is 3.6 kcal/mol for propane and 5.0 kcal/mol for *n*-butane; these values are smaller than the isosteric heats. As the monolayer coverage is approached, the adsorbed phase on the surface becomes more and more like the pure adsorbate liquid; hence, the isosteric heat should approach the vaporization heat. The decreasing values of the isosteric heat in feature (ii) show this tendency, and the fact that the isosteric heat is larger than the vaporization heat indicates that the adsorption measurements made in this study are well below monolayer region, which is consistent with the conclusion mentioned above.

Conclusions

The isotherms of propane and *n*-butane adsorbed on two polystyrene adsorbents with high and low surface areas can be represented either by the Freundlich isotherm or the Langmuir-Freundlich isotherm. The energetic heterogeneities of the adsorbents, characterized by the energy dispersion value σ, are different for the two polystyrenes although they have the same chemical nature. The polystyrene with the higher specific surface area is more energetically heterogeneous than the polystyrene with the lower specific surface area because the polystyrene with the larger surface area possesses a well-developed porous structure, which introduce extra heterogeneities into the polystyrene. The isosteric heats extracted from the dimensionless adsorption capacities depend on the solid-phase concentration for both adsorbents. This dependence indicates that both polystyrenes are energetically-heterogeneous adsorbents. The isosteric heats of adsorption on the adsorbent with the larger specific area are higher than that on the adsorbent with the smaller specific area also because the larger specific area adsorbent possesses a well-developed porous structure.

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