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A New Analytical Isotherm Equation for Adsorption on Heterogeneous Adsorbents

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Abstract

A new analytical isotherm equation is derived to describe the adsorption of gases on energetically heterogeneous surfaces. It is assumed that the local isotherm for a specific site is given by the Jovanovic model, and the distribution of energetically different sites is represented by a modified Morse-type energy distribution function. The model has four parameters, the saturation adsorption capacity and three energy distribution parameters. All the model parameters are related to the Henry's law constant. The new isotherm equation was tested against published equilibrium adsorption data on several hydrocarbon-silica gel, hydrocarbon-activated carbon, and hydrocarbon-zeolite systems at different temperatures and pressures. The new model provides an accurate and simple description of adsorption of gases on heterogeneous adsorbents, and gives an excellent correlation of the experimental adsorption data for the systems tested.

INTRODUCTION

It is well known that for a large class of solid-vapor systems the surface heterogeneity plays an important role in determining adsorption characteristics, and its effects require adequate treatment. The general approach

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used to describe adsorbent heterogeneity is to postulate that the heterogeneous surface exhibits a distribution of adsorptive potentials which are either grouped in patches or distributed randomly on the surface. Even small variations in the adsorption potential have been shown to influence the adsorption behavior.

As shown by Ross and Olivier (*1*), the overall isotherm is obtained by the integration of the contribution of each patch over the energy distribution range. The adsorption isotherm is thus given as

$$Q(P,T) = \int_0^\infty Q_1(P,T,e)E(e)\, de \tag{1}$$

where $E(e)$ is the energy distribution function and $Q(P,T)$ is the overall adsorption isotherm on the heterogeneous adsorbent. The term $Q_1(P,T,e)$ describes a specific adsorption isotherm for homotattic sites of adsorptive energy e. Both Q and Q_1 are amounts adsorbed per unit mass of adsorbent. Ross and Olivier (*1*) used a number of energy distributions for $E(e)$, including a Gaussian function, and employed the two-dimensional van der Waals equation of state for the adsorption isotherm $Q(P,T)$. They solved the model equations numerically and found that experimental isotherms of argon and nitrogen on various carbon blacks and a synthetic zeolite agreed well with the predicted values obtained when using a Gaussian distribution function. A detailed review of the studies which employ this approach is given by Jaroniec et al. (*2*).

The integration limits, zero and infinity, in Eq. (1) correspond respectively to a site which has a very weak adsorption potential ($e_{\min} \approx 0$) and a site which adsorbs irreversibly ($e_{\max} \approx \infty$). If $d\varepsilon_i$ represents the fraction of surface having energies between e and $e + de$, the summation of $d\varepsilon_i$ over all permitted values of e will be equal to unity. Thus

$$\int_0^\infty d\varepsilon_i = 1 \quad \text{or} \quad \int_0^\infty E(e)\, de = 1 \tag{2}$$

Any physically meaningful distribution function should satisfy the above normalization requirement.

The model represented by Eqs. (1) and (2) has three unknown functions: $Q(P,T)$, $Q_1(P,T,e)$, and $E(e)$. Only $Q(P,T)$, however, can be measured experimentally. By using an analytical expression for the local isotherm $Q_1(P,T,e)$, $E(e)$ can be calculated from the experimental adsorption data. As shown by Ross and Morrison (*3*) and by House and Jaycock (*4*), most of the published studies have focused on numerically estimating $E(e)$ from experimental $Q(P,T)$ data by assuming some well-known expression for $Q_1(P,T,e)$ such as the Langmuir model, the step isotherm, or the virial equation of state. However, analytical expressions for $E(e)$ were obtained by Misra (*5*) and Jaroniec (*6*) by assuming that the overall adsorption isotherm $Q(P,T)$ could be represented by some well-known isotherm model, and the local isotherm $Q_l(P,T,e)$ could be described by either the Langmuir model or the step isotherm.

Several studies have been carried out to develop analytical functions for $Q(P,T)$ since they are useful for data extrapolation and the modeling of separation processes by adsorption. Among these, Misra (*7, 8*) derived analytical expressions which employed expontential and constant energy distribution functions in conjunction with either the Langmuir or the Jovanovich isotherm equations to represent the specific isotherm for homotattic sites. The overall isotherm equations he obtained using different local isotherm equations were found to be comparable provided that the distribution functions were the same. Cerofolini et al. (*9*) used the Langmuir local isotherm but incorporated a condensation approximation as the adsorption energy distribution to obtain an expression for the overall isotherm equation. In contrast to the models developed by Misra, Cerofolini's equation reduced to the Henry's isotherm in the very low pressure limit. However, it was not tested using real data. Sircar (*10*) developed an expression for $Q(P,T)$ by assuming that the Langmuir model represented the local isotherm and the Gamma probability density function described the energetic heterogeneity of the adsorbent. Sircar's model successfully described the adsorption of various gases on activated carbons and zeolites over large ranges of pressure and temperature. However, the resulting equation is very cumbersome to use. Sircar (*11*) later developed a simpler model which had the same degree of versatility by using the Jovanovich local isotherm model in conjunction with the Gamma energy distribution function. Sircar analyzed the effect of the choice of the local isotherm on the adsorbent heterogeneity and reported that both models were capable of accurately describing the overall adsorption data. His work and the work of others suggests that the choice of the energy distribution function has a greater impact on the overall outcome than does the selection of the local isotherm equation.

In the present work a new analytical expression is developed to describe the adsorption of gases on energetically heterogeneous surfaces. The new isotherm equation is developed using the Jovanovic model as the local isotherm in conjunction with a modified Morse-type potential to describe the energy distribution of the heterogeneous surface. The new model is tested using published experimental data on several systems involving the adsorption of a number of hydrocarbons and carbon dioxide on silica gel, activated carbon, and Molecular Sieve.

THEORY

The general adsorption isotherm model for energetically heterogeneous surfaces developed in the present work is represented by Eqs. (1) and (2). The local isotherm for a homogeneous surface is given by the Jovanovic (*12*) equation as

$$Q_1(P,T,e) = A[1 - \exp(-bP)] \tag{3}$$

where A is the adsorption capacity at saturation and b is the Jovanovic parameter given by

$$b = b_0 \exp(q/RT) \tag{4}$$

The constant b_0 corresponds to the limiting value of b as the temperature approaches infinity. The terms q and R are the isosteric heat of adsorption and the universal gas constant, respectively. As the pressure approaches zero, the Jovanovic equation reduces to the Henry's law constant as follows:

$$\left[\frac{\partial Q_1}{\partial P}\right]_T = Ab = K_L(T) \quad \text{or} \quad K_L(T) = Ab_0 \quad \text{as} \quad T \rightarrow 0 \tag{5}$$

The value of b may vary from b_0 to infinity because, for an energetically heterogeneous adsorbent, the isosteric heat of adsorption can vary between zero to infinity. This corresponds to values for e of zero and infinity, respectively. Sircar (*11*) defined the following new energy parameter, g, which also varies between zero and infinity as q approaches zero and infinity, repsectively:

$$g = b_0[\exp(q/RT) - 1] \tag{6}$$

In the present work it is assumed that each site of the heterogeneous surface is characterized by a specific value of g, and the energy distribution on the adsorbent can be described by a modified Morse-type probability density function of g (*13*).

$$E(g) = K_0[e^{-K_1 g} - K_2 e^{-K_3 g}], \quad 0 \leq g \leq \infty \tag{7}$$

$$\int_0^\infty E(g)dg = 1 \tag{8}$$

In Eq. (7) the parameters K_0 through K_3 are all functions of temperature. However, following the normalization requirements imposed by Eq. (8), the constant K_0 can be expressed in terms of the other constant as

$$K_0 = \frac{K_1K_3}{K_3 - K_1K_2} \tag{9}$$

Equations (6) through (8) can be combined to give

$$\int_0^\infty K_0[e^{-K_1g} - K_2e^{-K_3g}][g + b_0]d\left(\frac{q}{RT}\right) = 1 \tag{10}$$

As shown by Eq. (10), the probability function for the heterogeneous adsorbent is written in terms of q/RT as

$$E\left(\frac{q}{RT}\right) = \frac{K_1K_3}{K_3 - K_1K_2}[e^{-K_1g} - K_2e^{-K_3g}][g + b_0] \tag{11}$$

where the relationship between q and q is given by Eq. (6). The above equation suggests that $E(q/RT)$ has a skewed Gaussian-like shape for positive values of K_1. Furthermore, since b_0 is usually a very small quantity [less than 10^{-4} for most practical systems as demonstrated by Sircar (*10*)], the function $E(q/RT)$ goes to zero as q approaches zero, even for K_1 approaching zero. In the limit as K_1 approaches infinity, the term $E(q/RT)$ approaches the Dirac-Delta function, representing an energetically homogeneous adsorbent. The distributions of $E(g)$ and $E(q/RT)$ for different values of K_1 are illustrated in Figs. 1 and 2, respectively. As shown in Fig. 2, a bimodel type energy distribution of the surface is observed for certain values of K_1. Also, the shape of the energy distribution curves exhibit a strong dependence on the values of K_1.

The overall isotherm can be obtained by expressing Eq. (1) as

$$Q = \int_0^\infty Q_1E(g)dg \tag{12}$$

Substituting Eqs. (3), (4), (6), (7), and (9) into Eq. (12) and integrating yields the following analytical expression for the overall adsorption isotherm for a heterogeneous adsorbent:

$$Q(P,T) = A\left[1 - e^{-b_0P}\frac{K_1K_3}{K_3 - K_1K_2}\left(\frac{1}{P + K_1} - \frac{K_2}{P + K_3}\right)\right] \tag{13}$$

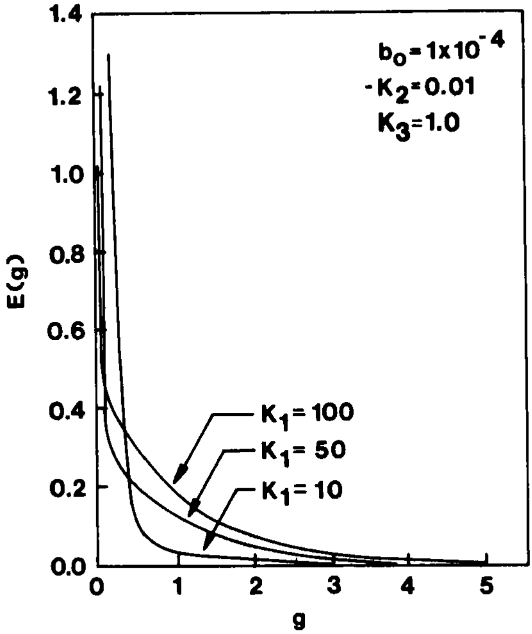

FIG. 1. Plots of energy distribution function for different K_1 in g domain.

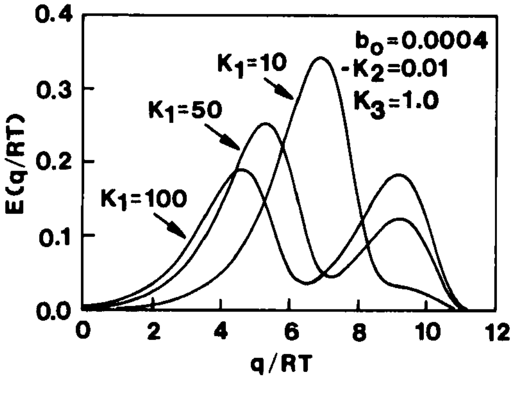

FIG. 2. Plots of probability function for different K_1 in q/RT domain.

It follows from Eq. (13) that

$$Q(P,T) = 0 \text{ as } P \to 0 \tag{14}$$

and

$$Q(P,T) = A \text{ as } P \to \infty \tag{15}$$

Also

$$\left[\frac{\partial Q}{\partial P}\right]_T = A\left[b_0 + \frac{K_3^2 - K_1^2 K_2}{K_1 K_3 (K_3 - K_1 K_2)}\right] = K_L(T) \text{ as } P \to 0 \tag{16}$$

It can be clearly seen from Eq. (16) that the adsorption isotherm given by Eq. (13) reduces to the linear isotherm in the limit as P approaches zero, and the model parameters are related to the Henry's law constant, K_L. Thus, the isotherm equation developed in the present work satisfies the requirements imposed by the physics of adsorption thermodynamics.

As the temperature approaches infinity, the local Jovanovic isotherm reduces to

$$Q_l = A[1 - \exp(-b_0 P)] \tag{17}$$

By substituting Eqs. (7), (9), and (17) into Eq. (12), it can be shown that the overall isotherm Q has the same form as Eq. (17) in the limit as T becomes infinitely large. The existence of such homogeneous type behavior for a heterogeneous adsorbent at high temperatures was previously pointed out by Ross and Olivier (*1*).

As the temperature increases and approaches infinity, the term Ab_0 in Eq. (16) becomes very small due to the fact that physical adsorption is an exothermic process. Because b_0 is usually a very small quantity ($b_0 \approx 10^{-4}$), $b_0 P$ is very small (much less than unity) in moderate pressure ranges. Thus, for all practical purposes, Eqs. (13) and (16) can be approximated as

$$Q(P,T) = A\left[1 - \frac{K_1 K_3}{K_3 - K_1 K_2}\left(\frac{1}{P + K_1} - \frac{K_2}{P + K_3}\right)\right] \tag{18}$$

and

$$A\left[\frac{K_3^2 - K_1^2K_2}{K_1K_3(K_3 - K_1K_2)}\right] = K_L(T) \text{ as } P \to 0 \tag{19}$$

The adsorption isotherm given by Eq. (18) describes the equilibrium adsorption data for gases on energetically heterogeneous adsorbents. The model has four parameters: the saturation capacity (A) and three energy distribution parameters (K_1, K_2, and K_3). All of these parameters are related the Henry's law constant through Eq. (19). Therefore, the number of adjustable parameters can be reduced to three provided that good low pressure isotherm data are available to determine the Henry's law constant.

TEST OF NEW ADSORPTION ISOTHERM

The accuracy of the new isotherm equation was evaluated by using published experimental data for several systems. Twelve adsorbate-adsorbent pairs over a wide range of pressures were correlated and the majority of the pairs were analyzed at three different temperatures. The four adjustable model parameters were determined for each data set by using a nonlinear regression analysis.

The systems used for testing the model included the adsorption data of methane, ethane, ethylene, and carbon dioxide on BPL activated carbon (*14, 15*); carbon dioxide on MSC V activated carbon (*16*); propane, propylene, ethylene,and acetylene on silica gel (*17, 18*); ethylene, carbon dioxide, and isobutane on 13X Molecular Sieve (*19*). In all of these systems the adsorbents appeared to be heterogeneous in nature as indicated by the inability to represent the experimental data with the available isotherm models for homogeneous surfaces. For illustrative purpose, the Jovanovic equation was used and a plot of ln $(1 - Q/A)$ vs P for various gases adsorbed on BPL activated carbon at 260.2 K is presented in Fig. 3. According to the Jovanovic model, such plots should yield a straight line for homogeneous adsorbents. The nonlinear nature of adsorption plots suggests that the adsorbent in question is heterogeneous.

Figures 4 through 12 show the experimental data and the best fit curves obtained from the new heterogeneous isotherm equation for the adsorption systems studied. From the analysis of these figures it is seen that the new heterogeneous model represents the isotherm data of several gases

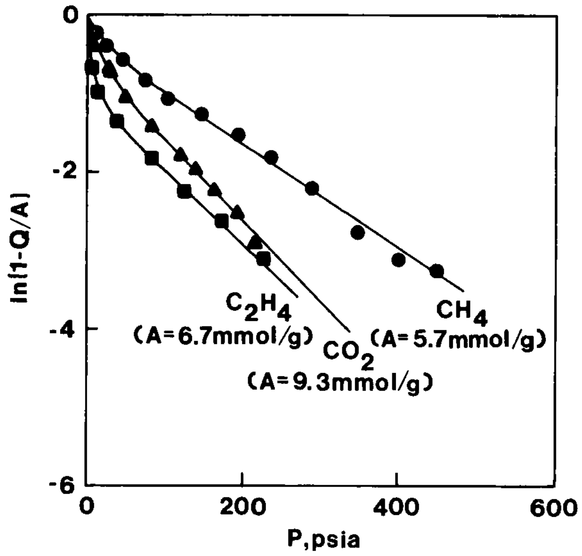

FIG. 3. Linearized Jovanovic plots for various gases adsorbed on BPL carbon at 260.2 K.

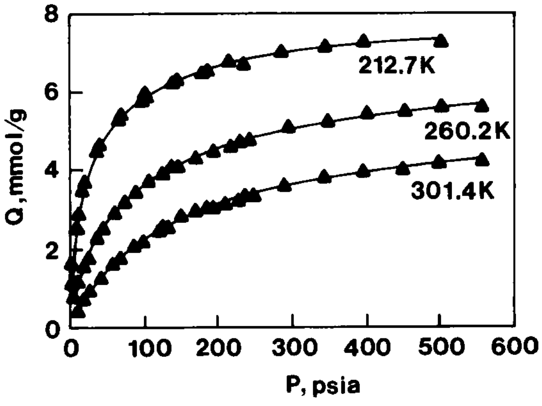

FIG. 4. Adsorption of methane on BPL activated carbon.

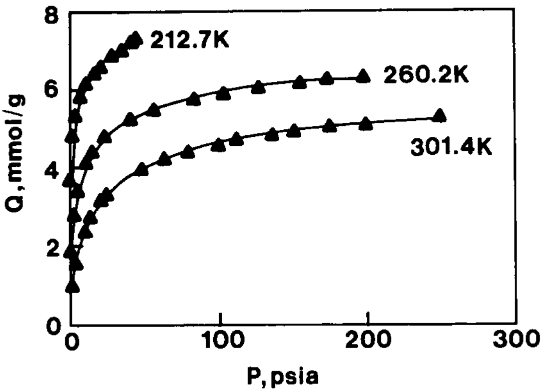

FIG. 5. Adsorption of ethane on BPL activated carbon.

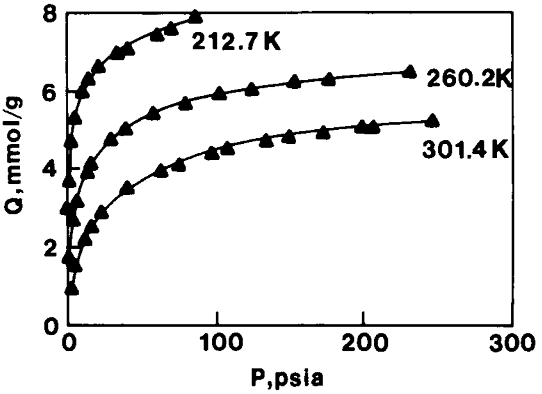

FIG. 6. Adsorption of ethylene on BPL activated carbon.

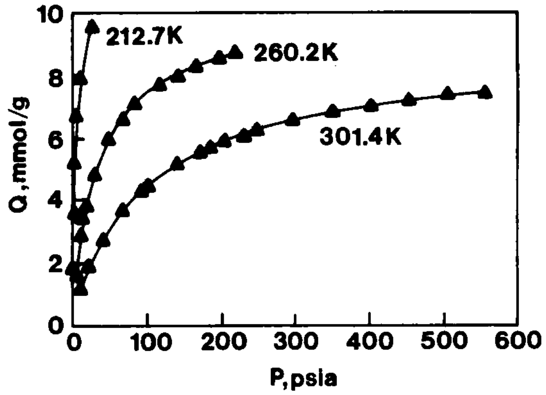

FIG. 7. Adsorption of carbon dioxide on BPL activated carbon.

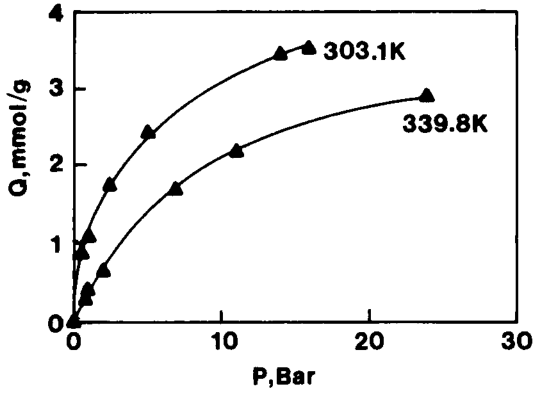

FIG. 8. Adsorption of carbon dioxide on MSC V activated carbon.

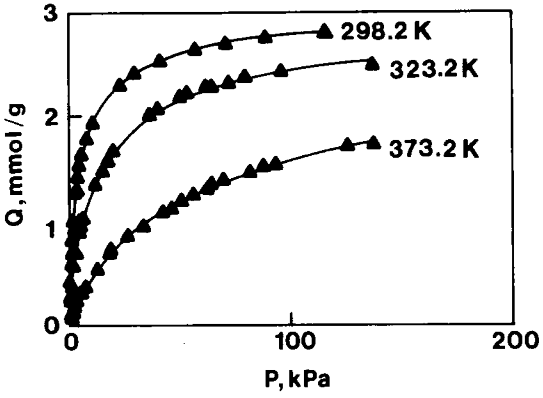

Fig. 9. Adsorption of ethylene on 13X Molecular Sieve.

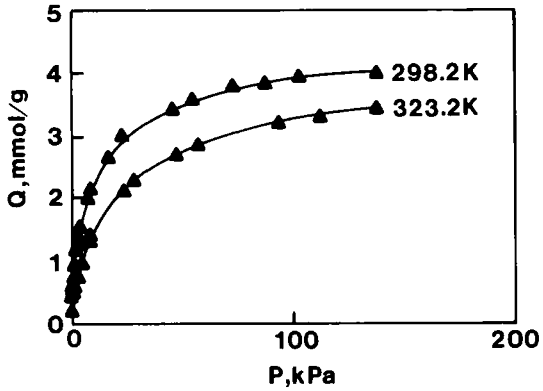

Fig. 10. Adsorption of carbon dioxide on 13X Molecular Sieve.

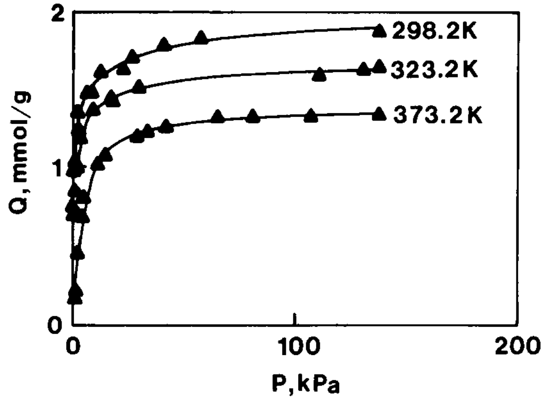

FIG. 11. Adsorption of isobutane on 13X Molecular Sieve.

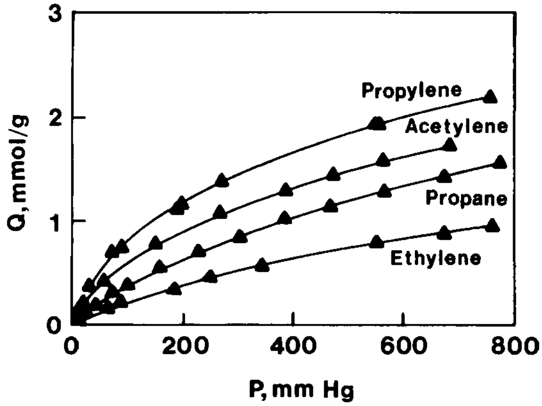

FIG. 12. Adsorption of ethylene, acetylene, propylene, and propane on silica gel at 298.2 K.

and vapors on various adsorbents extremely well over a wide range of pressure and temperature. For the majority of the adsorption systems, the average absolute percent deviation over the entire pressure range was found to be less than 2%.

The best fit model parameters for each system are presented in Tables 1 through 4. The parameters K_1 and K_3, which have units of pressure, were found to be strongly temperature-dependent, both increasing with increasing temperature. The parameter A, which has concentration units, also proved to be temperature-dependent. Its dependence, however, was observed to be weak, decreasing slightly with increasing temperature. On the other hand, the dimensionless parameter K_2 exhibited the least temperature-dependence. The consistency of the temperature-dependence of the model parameters suggests that these parameters may be correlated with respect to temperature so that the data can be interpolated from one temperature to another. The new heterogeneous adsorption equation reduces to a linear isotherm as pressure approaches zero. Thus Henry's law constants calculated for the systems employed are also presented in Tables 1 through 4.

The analysis of the numerical results summarized in Figs. 4 through 12 and in Tables 1 through 4 confirms that the heterogeneous adsorption model developed by employing the Jovanovic isotherm and a modified Morse-type energy distribution function provides an accurate and simple description of the adsorption characteristics of gases and vapors on heterogeneous adsorbents. In addition, it gives an excellent correlation of the experimental data for a large variety of systems.

CONCLUSIONS

A new analytical isotherm equation is developed to describe monolayer adsorption on energetically heterogeneous surfaces. The isotherm is developed by postulating that the adsorbent surface consists of a distribution of energetically different sites which can be represented by a Morse-type energy distribution function with the Jovanovic equation describing the specific isotherm for homotattic sites. The proposed equation reduces to a linear form in the low pressure region and can be used to obtain Henry's law constants.

The new model successfully represents experimental adsorption isotherm data for several gases and vapors on different heterogeneous adsorbents over a wide range of temperatures and pressures. Furthermore, it is

TABLE 1

The Model Parameters and the Henry's Law Constants for Methane, Ethane, and Ethylene on BPL Activated Carbon

Adsorbate	Temperature (K)	A (mmol/g)	K_1 (psia)	K_2 (−)	K_3 (psia)	K_L (mmol/g-psia)	Absolute average % deviation
Methane	301.4	5.41	161.07	−0.0057	12.23	0.0621	0.70
	260.2	6.50	112.76	−0.0235	10.14	0.1785	0.68
	212.7	7.97	78.05	−0.0422	4.33	0.8542	1.02
Ethane	301.4	5.56	31.54	−0.0424	2.54	0.8705	1.09
	260.2	6.55	25.40	−0.0434	1.00	3.5571	0.65
	212.7	7.10	3.62	−0.0442	0.12	34.6504	1.87
Ethylene	301.4	5.82	46.04	−0.0334	3.48	0.6002	0.72
	260.2	6.77	27.09	−0.0346	1.26	2.4353	0.66
	212.7	8.20	16.25	−0.0351	0.39	12.6922	0.86

TABLE 2
The Model Parameter and the Henry's Law Constants for Carbon Dioxide on BPL and MSC V Activated Carbons

Adsorbate	Temperature (K)	A (mmol/g)	K_1 (psia)	K_2 (-)	K_3 (psia)	K_L (mmol/g-psia)	Absolute average % deviation
CO_2-BPL Carbon	301.4	8.90	134.08	−0.0260	16.94	0.1448	0.35
	260.2	10.25	52.41	−0.0344	5.77	0.5719	0.13
	212.7	11.36	7.60	−0.0417	0.75	5.5084	0.23
CO_2-MSC V Carbon	339.8	3.93	7.78[a]	−0.0437	2.99[a]	0.4017[b]	1.03
	303.1	4.67	6.23[a]	−0.0018	0.07[a]	9.6715[b]	2.28

[a] Bar.

[b] mmol/g-bar.

TABLE 3
The Model Parameters and the Henry's Law Constants for Ethylene, Carbon Dioxide, and Isobutane on 13X Molecular Sieve.

Adsorbate	Temperature (K)	A (mmol/g)	K_1 (kPa)	K_2 (−)	K_3 (kPa)	K_L (mmol/g-kPa)	Absolute average % deviation
Ethylene	373.2	2.51	71.99	−0.0162	6.74	0.0847	2.42
	323.2	2.90	24.73	−0.0235	1.63	0.5541	2.40
	298.2	2.96	10.83	−0.0413	0.77	1.5903	1.54
Carbon dioxide	323.2	4.03	31.43	−0.0104	1.37	0.6702	1.98
	298.2	4.36	16.60	−0.0114	0.55	2.2390	1.61
Isobutane	373.2	1.41	279.00	−0.4380	3.47	0.3942	1.76
	323.2	1.70	27.23	−0.0613	0.43	3.1932	1.88
	298.2	1.99	25.36	−0.0145	0.15	9.6460	1.88

TABLE 4
The Model Parameters and Henry's Law Constants for Ethylene, Acetylene, Propylene, and Propane on Silica Gel at 298.2 K

Adsorbate	A (mmol/g)	K_1 (mmHg)	K_2 (−)	K_3 (mmHg)	K_L (mmol/g-mmHg)	Absolute average % deviation
Ethylene	2.35	1151.0	−0.0007	39.01	0.00320	2.02
Acetylene	3.70	1031.7	−0.0052	37.47	0.01550	2.02
Propylene	4.34	1149.7	−0.0152	67.4	0.01626	2.08
Propane	4.80	1982.7	−0.0041	103.5	0.00562	0.75

easy to use, and it provides a useful tool for accurately correlating isotherm data on heterogeneous surfaces.

NOTATION

A	saturation adsorption capacity
b	Jovanovic parameter as defined in Eq. (4)
b_0	limiting value of b at infinite temperature
e	energy of a particular site
E	energy distribution function
g	energy parameter defined by Eq. (6)
K_0	parameter in the energy distribution function
K_1	parameter in the energy distribution function
K_2	parameter in the energy distribution function
K_3	parameter in the energy distribution function
K_L	Henry's law constant
P	system pressure
Q	overall adsorption isotherm
Q_l	local adsorption isotherm
q	isosteric heat of adsorption
R	universal gas constant
T	system temperature

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