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Equations of Adsorption Isotherms for Heterogeneous Surfaces

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The paper gives the accurate equations of adsorption isotherms for a heterogeneous surface with the following distribution functions of sites with respect to the heats of adsorption:

$$N_g = \exp\ (-g/g_m) \qquad \text{(a)}$$

$$N_g = \exp\ (g/g_m) \qquad \text{(b)}$$

(g = heat of adsorption; g_m = a constant). It describes a method of determining adsorption isotherms in the case of symmetric and nonsymmetric distribution functions with a maximum, as obtained by combining distribution functions (a) and (b).

The corresponding relationships between heats of adsorption and surface coverages are given.

INTRODUCTION

The theoretical interpretation of the Freundlich adsorption isotherm was given by Zeldovitch (*1*) in 1935 on the basis of a model of a heterogeneous surface with a distribution function of sites with respect to the heats of adsorption:

$$Ng = \exp\ (-g/g_m) \qquad (1)$$

where g = heat of adsorption and g_m = a constant.

However, the problem of the theoretical form of the Freundlich equation and of the corresponding site distribution function is still being discussed in the literature.

The solution given by Zeldovitch (*1*) and later by Temkin and Levitch (*2*), Halsey and Taylor (*3*), and Halsey (*4*) on the basis of the distribution (1) is approximate since they performed integration over heats of adsorption from plus infinity to minus infinity.

Therefore the theoretical equation of the isotherm has a number of serious deficiencies; for instance, it gives infinitely large coverage for $p \to \infty$ (p = gas pressure).

Sips (*5,6*) chose to construct an "improved" isotherm by selecting mathematical expressions which reduce to the Freundlich isotherm at low pressures and unity at infinitely high pressures. Applying to these expressions a method similar to that of Temkin and Levitch (*2*), Sips found the appropriate site distribution functions, which differed from (1). It followed from (*1–6*) that Freundlich adsorption isotherms could be obtained for surfaces with different distribution functions of adsorption sites with respect to the heats of adsorption.

Thus, one cannot use Freundlich's approximate theoretical equations to give a sufficiently correct idea of the properties of the adsorbent surface.

A more reliable conclusion as to the nature of the distribution of sites with respect to the heats of adsorption based on experimental data can be drawn only with more accurate theoretical equations of adsorption isotherms for heterogeneous surfaces.

For this it is necessary, in the first place, to perform more accurate integration of the initial isotherm equations corresponding both to the site distribution function (1) and to other elementary site distribution functions.

This paper presents and discusses the accurate (in the mathematical sense) equations of the adsorption isotherm[1] corresponding to the distribution (1)[2] and of Temkin's (*8*) negative-power isotherm corresponding to the distribution function[3].

$$Ng = \exp\,(g/g_m). \qquad (2)$$

1. THE ACCURATE EQUATION OF THE ISOTHERM CORRESPONDING TO THE DISTRIBUTION FUNCTION (1)

The mean coverage θ of a heterogeneous surface is

$$\theta = \int_{g_l}^{g_u} \theta_g N_g dg \Big/ \int_{g_l}^{g_u} N_g dg, \qquad (3)$$

where θ_g is the coverage of sites with the heat of adsorption g, as given by Langmuir's equation

$$\theta_g = [1 + (p_o/p)\exp\,(-g/RT)]^{-1} \qquad (4)$$

Here $p_o = \exp\,(\Delta S/R)$ where ΔS (assumed to be independent of θ_g) is the entropy change in adsorption corresponding to $\theta_g = 0.5$. g_u and g_l are the boundary points of the distribution curve.

Write the initial isotherm equation as follows:

$$\theta = \int_{g_l}^{g_u} \exp\,(-g/g_m)\,[1 + (p_o/p)\exp\,(-g/RT)]^{-1}\,dg \Big/ \int_{g_l}^{g_u} \exp\,(-g/g_m)dg. \qquad (5)$$

Assuming $g_u = \infty$, $g_l = -\infty$ and $\int_{g_l}^{g_u} \exp\,(-g/g_m)dg = g_m$, we obtain Freundlich's theoretical isotherm (1-4). Perform more accurate integration.

Denoting $RT/g_m = \mu$, $\exp\,(g_u - g_l)/g_m = L$, $p_o\exp\,(-g_u/RT) = p_u$, $p_o\exp\,(-g_l/RT) = p_l$ and substituting $(p_o/p)\exp\,(-g/RT) = x$ we obtain

$$\theta = \mu\left[\frac{(p/p_l)^{\mu}\int_o^{x_l}(x^{\mu-1}/(1+x))\,dx}{1-L^{-1}} - \frac{(p/p_u)^{\mu}\int_o^{x_u}(x^{\mu-1}/(1+x))\,dx}{L-1}\right]. \qquad (6)$$

It has been found (*10*) that

$$\int_o^u \frac{x^{\mu-1}}{1+x}\,dx = \frac{u^{\mu}}{\mu}F(1,\mu;\,\mu+1;\,-u). \qquad (7)$$

Thus

$$\theta = \frac{F(1,\mu;\,\mu+1;\,-p_l/p)}{1-L^{-1}} - \frac{F(1,\mu;\,\mu+1;\,-p_u/p)}{L-1}. \qquad (8)$$

$F(1,\mu;\,\mu+1;\,-p_i/p)$ is a hypergeometric function expressed by the infinite series

$$F(1,\mu;\,\mu+1;\,-p_i/p) = 1 - \frac{\mu}{\mu+1}\left(\frac{p_i}{p}\right) + \frac{\mu}{\mu+2}\left(\frac{p_i}{p}\right)^2 \ldots = \sum_{n=0}^{n=\infty}(-1)^n\frac{\mu}{\mu+n}\left(\frac{p_i}{p}\right)^n \qquad (9)$$

The series (9) converges only for $p_i/p < 1$. Since p_l is always greater than p_u, Eq (9) yields the finite value of θ only in the range of $p > p_l$, i.e., for very high coverages. To obtain the isotherm equation in the range of $p < p_l$ it is necessary to find the analytical continuation of the

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[1] The consideration is based on the Langmuir equation (lateral interaction is ignored).

[2] A short communication was published [Ref. (*7*)].

[3] A short communication was published [Ref. (*9*)].

TABLE 1

Pressure range	Isotherm equations
$p < p_u$	$\theta = \dfrac{(p/p_l) \cdot [\mu/(\mu - 1)]F(1,1 - \mu; 2 - \mu; -p/p_l)}{1 - L^{-1}} - \dfrac{(p/p_u)[\mu/(\mu - 1)]F(1,1 - \mu; 2 - \mu; -p/p_u)^a}{L - 1}$
$p_u < p < p_l$	$\theta = \dfrac{(\mu\pi/\sin \mu\pi)(p/p_l)^{\mu} + (p/p_l)[\mu/(\mu - 1)] \cdot F(1,1 - \mu; 2 - \mu; -p/p_l)}{1 - L^{-1}}$ $\dfrac{F(1,\mu; \mu + 1; -p_u/p)}{L - 1}$
$p > p_l$	$\theta = \dfrac{F(1,\mu; \mu + 1; -p_l/p)}{1 - L^{-1}} - \dfrac{F(1,\mu; \mu + 1; -p_u/p)}{L - 1}$

[a] $(p/p_l)^{\mu}(1 - L^{-1})^{-1} - (p/p_u)^{\mu}(L - 1)^{-1} = 0.$

hypergeometric function[4] (9) into the range of $p < p_l$.

Below we give the final result of the calculation (7) made on the basis of Ref. (*11*). The analytical continuation of the hypergeometrical function $F(1, \mu;\ 1 + \mu;\ -p/p_l)$ is the function

$$\frac{\mu\pi}{\sin \pi\mu}\left(\frac{p}{p_l}\right)^{\mu} + \frac{\mu}{\mu - 1} F(1,1 - \mu;\ 2 - \mu;\ -p/p_l),$$

where

$$F(1,1 - \mu;\ 2 - \mu;\ -p/p_l) = 1 - \frac{1 - \mu}{2 - \mu}\left(\frac{p}{p_l}\right) + \frac{1 - \mu}{3 - \mu}\left(\frac{p}{p_l}\right)^2 \cdots = \sum_{n=0}^{n=\infty} (-1)^n \frac{1 - \mu}{n + 1 - \mu}\left(\frac{p}{p_l}\right)^n. \quad (10)$$

In its turn, the series (10) converges only at $p/p_l < 1$.

Thus, the isotherm can be described by three equations, each of which yields a finite value of θ only within a restricted pressure range:

(a) $p < p_u$, (b) $p_u < p < p_l$, (c) $p > p_l$.

All the equations are listed in Table 1. These equations completely describe the isotherm from $p = 0$ to $p = \infty$ and from $T = 0$ to $T = \infty$.

The isotherm equation for low coverages can be reduced to the Henry equation. The isotherm equation for high coverages can be reduced to the equation $\theta = 1 - (\mu/(\mu + 1))\ (p_l/p)$.

Consider the range $p_u < p < p_l$ more carefully. The coverage value corresponding to the lower boundary of this range depends on g_u and μ values, but the coverage value corresponding to the upper boundary of this range depends exclusively on μ. These (boundary) values of θ are as follows: 0.94 ($\mu = 0.1$); 0.85 ($\mu = 0.3$); 0.79 ($\mu = 0.5$); 0.71 ($\mu = 0.9$); 0.66 ($\mu = 1.3$).

In this important pressure range we obtain:

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[4] The hypergeometric functions tend to 1 as $p_l/p \to 0$ [Eq. (9)] and $p/p_l \to 0$ [Eq. (10)]. Already at $(p_l/p) = 0.1$ $F(1, \mu;\ \mu + 1;\ -p_l/p)$ is equal to 0.991 ($\mu = 0.1$); 0.968 ($\mu = 0.5$); 0.947 ($\mu = 1.3$). At $(p/p_l) = 0.1$ $F(1,\ 1 - \mu;\ 2 - \mu;\ -p/p_l)$ is 0.956 ($\mu = 0.1$); 0.961 ($\mu = 0.3$); 0.968 ($\mu = 0.5$); 0.991 ($\mu = 0.9$); 1.01 ($\mu = 1.1$). The only exceptions are cases where the values of μ are in the vicinity of integers 2, 3, 4, . . . , n. Thus, the hypergeometric functions on whose properties all these calculations are based can often be taken equal to 1.

$$\theta = \frac{(\mu\pi/\sin \mu\pi)(p/p_l)^{\mu}}{1 - L^{-1}} + \frac{(\mu/\mu - 1)\ (p/p_l)\ F(1,1-\mu;\ 2-\mu;\ -p/p_l)}{1 - L^{-1}} - \frac{F(1,\mu;\ 1+\mu;\ -p_u/p)}{L - 1}. \quad (11)$$

The upper limit g_u (as p_u) does not figure in the two first terms of this Eq. and the lower limit does not figure in the third term.

Therefore Eq. (11) can be easily simplified.

At $p \gg p_u$ F in the third term can be taken equal to 1:

$$\theta = (1 - L^{-1})^{-1} \left[\frac{\mu\pi}{\sin \mu\pi}\left(\frac{p}{p_l}\right)^{\mu} + \frac{\mu}{\mu - 1}\left(\frac{p}{p_l}\right) F\left(1,1-\mu;\ 2-\mu;-\frac{p}{p_l}\right) - L^{-1}\right]. \quad (12)$$

Further, if L is large enough, this term may be neglected, which corresponds to taking the upper limit equal to infinity

$$\theta = \frac{\mu\pi}{\sin \mu\pi}\left(\frac{p}{p_l}\right)^{\mu} + \frac{\mu}{\mu - 1}\left(\frac{p}{p_l}\right) F(1,1-\mu;\ 2-\mu;\ -p/p_l). \quad (13)$$

In many cases (if $p \ll p_l$) F in Eq. (13) can be taken to be equal to 1. Hence, the following equation may often be regarded as the sufficiently accurate approximation:

$$\theta = \frac{\mu\pi}{\sin \mu\pi}\left(\frac{p}{p_l}\right)^{\mu} + \frac{\mu}{\mu - 1}\,\frac{p}{p_l}. \quad (14)$$

Equation (14) may be interpreted as an algebraic sum of two terms: Freundlich's and Henry's.

At $\mu < 1$ Freundlich's term is always the larger; the smaller the ratio p/p_l and the smaller μ, the better is the Freundlich equation satisfied.

At $\mu > 1$ the Henry term is always the greater; the Henry equation is the better satisfied, the larger is μ and the smaller the ratio p/p_l.

Theoretical isotherms calculated by Eq. (13) (i.e., with the condition $g_u = \infty$) are given in Fig. 1a.

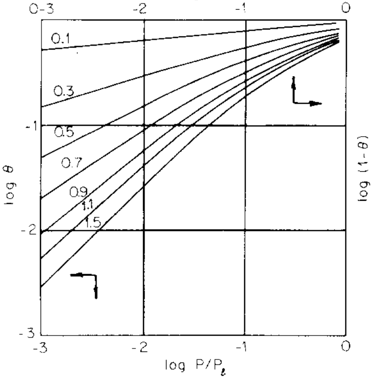

FIG. 1a. Adsorption isotherms for surface with exponential distribution of sites with respect to the heats of adsorption. Dependences of log θ on log (p/p_l) correspond to distribution function (1). Dependences of log $(1 - \theta)$ on log (p_u/p) correspond to distribution function (2). The values of μ are denoted by figures near the curves.

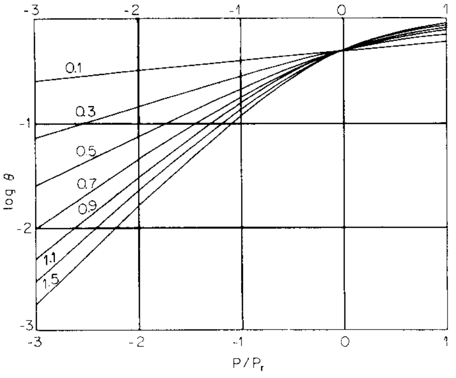

FIG. 1b. Adsorption isotherms for surface with symmetrical distribution function with a maximum of distribution of adsorption sites with respect to heats of adsorption. Dependences of log θ on log p/p_r. The values of μ are denoted by figures near the curves.

It can be seen that in the range $\theta > 0.1$ the isotherms in log coordinates are linear only at sufficiently low temperatures ($\mu \leqslant 0.1$ or $T \leqslant 0.1\ g_m/R$).

2. DIFFERENTIAL HEAT OF ADSORPTION–COVERAGE RELATIONSHIP FOR DISTRIBUTION FUNCTION (1)

Here, we give the dependence of g_d (differential heat of adsorption) on θ and p/p_l for the most important pressure range $p_u < p < p_l$ only.

A comparison of Eq. (13) ($g_u = \infty$) with the Clausius–Clapeyron equation shows that

$$\frac{g_d}{g_m} = \frac{g_l}{g_m} + \frac{\dfrac{\mu\pi}{\sin\mu\pi}\left[\dfrac{\cos\mu\pi\cdot\mu\pi}{\sin\mu\pi} - 1 - \ln\left(\dfrac{p}{p_l}\right)^{\mu}\right] + \left(\dfrac{p}{p_l}\right)^{1-\mu}\displaystyle\sum_{n=1}^{n=\infty}(-1)^{n+1}\left(\dfrac{p}{p_l}\right)^{n-1}\dfrac{n\mu}{(n-\mu)^2}}{\dfrac{\mu\pi}{\sin\mu\pi} + \left(\dfrac{p}{p_l}\right)^{1-\mu}\displaystyle\sum_{n=1}^{n=\infty}(-1)^{n+1}\left(\dfrac{p}{p_l}\right)^{1-\mu}\cdot\dfrac{n}{\mu-n}}. \qquad (15)$$

For $\mu \ll 1$ and $p/p_l \ll 1$ Eq. (15) reduce to Eq.

$$g_d = -g_m \ln\theta + g_l \qquad (16)$$

and for $\mu \gg 1$ and $p/p_l \ll 1$ to equation.

$$g_d = g_l + RT\ (1/(\mu - 1)). \qquad (17)$$

Figure 2a exhibits calculated $(g_d/g_m) - \log\theta$ relationships obtained from Eqs. (15) and (13). It can be seen that the shape and position of the curves depend on the temperature. The degree of temperature dependence is quite sufficient for comparing theoretical curves with calorimetric data.

3. ACCURATE EQUATION OF NEGATIVE-POWER ISOTHERM

M. I. Temkin (*8*) found that to the exponential distribution (2) there corresponds the negative-power isotherm

$$\theta = c - bp^{-\mu} \qquad (18)$$

(c and b = constants).

This equation is obviously an approximate one.

The accurate equations of the isotherm corresponding to the site distribution function (2) were found (*9*) by the method of analytical continuation of the function. These equations, however, can also be obtained from equations corresponding to the distribution function (1). To do this, it is sufficient to replace θ by $(I - \theta)$, p by p^{-1}, p_u by $(p_l)^{-1}$ and p_l by $(p_u)^{-1}$. Then the isotherm equation corresponding to the high-coverage range in the case of the distribution function (1) will correspond to the low-coverage range in the case of the distribution function (2), and vice versa.

For the low-coverage range ($p < p_u$) we get

$$\theta = I - \left[\frac{F(1,\mu;\ \mu + I;\ -p/p_u)}{I - L^{-1}} - \frac{F(1,\mu;\ \mu + 1;\ -p/p_l)}{L - 1}\right]. \qquad (19)$$

For most important middle pressure range $p_u < p < p_l$ we get:

$$\theta = 1 + \frac{(\mu/\mu - 1))(p_u/p)F(1,1-\mu;\ 2-\mu;\ -(p_u/p)) - (\mu\pi/\sin\mu\pi)(p_u/p)^{\mu}}{1 - L^{-1}} + \frac{F(1,\mu;\ 1+\mu;\ -(p/p_l)}{L - 1}. \qquad (20)$$

Here it is possible to obtain approximate equations similar to Eqs. (13) and (14). The equation (21) corresponding to the condition $g_l = -\infty$ and the equation (21a) corresponding to the condition $g_l = -\infty$ and $p \gg p_u$ may be considered the most important of them:

$$\theta = 1 + \frac{\mu}{\mu - 1} (p_u/p) F\left(1{,}1 - \mu; 2 - \mu; -\frac{p_u}{p}\right) - \frac{\mu\pi}{\sin \mu\pi} \left(\frac{p_u}{p}\right)^{\mu}. \quad (21)$$

(Equation (21) can be represented graphically with the aid of the curves of Fig. 1a. To do this, lay off log $(1 - \theta)$ on the y-axis and log (p_u/p) on the x-axis.)

$$\theta = 1 + \frac{\mu}{1 - \mu} \left(\frac{p_u}{p}\right) - \frac{\mu\pi}{\sin \mu\pi} \left(\frac{p_u}{p}\right)^{\mu}. \quad (21a)$$

For $\mu \ll 1$ and $p \gg p_u$ Eq. (21a) obviously reduce to a negative-power equation (18). For high coverage $(p > p_l)$ we have

$$\theta = 1 + \frac{\mu}{\mu - 1} \frac{(p_l/p) F(1{,}1 - \mu; 2 - \mu; -p_l/p)}{L - 1} - \frac{(p_u/p) F(1{,}1 - \mu; 2 - \mu; -p_u/p)}{I - L^{-1}}. \quad (22)$$

The calculated dependence of g_d on θ and p_u/p for the case of site distribution function (2) can be represented by the curves of Fig. 2a (for the middle pressure range and infinitely large lower limit $g_l = -\infty$).

In this case the value $(g_u - g_d)/g_m$ should be layed off on the y-axis and log $(1 - \theta)$ on the x-axis.

4. THE ISOTHERMS CORRESPONDING TO DISTRIBUTION FUNCTIONS WITH MAXIMUM

A symmetric or nonsymmetric site distribution function of cites with a maximum can be obtained by combining Eqs. (1) and (2).

(a) *The nonsymmetric distribution function.* For $g > g_r$ (g_r is the heat of adsorption corresponding to the maximum of site distribution curve).

$$N_g = \exp\ (-g/g_m); \ \mu = RT/g_m. \quad (23)$$

For $g < g_r$

$$N_g = \exp\ (g/g'_m); \ \mu' = RT/g'_m$$

(b) *The symmetric function* $(g_m = g'_m)$.

$$N_g = \exp\ [-|g - g_r|/g_m] \quad (24)$$

The curves of the distribution functions (23) and (24) have sharp peaks at $g = g_r$ in contrast to the normal distribution curve. The distribution functions (23) and (24), however, enable one to evaluate accurate integration and obtain isotherm equations which make it possible to vary the positions of the distribution maximum and limits over the heats of adsorption.

Furthermore in this case it is possible to vary pressure and temperature from zero to infinity (of course, it is a mathematical possibility) and to find the corresponding values of the coverage and the differential heat of adsorption.

The initial equation of isotherm, corresponding to the site distribution function (23) can be written as

$$\theta = A(1 + z^{-1})^{-1} + B(1 + z)^{-1}, \quad (25)$$

where A is Eq. (5); B is the initial equation of isotherm, corresponding to the site distribution function (2) and

$$z = \frac{g_m \exp\ ((g_r - g_l)/g_m)}{g'_m \exp\ ((g_u - g_r)/g'_m)}.$$

In the case of symmetry $(\mu = \mu_1$ and $g_u - g_r = g_r - g_l)$ $z = 1$. Then we find the solution as:

if $p < p_u$

$$\theta = A_1(1 + z^{-1})^{-1} + B_1(1 + z)^{-1}; \quad (26)$$

if $p < p < p_r$

$$\theta = A_2(1 + z^{-1}) + B_1(1 + z)^{-1}; \quad (26a)$$

if $p_r < p < p_l$

$$\theta = A_3(1+z^{-1})^{-1} + B_2(1+z)^{-1}; \quad (26b)$$

if $p > p_l$

$$\theta = A_3(1+z^{-1})^{-1} + B_3(1+z)^{-1}, \quad (26c)$$

where A_1, A_2, A_3 are the isotherm equations corresponding to the distribution function (1), B_1, B_2, B_3 are the isotherm equations corresponding to the distribution function (2).

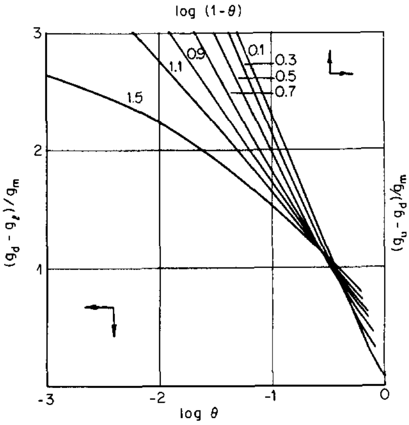

FIG. 2a. Heats of adsorption–coverage relationships for surface with exponential distribution of sites with respect to heats of adsorption. Dependence of $(g_d - g_l)/g_m$ on log θ correspond to distribution function (1). Dependences of $(g_u - g_d)/g_m$ on log $(1-\theta)$ correspond to distribution function (2). The values of μ are denoted by figures near the curves.

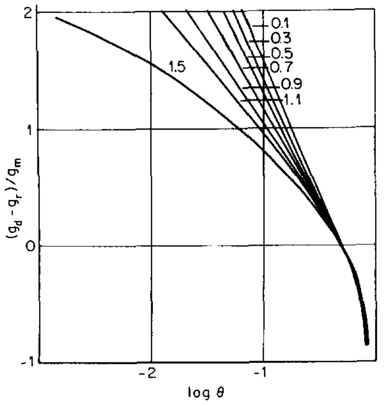

FIG. 2b. Heats of adsorption–coverage relationships in the case of symmetrical distribution function with a maximum. Dependence of $(g_d - g_r)/g_m$ on log θ. The values of μ are denoted by figures near the curves.

A_1 denotes the isotherm equation in the range $p < p_u$, A_2 and B_1, the isotherm equations in the range of $p < p_r$, A_3 and B_2, the isotherm equations in the range of $p_r < p < p_l$, and B_3, the isotherm equation in the range $p > p_l$.

Figure 1b shows isotherms for the most important pressure range in the case of symmetric distribution with a maximum and of infinitely large limits ($g_u = +\infty$, $g_l = -\infty$). The curves were calculated from Eq. (26a) and (26b), where A_2 is the isotherm described by Eq. (13), A_3 is the isotherm described by Eq. (8) for the case of $L = \infty$, B_1 is the isotherm described by Eq. (20) for the case of $L = \infty$, B_2 is the isotherm described by Eq. (21a). A comparison of the shapes of the isotherms in Fig. 1b and those in Fig. 1a does not reveal any evident difference between them. The differences in the nature of the heat of adsorption–coverage relationship are more visible.

Comparing Eq. (26a) and Eq. (26b) with the Clausius-Clapeyron equation we can find the $g_d - p$ relation (by analogy with Eq. (15)) and calculate the $g_d/g_m - \log\theta$ dependence. This dependence for the case infinitely large limits ($g_u = +\infty$ and $g_l = -\infty$) is given in Fig. 2b.

The difference between the curves in Fig. 2a and 2b in the range of relatively high coverages is obvious.

5. LIMIT TRANSITIONS OF ISOTHERM AS $g_m \to \infty$ AND $T \to 0$

Expanding the hypergeometric functions of the above isotherm equations into infinite series one can consider in detail a transition of them into Temkin's quasi-logarithmic form *(12)* $\theta = RT/(g_u - g_l)$ $[\ln(1 + p/p_u) - \ln(I + p/p_l)]$ as $g_m \to \infty$ and into equation

$$\theta = \mu \left[\frac{\ln (1 + p/p_u)}{L - 1} - \frac{\ln (1 + p/p_l)}{1 - L^{-1}} \right] \tag{27}$$

as $T \to 0$.

It is possible these transitions are a cause of prevalence of isotherm logarithmic forms.

CONCLUSION

The results obtained confirm that it is far from easy to establish the nature of surface heterogeneity from adsorption data. Simultaneous measurements of adsorption isotherms and determination of heats of adsorption by the calorimetric method in temperature ranges corresponding not only to $\mu < 1$ but also to $\mu > 1$ would be of great interest.

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