

Gas Adsorption on Solids with *Gaussian* Micropore-Size Distributions

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This paper presents a theoretical description of gas adsorption on microporous solids with *Gaussian* micropore-size distributions. This description comprises equations for the overall adsorption isotherm, the adsorption potential distribution, the differential molar enthalpy, and the differential molar entropy of adsorption. The truncated *Gaussian* micropore-size distribution is assumed to represent the structural heterogeneity of the micropores, whereas the adsorption potential distribution is used to characterize the energetic heterogeneity of these micropores.

(Keywords: Adsorption; Heterogeneous adsorbents, Micropore-size distributions)

Gasadsorption an Feststoffen mit Gaußscher Verteilung der Mikroporenrößen

Es wird eine theoretische Beschreibung der Gasadsorption an mikroporösen Feststoffen mit *Gaußscher* Verteilung der Porengrößen präsentiert. Diese Beschreibung beinhaltet Gleichungen für die Gesamtadsorptions-Isotherme, die Adsorptionspotential-Verteilung, die differentielle molare Enthalpie und die differentielle molare Entropie der Adsorption. Es wird angenommen, daß eine vereinfachte *Gauß*-Verteilung der Mikroporen die strukturelle Heterogenität wiedergibt, währenddessen eine Adsorptionspotential-Verteilung zur Darstellung der energetischen Heterogenität der Mikroporen verwendet wird.

Introduction

In the majority of papers [1] dealing with gas adsorption on heterogeneous solids, the theoretical considerations are based on the concept of the energetic heterogeneity of these solids. According to this

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concept, an energetically-heterogeneous solid possesses adsorption sites of different adsorption energies [2]. The sources of the energetic heterogeneity of a solid are various atoms and functional groups exposed at the solid surface, impurities bonded strongly to the surface, imperfections of the surface (surface heterogeneity), and a non-uniform microporous structure of the solid (structural heterogeneity) [1 e]. The differential distribution of the adsorption energy is usually used to characterize the energetic heterogeneity of the solids. For porous solids with large pores [3] and for non-porous solids, this energy distribution reflects only the surface heterogeneity because large pores do not cause an increase in the adsorption energy. For a microporous solid, this energy distribution reflects also the structural heterogeneity of the solid. For microporous solids, this discussion indicates that the physical interpretation of the energy distribution is more difficult because this function contains information about the surface heterogeneity of a solid and its microporous structure [4]. Recently, a method was proposed [4, 5] to separate the contributions to the total energy distribution from the surface and structural heterogeneities of a solid. Besides the energy distribution, which provides information about microporosity, the structural heterogeneity of microporous solids may be characterized by the micropore-size distribution [6], which provides direct information about the microporous structure of a solid.

Theoretical and experimental studies [6] showed that gas adsorption on a uniform microporous solid (e.g., activated carbon) is described satisfactorily by the *Dubinin* equation:

$$\theta = \exp(-mx^2A^2) \quad (1)$$

where

$$A = -\Delta G = RT \ln(p_0/p) \quad (2)$$

Here θ is the volume fraction of the uniform slit-like micropores of half-width x filled with adsorbate at the equilibrium pressure p and the absolute temperature T , A is the adsorption potential defined through Eq. (2) as the change of the *Gibbs* free energy taken with the minus sign, m is the coefficient established experimentally to be constant for a given vapor [6], p_0 is the saturation vapor pressure, and R is the universal gas constant. *Dubinin* [6] utilized Eq. (1) to formulate the isotherm equation for a non-uniform (heterogeneous) microporous solid with a *Gaussian* micropore-size distribution. It is worth noting that some authors [7] used another distribution function for characterizing the structural heterogeneity of microporous solids. Although this distribution does not provide direct

information about the heterogeneity of microporous solids, it may be recalculated to the micropore-size distribution.

The aim of this paper is to derive equations for the overall adsorption isotherm, the adsorption potential distribution, and the differential entropy for adsorption on microporous solids characterized by the *Gaussian* micropore-size distribution. *Dubinin's* approach [6] is modified to satisfy physical requirements. Since the physical meaning of the micropore-size distribution requires that each integration with respect to x be performed from zero, the part of *Gaussian* distribution from minus infinity to zero is omitted; consequently, the "truncated" *Gaussian* micropore-size distribution requires renormalization. The mean value \bar{x} and the dispersion σ_x for this truncated distribution are different from those that characterize the *Gaussian* distribution.

Results and Discussion

Properties of the Truncated Gaussian Micropore-Size Distribution

The *Gaussian* micropore-size distribution $J(x)$ may be written as follows:

$$J(x) = N \exp \left[-\frac{(x_0 - x)^2}{2\delta^2} \right] \quad (3)$$

where N is the normalization coefficient, and x_0 and δ are parameters of this distribution. The function $J(x)$ given by Eq. (3) is defined usually in the region of x from minus infinity to plus infinity; then the normalization coefficient $N = 1/(2\pi)^{1/2} \delta$, x_0 is equal to the mean value \bar{x} , and δ is the dispersion σ_x . Physical reality requires that the micropore-size distribution $J(x)$ be defined in the region of x from zero to plus infinity. Also this physically-meaningful distribution should satisfy the following normalization condition:

$$\int_0^{\infty} J(x) dx = 1 \quad (4)$$

To represent the micropore-size distribution, we assume here that a truncated-*Gaussian* distribution given by Eq. (3) for $x \geq 0$ satisfies the normalization condition [Eq. (4)]. The normalization coefficient N evaluated according to Eqs. (3) and (4) is:

$$N = \frac{1}{(2\pi)^{1/2} \delta} \cdot \frac{2}{1 + \operatorname{erf} \left(\frac{x_0}{\sqrt{2} \delta} \right)} = \frac{1}{(2\pi)^{1/2} \delta} \cdot \frac{2}{\operatorname{erfc} \left(-\frac{x_0}{\sqrt{2} \delta} \right)} \quad (5)$$

where $\text{erf}(z)$ is the known error function defined as follows [8]:

$$\text{erf}(z) = \frac{2}{\sqrt{\pi}} \int_0^z \exp(-t^2) dt \quad (6)$$

The complementary error function $\text{erfc}(-z)$ is given by:

$$\text{erfc}(-z) = \frac{2}{\sqrt{\pi}} \int_{-z}^{\infty} \exp(-t^2) dt = 1 + \text{erf}(z) \quad (7)$$

In view of Eq. (5), the truncated *Gaussian* micropore-size distribution is:

$$J(x) = \frac{1}{(2\pi)^{1/2} \delta} \cdot \frac{2}{\text{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right)} \exp\left[-\frac{(x_0 - x)^2}{2\delta^2}\right] \quad \text{for } x \geq 0 \quad (8)$$

The mean value \bar{x} and the dispersion σ_x of the truncated-*Gaussian* distribution [Eq. (8)] are different than those for the *Gaussian* distribution [Eq. (3) with $-\infty \leq x \leq \infty$]. The values of \bar{x} and σ_x associated with the distribution $J(x)$ may be calculated by means of the following equations:

$$\bar{x} = \int_0^{\infty} x J(x) dx \quad (9)$$

$$\sigma_x = \left[\int_0^{\infty} (x - \bar{x})^2 J(x) dx \right]^{1/2} = \left[\int_0^{\infty} x^2 J(x) dx - \bar{x}^2 \right]^{1/2} \quad (10)$$

Calculation of the integral expressed by Eq. (9) for the distribution $J(x)$ [Eq. (8)] gives:

$$\bar{x} = x_0 + \left(\frac{2}{\pi}\right)^{1/2} \frac{\delta}{\text{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right)} \exp\left(-\frac{x_0^2}{2\delta^2}\right) \quad (11)$$

The second term in Eq. (11) represents a correction to the *Gaussian* mean value x_0 . The dispersion σ_x for the truncated-*Gaussian* distribution $J(x)$ [Eq. (8)] calculated according to Eq. (10) is:

$$\sigma_x = \left[\delta^2 - \left(\frac{2}{\pi}\right)^{1/2} \frac{\delta x_0 \exp\left(-\frac{x_0^2}{2\delta^2}\right)}{\text{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right)} - \frac{2\delta^2 \exp(-x_0^2/\delta^2)}{\pi \left[\text{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right) \right]^2} \right]^{1/2} \quad (12)$$

The second and third terms in Eq. (12) define a correction to the variance δ^2 associated with the *Gaussian* distribution given by Eq.(3); this correction is the result of neglecting the part of *Gaussian* distribution in the region of x from minus infinity to zero. For small values of δ [i.e., for a narrow micropore-size distribution $J(x)$] and for relatively large values of x_0 (such that the maximum of this distribution is located relatively far from the origin), the correction terms appearing in Eqs. (11) and (12) become negligible; however, for a broad micropore-size distribution $J(x)$ (which corresponds to strong heterogeneity of the microporous structure), these terms give significant contributions to the mean values \bar{x} and the dispersion σ_x of this distribution.

Adsorption Equation for Solids Characterized by the Truncated Gaussian Micropore-Size Distribution

For a heterogeneous microporous solid, the fraction of filling of the non-uniform slit-like micropores is expressed by [6]:

$$\theta(A) = \int_0^{\infty} \exp(-mx^2A^2)J(x)dx \quad (13)$$

The function $\theta(A)$ is called the characteristic adsorption curve; however, the dependence θ vs. p denotes the adsorption isotherm. *Dubinin* [6] discussed an adsorption equation obtained by solving the integral Eq. (13) for the *Gaussian* distribution given by Eq.(3) with $N = 1/(2\pi)^{1/2}\delta$. As mentioned earlier, this distribution does not satisfy the normalization condition [Eq.(4)], which is necessary for a physically-meaningful micropore-size distribution.

Evaluation of the integral Eq. (13) for the truncated micropore-size distribution gives:

$$\theta(A) = v(A) \exp(-mx_0^2A^2[v(A)]^2) \frac{\operatorname{erfc}\left[-\frac{x_0v(A)}{\sqrt{2}\delta}\right]}{\operatorname{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right)} \quad (14)$$

where

$$v(A) = (1 + 2m\delta^2A^2)^{-1/2} \quad (15)$$

and

$$\operatorname{erfc}\left[-\frac{x_0v(A)}{\sqrt{2}\delta}\right] = 1 + \operatorname{erf}\left[-\frac{x_0v(A)}{\sqrt{2}\delta}\right] \quad (16)$$

For $\delta = 0$ (i.e., a uniform microporous solid), the function $v(A) = 1$ and the ratio of the complementary error functions in Eq. (14) is equal to unity; in this case, Eq. (14) reduces to the *Dubinin* Eq. (1).

Adsorption Potential Distribution Associated with the Characteristic Adsorption Eq. (14)

Microporous structure is the source of an additional energetic heterogeneity, which may be characterized by the adsorption potential distribution $X(A)$ [1 e]. This distribution satisfies the following normalization condition:

$$\int_0^{\infty} X(A) dA = 1 \quad (17)$$

A simple way of calculating the distribution $X(A)$ is offered by the condensation-approximation method [1 g]; accordingly, the $X(A)$ -distribution is given by:

$$X(A) = -d\theta(A)/dA \quad (18)$$

Using Eq. (18) to calculate the adsorption potential distribution $X(A)$ associated with the characteristic adsorption curve given by Eq. (14), we obtain:

$$\begin{aligned} X(A) = & \frac{2mA[v(A)]^3}{\operatorname{erfc}\left(-\frac{x_0}{\sqrt{2}\delta}\right)} \exp\{-mx_0^2A^2[v(A)]^2\} \\ & \cdot \left[\operatorname{erfc}\left[-\frac{x_0v(A)}{\sqrt{2}\delta}\right] \cdot (\delta^2 + x_0^2[v(A)]^2) \right. \\ & \left. + \left(\frac{2}{\pi}\right)^{1/2} \delta x_0 v(A) \exp\left(-\frac{x_0^2[v(A)]^2}{2\delta^2}\right) \right] \quad (19) \end{aligned}$$

The average adsorption potential \bar{A} associated with the $X(A)$ -distribution is defined as follows:

$$\bar{A} = \int_0^{\infty} AX(A) dA \quad (20)$$

Another equation for \bar{A} may be obtained either by calculating the integral Eq. (20) for the $X(A)$ -distribution given by Eq. (19), or in a simpler way from the following integral representation of $X(A)$:

$$X(A) = \int_0^{\infty} 2mx^2A \exp(-mx^2A^2)J(x) dx \quad (21)$$

Equation (21) was obtained by differentiating Eq. (13) according to Eq. (18). Equations (20) and (21) give a general expression for the average adsorption potential \bar{A} [9]:

$$\bar{A} = \frac{1}{2} \left(\frac{\pi}{m} \right)^{1/2} \int_0^{\infty} \frac{J(x)}{x} dx \quad (22)$$

Substitution of the micropore-size distribution $J(x)$ [Eq. (8)] into Eq. (22) gives:

$$\bar{A} = \left[\delta (2m)^{1/2} \operatorname{erfc} \left(-\frac{x_0}{\sqrt{2}\delta} \right) \right]^{-1} \int_0^{\infty} \frac{1}{x} \exp \left[-\frac{(x_0 - x)^2}{2\delta^2} \right] dx \quad (23)$$

The dispersion σ_A associated with the $X(A)$ -distribution given by Eq. (21) is:

$$\begin{aligned} \sigma_A &= \left[\int_0^{\infty} (A - \bar{A})^2 X(A) dA \right]^{1/2} = \left[\int_0^{\infty} A^2 X(A) dA - \bar{A}^2 \right]^{1/2} \\ &= \frac{1}{\sqrt{m}} \left(\int_0^{\infty} \frac{J(x)}{x^2} dx - \frac{\pi}{4} \left[\int_0^{\infty} \frac{J(x)}{x} dx \right]^2 \right)^{1/2} \end{aligned} \quad (24)$$

For the $J(x)$ -distribution given by Eq. (8), we obtain:

$$\begin{aligned} \sigma_A^2 &= \frac{2}{m\delta(2\pi)^{1/2} \operatorname{erfc} \left(-\frac{x_0}{\sqrt{2}\delta} \right)} \int_0^{\infty} \frac{1}{x^2} \exp \left[-\frac{(x_0 - x)^2}{2\delta^2} \right] dx \\ &\quad - \frac{1}{2m\delta^2 \left[\operatorname{erfc} \left(-\frac{x_0}{\sqrt{2}\delta} \right) \right]^2} \left[\int_0^{\infty} \frac{1}{x} \exp \left[-\frac{(x_0 - x)^2}{2\delta^2} \right] dx \right]^2 \end{aligned} \quad (25)$$

The mean adsorption potential \bar{A} and the dispersion σ_A characterize the adsorption potential distribution $X(A)$ given by Eq. (19).

Thermodynamic Functions Associated with the Characteristic Adsorption Eq. (14)

The differential molar enthalpy ΔH and the differential molar entropy ΔS for a gaseous compound adsorbed on a microporous solid may be expressed in terms of the characteristic adsorption curve $\theta(A)$ and the adsorption potential distribution $X(A)$ defined by Eq. (18) [10]:

$$\Delta H = -A - \alpha T \theta(A)/X(A) \quad (26)$$

$$\Delta S = -\alpha \theta(A)/X(A) \quad (27)$$

Here α is the negative of the thermal coefficient of the logarithm of the maximum amount a° adsorbed in the micropores; that is,

$$\alpha = - (d \ln a^\circ / dT) \quad (28)$$

Substituting Eqs. (14) and (19) into Eq. (26), we obtain:

$$\begin{aligned} \Delta H = -A - \frac{\alpha T \operatorname{erfc} \left[-\frac{x_0 v(A)}{\sqrt{2} \delta} \right]}{2mA [v(A)]^2} & \left[\operatorname{erfc} \left[-\frac{x_0 v(A)}{\sqrt{2} \delta} \right] \cdot (\delta^2 + x_0^2 [v(A)]^2) \right. \\ & \left. + \left(\frac{2}{\pi} \right)^{1/2} \delta x_0 v(A) \exp \left(-\frac{x_0^2 [v(A)]^2}{2\delta^2} \right) \right]^{-1} \end{aligned} \quad (29)$$

The differential molar entropy ΔS of adsorption associated with the functions $\theta(A)$ [Eq.(14)] and $X(A)$ [Eq.(19)] may be expressed as follows:

$$\begin{aligned} \Delta S = - \frac{\alpha \operatorname{erfc} \left[-\frac{x_0 v(A)}{\sqrt{2} \delta} \right]}{2mA [v(A)]^2} & \cdot \left[\operatorname{erfc} \left[-\frac{x_0 v(A)}{\sqrt{2} \delta} \right] \right. \\ & \cdot (\delta^2 + x_0^2 [v(A)]^2) + \left(\frac{2}{\pi} \right)^{1/2} \delta x_0 v(A) \cdot \exp \left[-\frac{x_0^2 [v(A)]^2}{2\delta^2} \right] \left. \right]^{-1} \end{aligned} \quad (30)$$

Also, we can express the immersion enthalpy ΔH^{im} of a microporous solid in a liquid adsorbate by means of the parameters that characterize the adsorption of its vapor. This enthalpy is related to the average adsorption potential \bar{A} [11]:

$$\Delta H^{\text{im}} = - (1 + \alpha T) \bar{A} \quad (31)$$

The average adsorption potential \bar{A} for the $X(A)$ -distribution given by Eq. (19) is expressed by Eq. (23); hence, substituting this equation into Eq. (31), we obtain:

$$\Delta H^{\text{im}} = - \left[(1 + \alpha T) \left/ \left[\delta (2m)^{1/2} \operatorname{erfc} \left(- \frac{x_0}{\sqrt{2}\delta} \right) \right] \right. \right] \cdot \int_0^{\infty} \frac{1}{x} \exp \left[- \frac{(x_0 - x)^2}{2\delta^2} \right] dx \quad (32)$$

The immersion enthalpy ΔH^{im} may be expressed also by means of the geometric surface area S^* related to the micropores. This surface area is defined as follows [12]:

$$S^* = V \int_0^{\infty} \frac{J(x)}{x} dx \quad (33)$$

where V is the micropore volume. For the truncated-*Gaussian* micropore-size distribution $J(x)$ [Eq. (8)], we have:

$$S^* = \frac{V}{(2\pi)^{1/2} \delta} \cdot \frac{2}{\operatorname{erfc} \left(- \frac{x_0}{\sqrt{2}\delta} \right)} \int_0^{\infty} \frac{1}{x} \exp \left[- \frac{(x_0 - x)^2}{2\delta^2} \right] dx \quad (34)$$

Combination of Eqs. (32) and (34) gives:

$$\Delta H^{\text{im}} = - \frac{1}{2} (1 + \alpha T) \left(\frac{\pi}{m} \right)^{1/2} (S^*/V) \quad (35)$$

It is noteworthy that Eq. (35) is valid for gas adsorption on a non-uniform microporous solid described by the integral Eq. (13) with an arbitrary micropore-size distribution $J(x)$. For this integral equation, the ratio of the average adsorption potential \bar{A} [Eq. (22)] and the surface area S^* [Eq. (33)] does not depend on the function $J(x)$:

$$\bar{A}/S^* = \frac{1}{2V} \left(\frac{\pi}{m} \right)^{1/2} \quad (36)$$

Since the methods for calculating the adsorption potential distribution $X(A)$ were discussed by many authors (e.g., see review [1 e] and references therein), the value \bar{A} may be calculated easily by numerical or analytical integration of Eq. (20). Next, the geometric surface area S^* of the micropores may be evaluated through Eq. (36); in other words, the

calculation of the S^* -value through Eq. (36) does not require the evaluation of the micropore-size distribution $J(x)$.

Illustrative Calculations

In this section, we present the results of illustrative calculations of the truncated-*Gaussian* micropore-size distribution $J(x)$ and the functions associated with this distribution. The micropore-size distributions in Fig. 1 were calculated from Eq. (8) with $x_0 = 0.8$ nm for two values of $\delta = 0.3$ and 0.5 nm. These values are analogous to those obtained from benzene adsorption isotherms measured on various types of activated carbons [7e]. The micropore size distribution $J(x)$ calculated for $\delta = 0.3$ nm is relatively narrow; in this case, the factor $2/\text{erfc}(-x_0/\sqrt{2}\delta)$ in Eq. (8) is equal to 0.99, and the normalization factor reduces to $1/(2\pi)^{1/2}\delta$. For a broad micropore-size distribution $J(x)$, the factor $2/\text{erfc}(-x_0/\sqrt{2}\delta)$ in Eq. (8) differs from unity and can not be neglected; that is, the use of a truncated *Gaussian* distribution with the simple normalization factor $1/(2\pi)^{1/2}\delta$ can not be justified as suggested elsewhere [6, 13]. For $x_0/\delta > 2.3$, it is easy to show that the above factor may be replaced by unity; however, for $x_0/\delta < 2.3$ its value differs from unity and the full form of Eq. (8) should be used to represent micropore-size distribution. For several heterogeneous microporous solids, it is noteworthy that the ratio x_0/δ is smaller than 2.3. For these solids, the normalized truncated-*Gaussian* distribution $J(x)$ given by Eq. (8) ensures

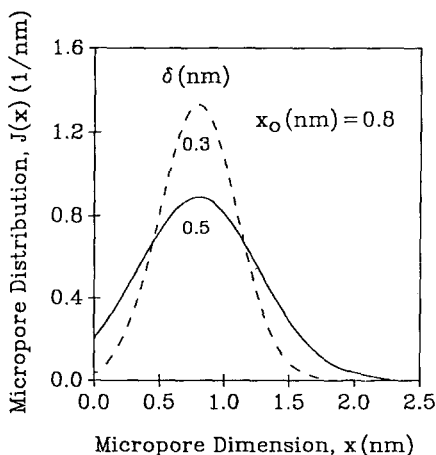


Fig. 1. The truncated-*Gaussian* micropore-size distribution $J(x)$ calculated according to Eq. (8) for $x_0 = 0.8$ nm, $\delta = 0.3$ nm (---) and 0.5 nm (—)

a correct evaluation for the micropore volume, the average micropore dimension \bar{x} , and the dispersion σ_x .

Figure 2 presents the characteristic adsorption curves $\theta(A)$ associated with the truncated-*Gaussian* micropore-size distributions shown in Fig. 1. These curves were calculated according to Eq. (14). The $\theta(A)$ -curves are decreasing functions from unity to zero. Comparison of the two $\theta(A)$ -curves, which are associated with narrow and broad micropore-size distributions, shows that the $\theta(A)$ -curve for the narrow distribution lies below that for the broad distribution when A is larger than about 20 kJ/mol. The two curves are almost the same for smaller values of A . Associated with the $\theta(A)$ -curves shown in Fig. 2 are the adsorption potential distributions $X(A)$ and the ratio of the differential molar enthalpy $\Delta S(A)$ to the thermal coefficient α ; the $X(A)$ -distribution curves are presented in Fig. 3, and the ratio $\Delta S(A)/\alpha$ is presented in Fig. 4. The $X(A)$ -distribution is a function with a single peak that is broadened in the direction of high values of A . This function does not change significantly as the structural heterogeneity of the micropores increases, which is represented by a broader micropore-size distribution. A characteristic feature of the $X(A)$ -distribution is a long tail, which increases with increasing heterogeneity of the micropores (cf. Fig. 3).

The dependence $\Delta S/\alpha$ vs. A shows a maximum, which becomes more pronounced for strongly-heterogeneous microporous solids. The maximum value of $\Delta S/\alpha$ increases as the micropore-size distribution becomes narrower.

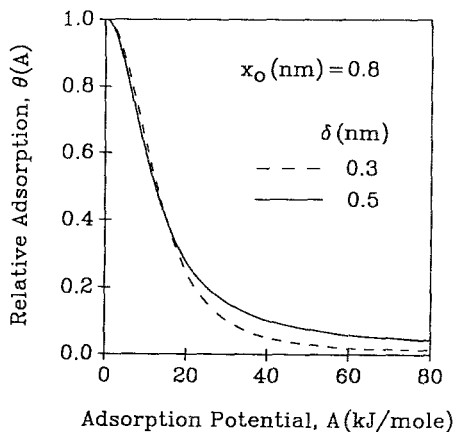


Fig. 2. The characteristic adsorption curves $\theta(A)$ associated with the truncated micropore-size distributions, calculated according to Eq. (14) for $x_0 = 0.8$ nm, $m = 0.00694$ (nm · kJ/mol) $^{-2}$, $\delta = 0.3$ nm (-----) and 0.5 nm (—————)

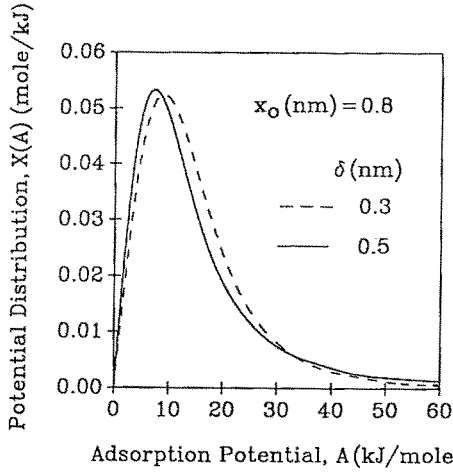


Fig. 3. The adsorption potential distribution $X(A)$ calculated according to Eq. (19) for $x_0 = 0.8$ nm, $m = 0.00694$ (nm · kJ/mol)⁻², $\delta = 0.3$ nm (-----) and 0.5 nm (————)

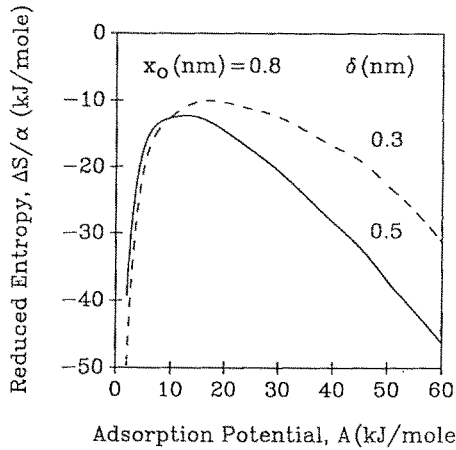


Fig. 4. The ratio $\Delta S/\alpha$ of the differential molar entropy ΔS to the thermal coefficient α plotted as function of the adsorption potential A , calculated according to Eq. (30) for $x_0 = 0.8$ nm, $m = 0.00694$ (nm · kJ/mol)⁻², $\delta = 0.3$ nm (-----) and 0.5 nm (————)

Conclusion

Our description of gas adsorption on heterogeneous microporous solids offers a set of expressions for the most important thermodynamic

quantities that are used for describing the volume filling of the micropores and for characterizing gas/solid adsorption systems. These expressions are generated by the truncated-*Gaussian* micropore-size distribution defined in the region from zero to plus infinity.

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