Adsorption of gases on adsorbents of various nature and porous structure at high pressures

A. A. Pribylov,* O. G. Larionov, L. G. Shekhovtsova, I. A. Kalinnikova, and L. D. Belyakova

A. N. Frumkin Institute of Physical Chemistry and Electrochemistry, Russian Academy of Sciences,
31 Leninsky prosp., 119991 Moscow, Russian Federation.
Fax: +7 (495) 952 5308. E-mail: pribylov@phyche.ac.ru, pribylov_34@mail.ru

The isotherms of excess adsorption of argon, nitrogen, and carbon dioxide on adsorbents with various porous structures (active carbon AU-71, polymeric sorbent MN-200, and synthetic zeolite NaA) were measured on the precision volumetric-gravimetric device in the pressure range of 0.1—150 MPa and at temperatures 300—400 K. The results of determination of the adsorption volumes of the studied adsorbents were compared using two methods. The first method is based on the Dubinin-Radushkevich equation, whereas the second one involves a conditional division of the weight of the substance in an ampule into the adsorbed portion and the part existing in the gas phase. The behavior of the adsorbed substance is described by the equation of adsorption of complete content corresponding to the physics of the adsorption process. The relation of the parameters of the empirical Dubinin—Radushkevich equation to the energy characteristics of the system was established.

Key words: adsorption, carbon sorbents, polymeric sorbents, synthetic zeolites, isotherm of excess adsorption.

The purpose of the present work is to develop a method for determination of the structural characteristics of adsorbents of various nature and porosity. Experimental data on adsorption of vapor of various substances are usually used for this purpose. However, when vapor is adsorbed on adsorbents with a broad pore size distribution. condensation processes in large pores can result in errors in determination of the adsorption volume. Investigations of the structural characteristics of microporous adsorbents widely use Dubinin´s method assuming that the adsorbate condensed in pores has the density equal to that of the liquid. This assumption does not always correspond to real observations. The results of studies of adsorption of gases rather than vapors on adsorbents of various porosity made it possible to develop a new method for the determination of the adsorption volume of the adsorbent, which includes no assumptions on the state of adsorbed substance. This method uses the experimental data: the weight of the gas existing in the adsorber filled with the adsorbent and the density of the equilibrium gaseous phase under known P and T values. The necessary condition for applicability of this method is the choice of the analytical expression for adsorption of complete content, which would correspond to the physics of the adsorption process. In this report, we describe adsorption of argon, and nitrogen, and carbon dioxide on the adsorbents of various nature and porous structure: supermicroporous carbon AU-71, polystyrene supercross-linked adsorbent MN-200, and synthetic zeolite NaA. The studies were carried out in the temperature interval 300—400 K and in the pressure range 0.1—90 MPa, and the studies on zeolite were carried out under pressures below 160 MPa. The results of determination of the structural and energetic characteristics of the studied sorbents by our method and the method based on the Dubinin—Radushkevich equation were compared.

Experimental

All measurements of argon and nitrogen adsorption were carried out by the combined volumetric gravimetric method on the experimental high-pressure device developed by the authors and described in detail earlier.[1] Argon (99.99%), nitrogen, and carbon dioxide (99.99%) were chosen as adsorptives. When measuring the adsorption isotherms, the temperature of the autoclave filled with the adsorbent in some cases deviated from the temperature of the gauge gravimetric manometer used for the determination of the pressure in the apparatus. Accordingly, the argon density in the autoclave and the pressure in the apparatus were calculated by the multiparametric equation of state for argon $F(P,T,\rho)^2$, which makes it possible to determine these parameters with an inaccuracy not higher than 1%. Synthetic zeolite NaA was used as the adsorbent with a regular porous structure, and active carbon AU-71 obtained from brown coal and classified as a supermicroporous carbon served as the adsorbent with a nonregular porous structure. Supercross-linked polystyrene polymer MN-200 was also studied.

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 4, pp. 707—712, April, 2009.

The isotherms of excess adsorption of argon on zeolite NaA at $P < 150$ MPa and 334, 373 K and on AU-71 and MN-200 under pressures of 0.1—90 MPa and different temperatures are shown in Figs 1—3. The excess adsorption values Γ were calculated by the equation

$$\Gamma = (m - \rho_g V_{acc})/m_{ads}, \quad (1)$$

where m is the weight of the gas (argon) existing in the autoclave filled with the adsorbent and measured with a balance, ρ_g is the density of the density of the equilibrium gaseous phase at given P and T, V_{acc} is the accessible volume calculated as the volume of the empty autoclave minus the volume of the adsorbent placed in the autoclave, and m_{ads} is the weight of the adsorbent. The volume of the empty autoclave was precalibrated. The adsorbent volume was determined by the division of the weight of the adsorbent into its density. For the active carbon the density was 2 g cm^{-3}, for the polymer it was 1.1 g cm^{-3}, and in the case of zeolite NaA its crystallographic density[3] equal to 1.99 g cm^{-3} was used.

Results and Discussion

Routine methods applied for the determination of the porous structure parameters of the adsorbents with the nonregular structure, particularly, active carbons make use of experimental data on vapor adsorption. The methods of approaching evaluation of the structure parameters of microporous adsorbents are most clearly presented in the theory of volumetric filling of microporous (TVFM). This theory is based on the Dubinin—Radushkevich (DR) thermal equation of adsorption[4]

$$a = a_0(T)\exp(-\{(RT/E)\ln[P_s(T)/P]\}^2) \quad (2)$$

and the Dubinin—Nikolaev equation

$$a_0(T) = a_0{}^0\exp[-\alpha(T - T_0)]. \quad (3)$$

Here a is the amount adsorbed at the temperature T and current P values, $a_0(T)$ is the limiting amount adsorbed at the temperature T, $a_0{}^0$ is the limiting amount adsorbed for the given system at T_0, E is the characteristic energy, P_s is the saturation vapor pressure of the adsorptive at the temperature T, T_0 is the boiling point under normal conditions, α is the thermal coefficient of limiting adsorption, and P is the equilibrium pressure of the adsorptive. These equations are widely used for studying the adsorption of vapors on various adsorbents, when the excess adsorption values Γ only slightly differ from the values of adsorption of complete content a at the same P values.

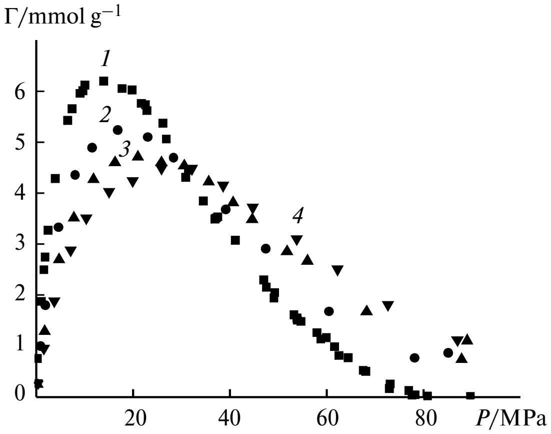

Fig. 2. Isotherms of excess adsorption of argon on active carbon AU-71 at 303 (*1*), 343 (*2*), 373 (*3*), and 400 K (*4*).

Determination of the specific adsorption volume. The method for the determination of the specific adsorption volume W by the results of measuring gas adsorption on adsorbents of various nature performed in a wide pressure interval and presented as excess isotherms has been described earlier.[5] The modification of the method[5] assumes that the initial experimental data on adsorption (P, m, ρ_g, T) are presented as the pressure dependence of m/ρ_g

$$m = m_a + m_g. \quad (4)$$

Here m is the weight of the gas existing in the autoclave of volume $V_a{}'$ filled with the adsorbent, m_a is the weight of

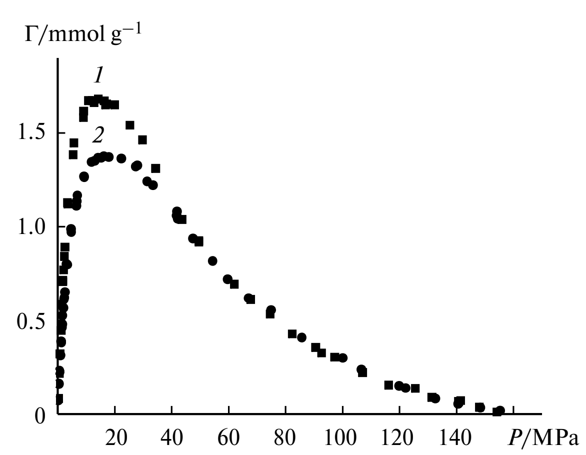

Fig. 1. Isotherms of excess adsorption of argon on zeolite NaA at 343 (*1*) and 373 K (*2*).

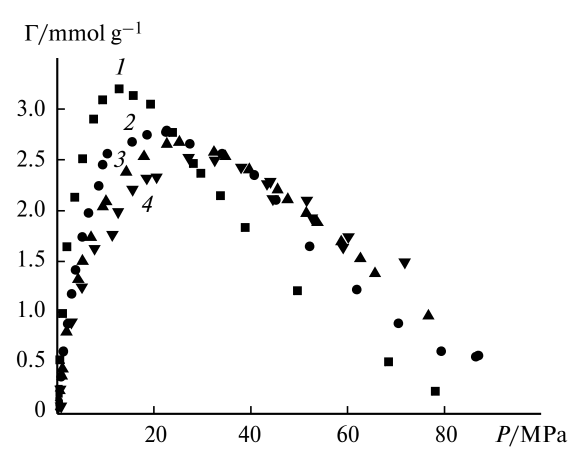

Fig. 3. Isotherms of excess adsorption of argon on polymer MN-200 at 303 (*1*), 343 (*2*), 373 (*3*), and 400 K (*4*).

the gas in the adsorbed state in the volume w (referred to the whole weight of the adsorbent) with the density ρ_a, and m_g is the weight of the gas existing in some volume V inside the autoclave with the density of the equilibrium gaseous phase ρ_g. Let us present dependence (4) as the equation

$$m/\rho_g = m_a/\rho_g + V = a/\rho_g + V. \qquad (5)$$

There is a problem of choosing such an analytical expression for a that would correspond to the physics of the adsorption process. The equation for the description of the isotherm of adsorption of complete content on porous adsorbents with the regular structure was earlier[6] derived

$$a = N/B = \frac{PQ_1 + 2P^2Q_2 + \ldots}{1 + PQ_1 + 2P^2Q_2 + \ldots} \qquad (6)$$

Here B is the number of cavities per 1 g of the adsorbent, in which N particles are adsorbed, Q_i is the configurational integral of the system consisting of i adsorbed particles in the cavity, and the adsorption of complete content N/B is expressed through the number of particles in a cavity. When carbon adsorbents with a nonregular structure are studied, the amount adsorbed is usually expressed in mmol g^{-1} and, hence, Eq. (6) should be used in the form

$$a = K_0\frac{PK_1 + 2P^2K_2 + \ldots + nP^nK_n}{1 + PK_1 + 2P^2K_2 + \ldots + P^nK_n}. \qquad (7)$$

Here the values K_0, K_1, ... K_n depend only on the temperature and are pressure-independent, and n is constant for the given adsorption system and is independent of either T or P. These coefficients can easily be selected under the conditions of best agreement between the experimental data and the curve obtained by Eq. (7) in the considered pressure intervals. The density of the equilibrium gaseous phase ρ_g can also be presented in form (7) with the intrinsic coefficients K_i^g. An n value of 3 can be adopted. An increase in n does not improve the description of experimental data. Now the function m/ρ_g (5) can be written in the form

$$m/\rho_g = wK_0{}^0A/B + V' - m_{ads}/\rho_{ads} - w, \qquad (8)$$

where

$$A = \frac{(K_1P + 2K_2P^2 + 3K_3P^3)}{(1 + K_1P + 2K_2P^2 + 3K_3P^3)},$$

$$B = \frac{K_0^g(K_1^gP + 2K_2^gP^2 + 3K_3^gP^3)}{(1 + K_1^gP + 2K_2^gP^2 + 3K_3^gP^3)}.$$

Here the coefficients K_i^g were determined beforehand for each isotherm, and K_i and w are selected by the Newton method. Expression (8) corresponds to the assumption that in the volume w (referred to the whole m_{ads}) the adsorbate density ρ_a is described by the equation of type (7) and the adsorptive with the density ρ_g is in the considered volume V inside the autoclave. In this case, the equality is fulfilled

$$V = V' - m_{ads}/\rho_{ads} - w. \qquad (9)$$

A modified method for the determination of the adsorption volume described above was first applied to the systems with active carbon AU-71 and with Ar and CO_2 as adsorptives. The w value was determined for all four temperatures (303, 343, 373, and 400 K) by the initial experimental data (P, m, ρ_s) on the adsorption of Ar on carbon AU-71. The model presented by Eq. (8) dictates that w is temperature-independent. The specific adsorption volume W was determined as

$$w/m_{ads} = W. \qquad (10)$$

The values of the specific adsorption volume for the argon—carbon AU-71 system evaluated by our method are given in Table 1.

The specific adsorption volume of the sample was determined by our method from the initial experimental data on CO_2 adsorption on carbon AU-71 for four temperatures (298, 343, 373, and 400 K) (Table 1). The obtained W values agrees satisfactorily with the values determined from vapor adsorption by the Dubinin—Radushkevich method using CO_2 (T = 298 K) (Fig. 4), C_6H_6 (T = 293 K), and N_2 (T = 77 K)[8] as adsorptives.

Adsorption of argon on the polymeric adsorbent MN-200 was studied similarly (see Figs 3 and 5). Four isotherms were considered: T = 303, 343, 373, and 400 K. The specific adsorption volume of the adsorbent equal to 0.45 cm^3 g^{-1} was found for this system. This value coincided satisfactorily with W = 0.5 cm^3 g^{-1} obtained by the calibration by helium and mercury with allowance for the assumption that the He atoms penetrate in all pores of adsorbent, but mercury does not diffuse in the pores. It should be mentioned that the adsorbent MN-200 exhibits good stability and reproducibility of the results obtained by multiple measurements of the adsorption and desorption of argon and nitrogen at high pressures (up to ~100 MPa) and temperatures below 450 K.

The application of this method to the adsorption system Ar—NaA and measurements at T = 343 and 373 K

Table. 1. Values of the specific adsorption volume obtained by our method (I) and using the Dubinin—Radushkevich method (II)

Adsorbent	Ar,	N_2		O_2,	CO_2		C_6H_6,
	I	I	II	II	I	II	II
AU-71	0.89	0.9	1.01	—	0.9	0.92	0.89
NaA	0.19	0.23	—	0.24*	—	—	—

* Earlier values.[7]

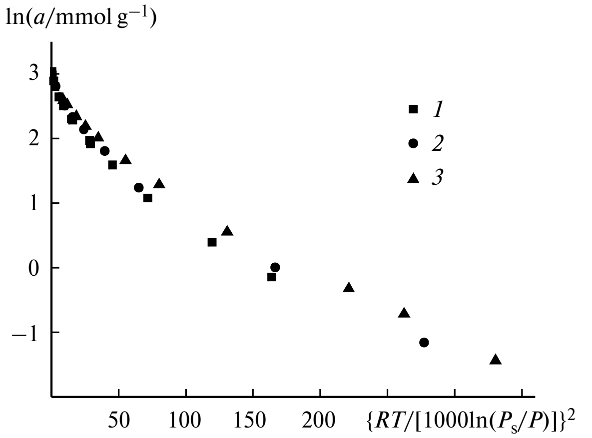

Fig. 4. Isotherms of excess adsorption of carbon dioxide on carbon AU-71 in the coordinates of the Dubinin—Radushkevich equation at 298 (*1*), 323 (*2*), and 347 K (*3*).

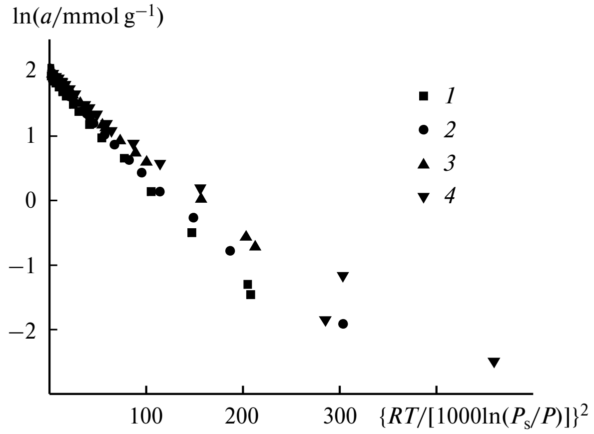

Fig. 5. Isotherms of excess adsorption of argon on MN-200 in the coordinates of the Dubinin—Radushkevich equation at 303 (*1*), 343 (*2*), 373 (*3*), and 400 K (*4*).

and pressures below 160 MPa (see Fig. 1) made it possible to determine W (see Table 1) that satisfactorily agrees with the earlier[9] reported value ($W = 0.185$ cm^3 g^{-1}). The processing of the data on N_2 adsorption on the same zeolite at temperatures 305, 334, and 373 K gave the value (see Table 1) well consistent with the published value ($W = 0.23$ cm^3 g^{-1}).[1] Using the adsorption volume values for all considered systems, one can calculate the isotherms of adsorption of complete content by the formula

$$a = [m - (V' - m_{\text{ads}}/\rho_{\text{ads}} - W)\rho_{\text{g}}]/m_{\text{ads}}. \qquad (11)$$

Relationship between the descriptions of adsorption by the statistical thermodynamics method and Dubinin—Radushkevich equation. For vapor adsorption the isotherms of excess adsorption and complete content almost coincide and increase monotonically in the entire pressure range. The isotherms of excess adsorption of gases in a wide pressure range, unlike the isotherms of vapor adsorption, pass through the maximum and then reach zero values with the pressure increase, whereas the shape of the isotherms of adsorption of complete content is similar to that of the isotherms of vapor adsorption.[1] It is known that the isotherms of vapor adsorption and the isotherms of adsorption of complete content of gases on the porous adsorbents are rather well described by both Eq. (2) and Eq. (6). Therefore, let us try to compare these equations describing the isotherms of adsorption of complete content. This allows one to obtain the relationship of the parameters in Eqs (2) and (6). However, to describe the isotherms of gas adsorption by Eq. (2), first of all it is necessary to determine an analog of P_s for the gaseous adsorptive, namely, «P_s». The tabulated data[10] for the dependence of lnP_s on $1/T$ for various gases are well approximated by the section of the plot of the linear function. Using this function, one can find the «P_s» values for the temperatures exceeding the critical point (Table 2).

An adsorption system on porous adsorbents with the regular structure has earlier[6] been described by the equations

$$N = \left.\frac{\partial(\ln \Xi)}{\partial(\ln P)}\right|_T, \quad U_{\text{av}} = -k\left.\frac{\partial(\ln \Xi)}{\partial(1/T)}\right|_P, \qquad (12)$$

$$\Xi = (1 + Q_1P + \ldots + Q_mP^m)^B,$$

where U_{av} is the average potential energy of N particles, k is the Boltzmann constant, and Ξ is the statistical sum of a large canonical ensemble for the adsorption system. Equation (6) was successfully applied[5,11,12] to the adsorbents with an arbitrary porous structure.

Let us consider the equations for the isotherms of adsorption (2) and (6) describing the same experimental data in the coordinates a—ln$P$. In Eq. (2) the amount adsorbed of complete content will be designed as a_1, and in Eq. (6) the same value is designed as a_2. To reveal the relation between the constants and average values for Eqs (2) and (6), we specify two conditions: a_1 and a_2 are equal at $P = $ «$P_s(T)$» and the integrals I_1 and I_2 coincide within the indicated range with good accuracy.

$$I_1(T) = \int\limits_{-\infty}^{P_s(T)} a_1(P,T)\text{d}(\ln P) = a_0(T)\frac{E}{RT}\frac{\sqrt{\pi}}{2} \qquad (13)$$

Table 2. Values «P_s» for the temperatures higher than the critical point

Gas	«P_s»/MPa (T/K)			
Ar	67.6 (303)	92.0 (343)	111.3 (373)	129.3 (400)
N_2	86.0 (303)	112.8 (343)	133.1 (373)	151.2 (400)
CO_2	6.3 (298)	10.5 (323)	16.1 (343)	—

$$I_2(T) = \int_{-\infty}^{P_s(T)} a_2(P,T)\mathrm{d}(\ln P) = \int_{-\infty}^{P_s(T)} N/B\mathrm{d}(\ln P) = \\ = \frac{1}{B}\ln \Xi(P_s(T),T) \tag{14}$$

Accepting that $I_1(T) \approx I_2(T)$, let us compare the full derivative with respect to $1/T$ for I_1 and I_2.

$$\frac{\mathrm{d}I_1(T)}{\mathrm{d}(1/T)} = \frac{\mathrm{d}}{\mathrm{d}(1/T)}(a_0(T)\frac{E}{RT}\frac{\sqrt{\pi}}{2}) = \\ = \frac{\sqrt{\pi}}{2}\frac{E}{R}a_0(T)(1 - T\frac{\mathrm{d}\ln a_0(T)}{\mathrm{d}T}) = \frac{\sqrt{\pi}}{2}\frac{E}{R}a_0(T)(1+\alpha T)$$

$$\frac{\mathrm{d}I_2(T)}{\mathrm{d}(1/T)} = \frac{1}{B}\left\{\frac{\partial \ln \Xi[P_s(T),T]}{\partial(\ln P_s)}\bigg|_T\right\}\frac{\mathrm{d}\ln P_s(T)}{\mathrm{d}(1/T)} + \\ + \frac{\partial \ln \Xi[P_s(T),T]}{\partial(1/T)}\bigg|_{\mathrm{av}} = \frac{1}{B}\left\{N_0\frac{\mathrm{d}\ln P_s(T)}{\mathrm{d}(1/T)} - \frac{1}{k}U_{\mathrm{av}}[P_s(T)]\right\} = \\ = \frac{N_0}{B}\left(A - \frac{u_{\mathrm{av}}}{k}\right) \tag{15}$$

Here U_{av} is the average potential energy of N_0 particles adsorbed under the pressure $P_s(T)$, and u_{av} is the average potential energy based on one particles under the same pressure,

$$\frac{\mathrm{d}\ln P_s(T)}{\mathrm{d}(1/T)} = A = \mathrm{const}$$

for all known cases. Equalizing the derivatives, we have

$$\frac{\sqrt{\pi}}{2}\frac{E}{R}a_0(T)(1+\alpha T) = \frac{N_0}{B}\left(A - \frac{u_{\mathrm{av}}}{k}\right). \tag{16}$$

According to the first condition, $a_0(T) = N_0/B$. Then Eq. (16) can be rewritten in the form

$$E = \frac{2}{\sqrt{\pi}}(AR - U_{\mu})/(1+\alpha T). \tag{17}$$

Let us calculate the system benzene—active carbon AG-40 (see Ref. 4) from Eq. (17). For this system $E = 20.7$ kJ mol^{-1}, $\alpha = 1.2 \cdot 10^{-3}$ K^{-1}, $A = -3.8 \cdot 10^3$ K^{-1}, $T = 280$ K, and the average isosteric heat of adsorption $q = 58.5$ kJ mol^{-1}. To determine U_{μ}, we use the equations

$$q_{\mathrm{st}} = -\frac{\partial U}{\partial N} + RT, \\ -U_{\mu} = -\frac{U}{N} = \frac{1}{a_m}\int_0^{a_0} q_{\mathrm{st}}\mathrm{d}a - RT = q - RT, \tag{18}$$

where U is the full potential energy of N moles of the adsorbed particles. In this case, the right side of Eq. (17) is equal to 1.13[−31.58 + (58.5 − 2.33)]/1.336 = 20.8 kJ mol^{-1}, which nearly coincides with $E = 20.7$ kJ mol^{-1}.

In some cases, Eq. (17) is substantially simplified. For the description of the isotherms of adsorption of complete content of gases by Eq. (2) with the parameters $a_0(T)$, E, and «P_s» in the pressure interval from zero to «P_s» for a certain range of the systems, $a_0(T) \approx$ const, *i.e.*, $\alpha \approx 0$. For instance, for the systems Ar and N$_2$—zeolite NaA (Figs 6 and 7) a_0 and E are almost temperature-independent, *i.e.*, $\alpha \approx 0$. Note that E is temperature-independent for the adsorption of vapors on microporous adsorbents. As can be seen from the data in Figs 4 and 8, for the systems Ar and CO$_2$—supermicroporous carbon AU-71, a_0 is also temperature-independent and the E value cannot already be considered constant. This fact indicating that the invariance of the characteristic curve is not valid has been considered earlier.[13] The thermodynamic criterion of applicability of the potential theory of adsorption was determined[13]: the change in the entropy ΔS of the adsorbate should be lower than zero. As an example, we calculated the change in the entropy of CO$_2$ adsorbed on carbon AU-71 (Fig. 9). For this system, invariance of the characteristic curve is fulfilled at $a > 7$ mmol g^{-1}, which is well

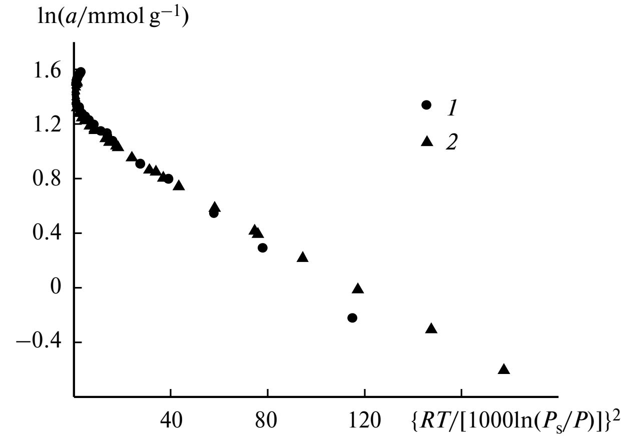

Fig. 6. Isotherms of excess adsorption of argon on zeolite NaA in the coordinates of the Dubinin—Radushkevich equation at 334 (*1*) and 373 K (*2*).

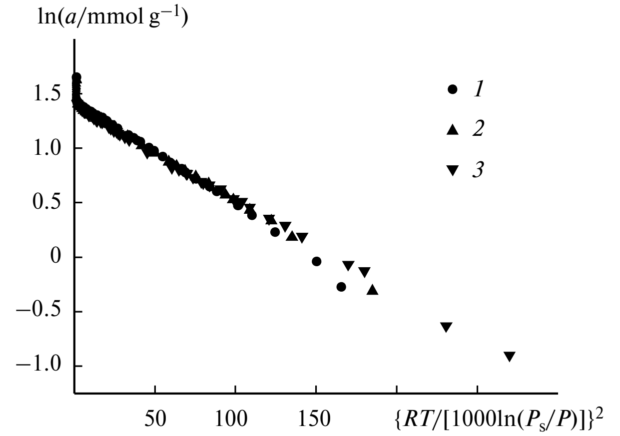

Fig. 7. Isotherms of excess adsorption of nitrogen on zeolite NaA in the coordinates of the Dubinin—Radushkevich equation at 305 (*1*), 334 (*2*), and 373 K (*3*).

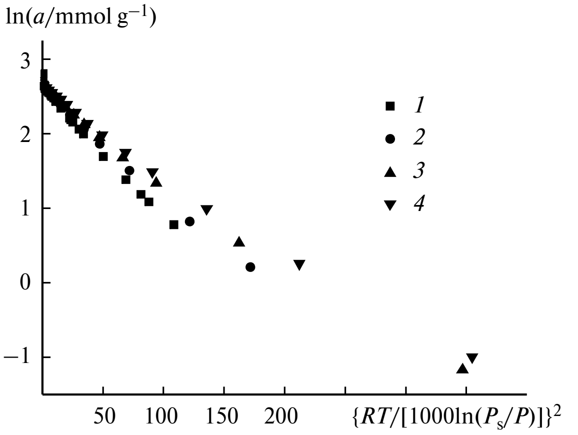

Fig. 8. Isotherms of excess adsorption of argon on carbon AU-71 in the coordinates of the Dubinin—Radushkevich equation at 303 (*1*), 343 (*2*), 373 (*3*), and 400 K (*4*).

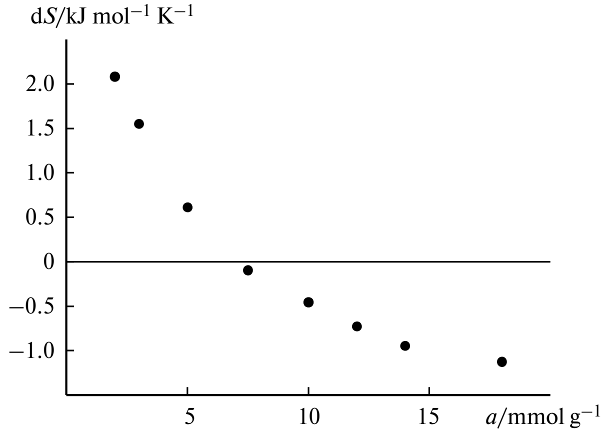

Fig. 9. Change in the entropy of adsorption of carbon dioxide on carbon AU-71 at different amounts adsorbed.

seen in Fig. 4. The systems Ar and N_2—polymeric adsorbent MN-200 are similar to the systems with AU-71 by the behavior of the isotherms (Figs 5 and 10).

Let us consider a specific feature of the experimental curves for the adsorption of gases (see Figs 6—8). The value of the argument $[RT\ln(«P_s»/P)]^2$ for the pressures higher than $«P_s»$ is equal to the argument for some $P < «P_s»$. Thus, at $P > «P_s»$ the function in the indicated coordinates becomes multivalued.

The right side of Eq. (17) at $\alpha = 0$ is defined only by the A and U_μ values. Then this equation makes it possible to determine the parameter E without considering the family of isotherms but by calculating the potential energy of adsorbate—adsorbent and adsorbate—adsorbate interactions for the system involved. Probably, this possibility is real for adsorbents of the zeolite type,[14] and after the transformation of Eq. (17)

$$E = \frac{2}{\sqrt{\pi}}(AR - U_\mu) \qquad (19)$$

becomes appropriate for the calculation of the E constant in Eq. (2).

Thus, the new method for the determination of the adsorption volume of porous adsorbents by measuring the adsorption of gases gives the values that differ slightly from those obtained by the Dubinin—Radushkevich equation for adsorption of vapors. However, the method proposed makes it possible to avoid the assumption used in the TVFM about the equality of the density of the adsorbed phase and the density of the liquid. This allows one to study systems inaccessible for investigations by the TVFM method.

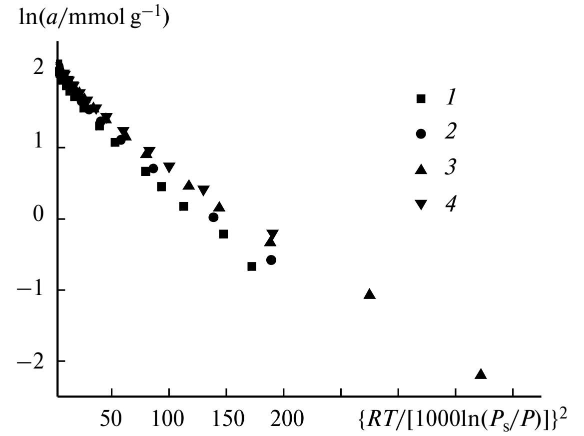

Fig. 10. Isotherms of adsorption of nitrogen on MN-200 in the coordinates of the Dubinin—Radushkevich equation at 303 (*1*), 343 (*2*), 373 (*3*), and 400 K (*4*).

The relationship between the parameters of the empirical Dubinin—Radushkevich equation to the energy characteristics of the system was established on the basis of approaching methods of statistical thermodynamics. This relationship serves to find the characteristic energy value (parameter E in the Dubinin—Radushkevich equation) without considering the family of isotherms but by calculating the average potential energy of adsorbate—adsorbent and adsorbate—adsorbate interactions by Eq. (18).

The authors are grateful to V. A. Davankov and M. P. Tsyurupa (A. N. Nesmeyanov Institute of Organoelement Chemistry, Russian Academy of Sciences) for the kindly presented samples of polymer MN-200.

This work was financially supported by the Division of Chemistry and Materials Science of the Russian Academy of Sciences (Grant 02.2.8).

References

1. A. A. Pribylov, S. M. Kalashnikov, V. V. Serpinskii, *Izv. Akad. Nauk SSSR*, *Ser. Khim.*, 1990, 1238 [*Bull. Acad. Sci. USSR*, *Div. Chem. Sci.* (*Engl. Transl.*), 1990, **39**, 1105].
2. *Teplofizicheskie svoistva neona, argona, kriptona i ksenona* [*Thermophysical Properties of Neon, Argon, Krypton, and Xe-*

non], Ed. V. A. Rabinovich, Izd-vo Standartov, Moscow, 1976, 636 pp. (in Russian).

3. D. W. Breck, *Zeolite Molecular Sieves*, Wiley-Inerscience, New York—London—Sidney—Toronto, 1974.
4. M. M. Dubinin, *Adsorbtsiya v mikroporakh. Tr. 5-i konf. po teoreticheskim voprosam adsorbtsii* [*Adsorption in Micropores. Abstrs 5th Conf. on Theoretical Problems of Adsorption*], Nauka, Moscow, 1983, 186 pp. (in Russian).
5. T. S. Yakubov, L. G. Shekhovtsova, A. A. Pribylov, *Izv. Akad. Nauk*, *Ser. Khim.*, 1995, 2381 [*Russ. Chem. Bull.* (*Engl. Transl.*), 1995, **44**, 2277].
6. V. A. Bakaev, *Dokl. Akad. Nauk SSSR*, 1966, **167**, 369 [*Dokl. Chem.* (*Engl. Transl.*), 1966].
7. A. M. Arkharov, B. P. Bering, I. A. Kalinnikova, V. V. Serpinskii, *Izv. Akad. Nauk SSSR*, *Ser. Khim.*, 1972, 1434 [*Bull. Acad. Sci. USSR*, *Div. Chem. Sci.* (*Engl. Transl.*), 1972, **21**, 1389.
8. A. A. Pribylov, L. G. Shekhovtsova, I. A. Kalinnikova, *Izv. Akad. Nauk*, *Ser. Khim.*, 2005, 2219 [*Russ. Chem. Bull.*, *Int. Ed.*, 2005, **54**, 2290].
9. A. A. Pribylov, *Izv. Akad. Nauk*, *Ser. Khim.*, 1996, 574 [*Russ. Chem. Bull.* (*Engl. Transl.*), 1996, **45**, 534].
10. N. B. Vargaftik, *Spravochnik po teplofizicheskim svoistvam gazov i zhidkostei* [*Manual on the Thermophysical Properties of Gases and Liquids*], Nauka, Moscow, 1972, 720 pp. (in Russian).
11. L. G. Shekhovtsova, A. A. Fomkin, *Izv. Akad. Nauk SSSR*, *Ser. Khim.*, 1992, 19 [*Bull. Russ. Acad. Sci.*, *Div. Chem. Sci.* (*Engl. Transl.*), 1992, **41**, 10].
12. L. G. Shekhovtsova, A. A. Fomkin, V. A. Bakaev, *Izv. Akad. Nauk SSSR, Ser. Khim.*, 1987, 2347 [*Bull. Acad. Sci. USSR*, *Div. Chem. Sci.* (*Engl. Transl.*), 1987, **36**, 2176].
13. B. P. Bering, V. V. Serpinskii, *Dokl. Akad. Nauk SSSR*, 1963, **148**, 1331 [*Dokl. Chem.* (*Engl. Transl.*), 1963].
14. V. P. Kalganov, N. A. Potolokov, V. A. Fedorov, A. M. Tolmachev, *Zh. Fiz. Khim.*, 2005, **79**, 1295 [*Russ. J. Phys. Chem.* (*Engl. Tansl.*), 2005, **79**].

Received August 29, 2007