Error in the Estimation of Effective Diffusion Coefficients from Sorption Measurements*

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Received 19 May 2000

A gravimetric sorption method was employed for the estimation of effective diffusion coefficients of water in silica gel particles. Air with a certain concentration of water passed through an adsorber with a fixed bed of silica gel particles with a height equal to the diameter of a single particle. The adsorber was weighed at chosen time intervals. This method is based on the assumption of zero mass transfer resistance in the fluid phase surrounding the adsorbent particles. From the two-film theory it follows that for the measurement conditions employed here, the maximum resistance in the fluid phase is 12.5 %. Hence, the resistance in the solid phase is loaded by a maximal error of 12.5 %. The total error of the estimated values of the effective diffusion coefficients is not greater than 40 %. The values of the effective diffusion coefficients obtained by the measurements of the stepwise adsorption and the values of the effective diffusivity obtained by the measurements of the stepwise desorption are within the range 0.90×10^{-10}—2.1×10^{-10} m^2 s^{-1}. Data obtained in this way are in agreement with data published in the literature. Comparison of the estimated values of the effective diffusion coefficients with those calculated under the assumption that the water transport in silica gel particles takes place solely by molecular diffusion or by Knudsen diffusion demonstrates that the latter does not occur under the conditions investigated.

In designing adsorbers mass transfer is usually described by unsteady diffusion, in which parameter is the effective diffusion coefficient. The values of effective diffusion coefficients can be obtained experimentally. The method used for the estimation of effective diffusion coefficients is described in more details in papers [1, 2]. Diffusion of gases in porous solids at higher temperatures up to 700 ℃ was reported by *Yang* and *Liu* [3]. Sorption and diffusion of pentane in particles of molecular sieve 5A was described in work [4]. Simultaneous diffusion and adsorption of hexane in particles of molecular sieve 5A is given in Ref. [5]. Adsorption kinetics of hydrocarbons in fixed active carbon or silica gel beds was reported by *Malek* and *Farooq* [6]. The dynamic-column breakthrough method has been effectively used to determine mass transfer data for methane, ethane, and propane adsorption in activated carbon and silica gel beds. The advantage of this method over other gravimetric or volumetric experimental methods is that more representative data are obtained since a relatively large amount of adsorbent is used. In studying sorption of ethylbenzene in crystals of molecular sieve H-ZSM-5, *Schumacher* and *Karge* [7] estimated the values of effective diffusion coefficients ranging from 1.5×10^{-11} to 2×10^{-10} m^2 s^{-1} at temperatures within 315—395 K. *Krückels* [8] experimentally investigated the step adsorption of water vapours on silica gel particles. Measurements were performed by means of Cahn electronic microweights in the absence of an inert gas. The calculated values of the effective diffusion coefficient obtained by the solution derived by Crank for low values of adsorption time and simplified by Schilling were ranging from 2×10^{-11} to 2×10^{-10} m^2 s^{-1}. *Pesaran* and *Mills* [9, 10] examined the transport mechanism of humidity in silica gel particles. From measurements of the adsorption of water from air in a fixed silica gel bed the authors found out that in the adsorption on a microporous silica gel, surface diffusion is the determining mechanism, while for macroporous silica gel, both Knudsen and surface diffusion are important. The kinetics and mechanism of water vapour adsorption on silica gel have been investigated using thermogravimetry under controlled temperature and water vapour pressure [11]. From experimental data by taking into account simultaneous mass and heat transfer effective diffusivity of 8×10^{-10} m^2 s^{-1} was calculated. By studying the kinetics of water vapour sorption in an apparatus

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*Presented at the 27th International Conference of the Slovak Society of Chemical Engineering, Tatranské Matliare, 22—26 May 2000.

with a constant volume and changing adsorptive pressure *Lu et al.* [12] revealed that the adsorption rate is dependent upon the silica gel particle diameter. Calculated values of the effective diffusion coefficient were within the range 2×10^{-9}—2.5×10^{-10} m^2 s^{-1}. In modelling nonisothermal drying by adsorption in a silica gel bed, *San* and *Jiang* [13] suggested a mass transfer resistance in the fluid phase with consideration of a fluid friction effect.

The authors of monographs [1, 2] reported the differences in the order of magnitude of effective diffusion coefficients of hydrocarbons on molecular sieve crystals obtained by various methods, but did not concern with the causes of these discrepancies in connection with the errors of individual methods. In the investigation of diffusion of chlorinated hydrocarbons in active carbon particles by a gravimetric method, the calculated error of estimated values of effective diffusion coefficients caused by neglecting the mass transfer resistance in the fluid phase directed towards the outer surface of adsorbent particle did not reach 20 % [14].

THEORETICAL

For unsteady mass transfer in a porous material the following equation can be derived from the balance of component A

$$\beta\frac{\partial c_\mathrm{A}}{\partial t} - R_\mathrm{A} = \nabla \cdot D''\nabla c_\mathrm{A} \qquad (1)$$

where c_A is the concentration of component A in the gas phase, R_A is the rate of production of component A in the volume of the porous body, D'' is the equivalent diffusion coefficient, β is porosity, and t is time. If the diffusion coefficient is equal in all directions, the porous material is isotropic in terms of mass transfer and eqn (*1*) can be rearranged into the following form

$$\beta\frac{\partial c_\mathrm{A}}{\partial t} - R_\mathrm{A} = D''\nabla^2 c_\mathrm{A} \qquad (2)$$

R_A represents a negative adsorption rate related to the volume of the porous adsorbent for parallel diffusion and adsorption in the porous material. Thus, eqn (*2*) becomes

$$\beta\frac{\partial c_\mathrm{A}}{\partial t} + \frac{\partial q_\mathrm{A}}{\partial t} = D''\nabla^2 c_\mathrm{A} \qquad (3)$$

For linear adsorption isotherms

$$q_\mathrm{A} = Kc_\mathrm{A} \qquad (4)$$

or

$$q_\mathrm{A} = K'c_\mathrm{A} + e \qquad (5)$$

Eqn (*3*) can be rearranged to the following expression

$$\frac{\partial c_\mathrm{A}}{\partial t} = D_\mathrm{e}\nabla^2 c_\mathrm{A} \qquad (6)$$

The effective diffusion coefficient is defined as

$$D_\mathrm{e} = \frac{D''}{\beta + K} \qquad (7)$$

The value of this coefficient depends on the diffusion mechanism in the pores of the solid phase and the properties of the system. If the transport of component A is carried out solely by molecular diffusion, then [15]

$$D_\mathrm{e} = \frac{D_\mathrm{AB}}{\beta + K}\frac{\beta}{k^2} \qquad (8)$$

In the case of Knudsen diffusion

$$D_\mathrm{e} = \frac{D_\mathrm{K}}{\beta + K}\frac{\beta}{k^2} \qquad (9)$$

For a symmetric diffusion in a spherical particle, eqn (*6*) has the following form

$$\frac{\partial c_\mathrm{A}}{\partial t} = D_\mathrm{e}\left[\frac{\partial^2 c_\mathrm{A}}{\partial r^2} + \frac{1}{r}\frac{\partial c_\mathrm{A}}{\partial r}\right] \qquad (10)$$

where r is the radial coordinate.

Initial and boundary conditions of the stepwise adsorption and/or desorption are as follows

$$\begin{array}{llll} t = 0 & 0 \leqq r \leqq r_\mathrm{o} & c_\mathrm{A} = c_\mathrm{Ao} & q_\mathrm{A} = q_\mathrm{Ao} \\ t > 0 & r = r_\mathrm{o} & c_\mathrm{A} = c_\mathrm{A1} & \end{array} \qquad (11)$$

Solutions of eqn (*10*) for condition (*11*) $c_\mathrm{A} = f(r,t)$ can be found in monograph [16]. For the sake of experimental determination of the effective diffusion coefficients, the solution of the stepwise successive adsorption is

$$\gamma_t = \frac{\Delta q_t}{\Delta q_\infty} = \frac{\Delta m_t}{\Delta m_\infty} = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-\frac{n^2\pi^2 D_\mathrm{e}t}{r_\mathrm{o}^2}\right) \qquad (12)$$

and of the stepwise successive desorption

$$\gamma_t = 1 - \frac{\Delta m_t}{\Delta m_\infty} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-\frac{n^2\pi^2 D_\mathrm{e}t}{r_\mathrm{o}^2}\right) \qquad (13)$$

where Δm_t, Δm_∞ and Δq_t, Δq_∞ are the increments of the adsorbed amount and adsorbate concentration at time t, and in equilibrium, respectively, and r_o is the particle radius.

EXPERIMENTAL

The silica gel employed, Davison – type H, consisted of particles of the following characteristics: mesh fraction 3—4 mm, mean diameter d_p = 3.46 mm, specific surface area S_p = 806 m^2 g^{-1}, specific volume of the transport pores V_p = 0.522 cm^3 g^{-1}, particle density ρ_p = 1.490 g cm^{-3}, porosity β = 0.778 cm^3 cm^{-3}.

Twice distilled water was used.

Prior to the measurement, all adsorbents were activated. The adsorbers filled with silica gel and activated carbon were heated in a laboratory oven to a temperature of 150 °C, while a small stream of air was

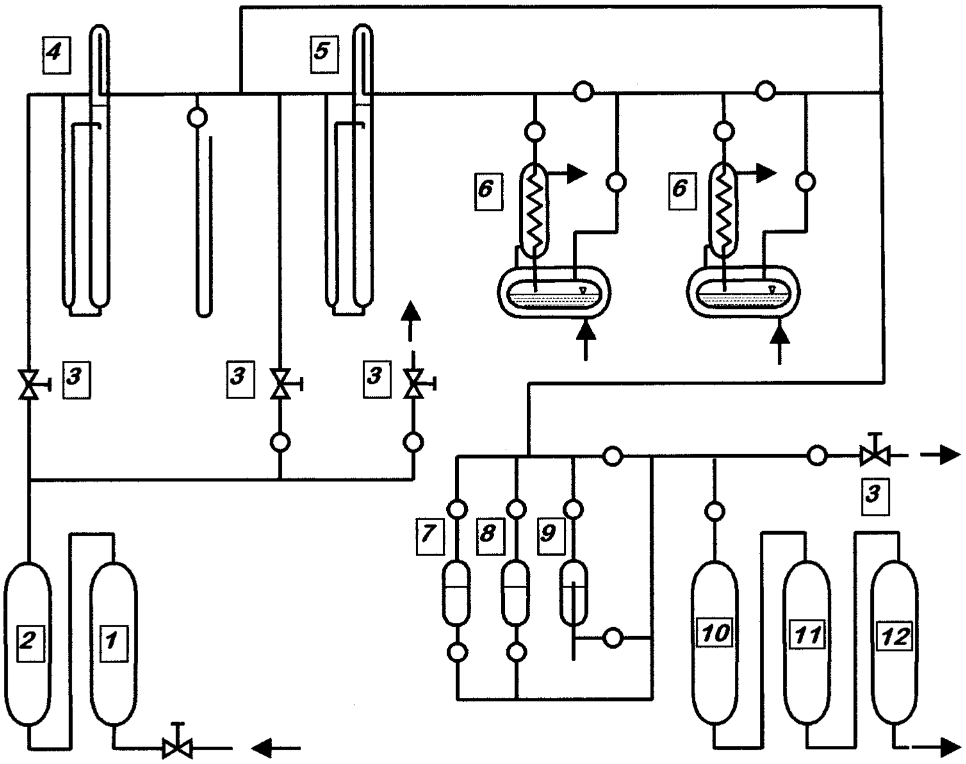

Fig. 1. Schematic diagram of apparatus employed: *1.* adsorber filled with silica gel, *2.* adsorber filled with molecular sieve, *3.* distribution valves, *4, 5.* flowmeters, *6.* saturators, *7, 8.* adsorbers containing differential bed of adsorbent, *9.* adsorber containing differential bed of adsorbent with thermocouple, *10, 11, 12.* adsorbers filled with silica gel and molecular sieve.

directed through these adsorbers for about 3 h. The adsober filled with a molecular sieve was heated in an oven to a temperature of 450 ℃ and thus activated at small flow-rates of air for about 2.5 h.

In investigating the course of the multistage adsorption and desorption a glass apparatus, schematically drawn in Fig. 1, was used. Air fed into the apparatus was purified in adsorbers *1* and *2* by entrapping organic impurities and water. The purified air was divided into two parts. In the first stream, air was saturated in the saturators *6* by evaporation of water so that after its mixing with an unsaturated stream, the resulting stream contained the required concentration of water. Air containing water vapour was released into the atmosphere until the system of saturators reached a steady state (in about 20 min). After reaching constant values of flow-rates and adsorptive concentration, the stream of saturated air was switched to adsorber *7* and adsorbers *10—12*. When a chosen adsorption time elapsed, the stream of air was switched to adsorber *8*, and adsorber *7* was weighed. When an equal adsorption time elapsed also for adsorber *8*, the stream was switched to adsorber *7* and this procedure was repeated until an equilibrium was reached. The concentration of adsorptive in the stream of air was then increased by a change of the ratio of the stream of air through the saturators and the stream of air outside of the saturators, or by the change of temperature in the saturators, whereby mixed streams of air outside of adsorbers were directed into the atmosphere. When the adsorptive concentration had reached a steady-state value, the second adsorption stage followed. After the completion of the second stage of adsorption, a further adsorption or desorption stage could follow using a stream of air with a chosen adsorptive concentration or a stream of pure air. The results of these measurements represent repeated sets of measurements $\{\gamma_{ti}; t_i\}$ for individual adsorption and desorption stages.

In order to evaluate the isothermal course of the sorption process, adsorber *9* was designed (Fig. 2) which was of the same size as adsorbers *7*, *8* but it moreover contained a single adsorbent particle with a bored hole in which a thermocouple was placed. The measurements were carried out in one of the adsorbers *7* or *8* and in adsorber *9* in equal adsorption or desorption times. Adsorber *9*, however, was not weighed, only the temperature in the adsorbent particle was examined during the measurements.

RESULTS AND DISCUSSION

The course of the adsorption of water from a stream of air and its desorption by a stream of pure air are depicted in Fig. 3. The courses of three adsorption stages and two desorption stages are in Fig. 4. Using the experimentally obtained set of data $\{\gamma_{ti}; t_i\}$ and employing eqn (*12*) for adsorption and eqn (*13*)

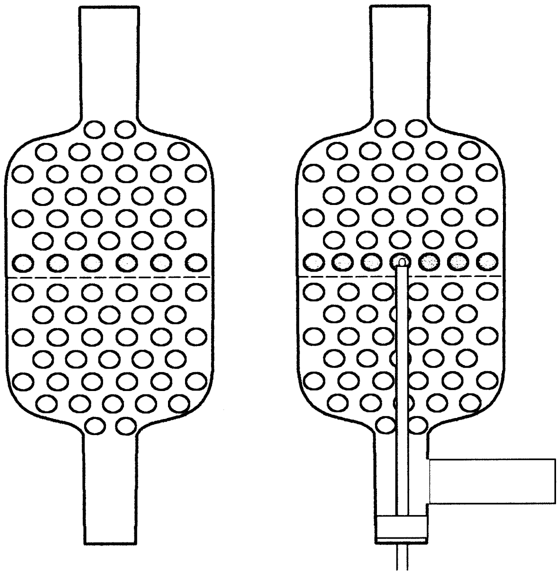

Fig. 2. Scheme of adsorbers with differential bed of adsorbent.

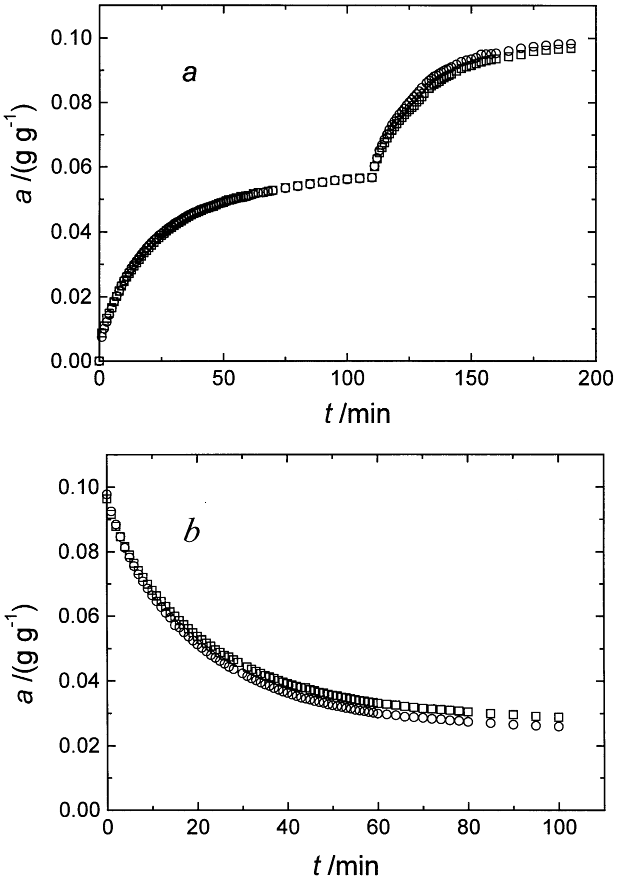

Fig. 3. Courses of two adsorption stages (*a*) and desorption (*b*): ☐ adsorber *7*, ○ adsorber *8*.

for desorption, it was possible to calculate the values of the effective diffusion coefficients by optimization methods. The following relationship was used as the optimization function

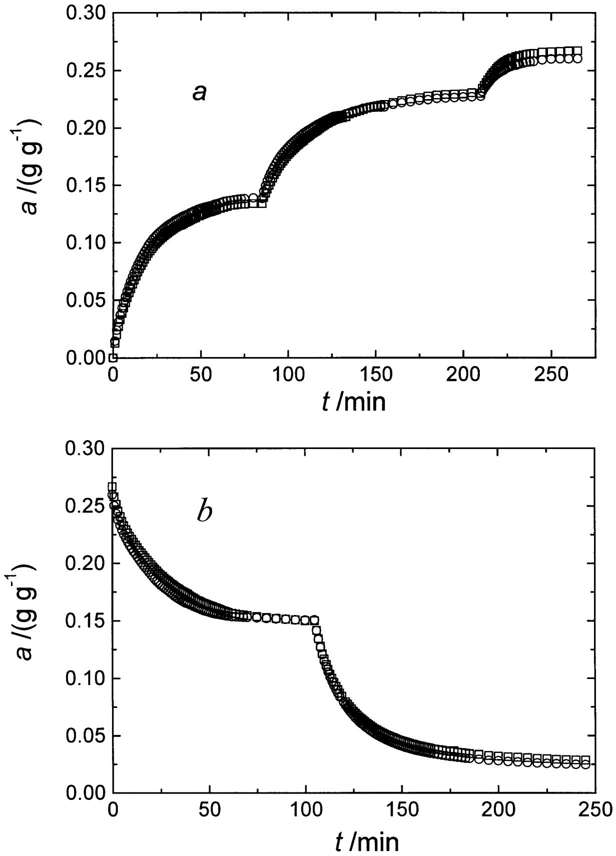

Fig. 4. Courses of three adsorption stages (*a*) and two desorption stages (*b*): ☐ adsorber *7*, ○ adsorber *8*.

$$F = \sum_{n=1}^{N} \left[(\gamma_t)_{\text{meas}} - (\gamma_t)_{\text{calc}}\right]^2 \qquad (14)$$

where $(\gamma_t)_{\text{meas}}$, $(\gamma_t)_{\text{calc}}$ are γ_t obtained by measurement and calculation, respectively, for the same time.

The results of these calculations together with experimental conditions are listed in Table 1. The calculated values of the effective diffusion coefficients are ranging from 0.747×10^{-10}—2.867×10^{-10} m^2 s^{-1}. They are of the same order of magnitude as values of the apparent diffusivity published in papers [8, 12] and the effective diffusivity measured in an atmosphere of pure water vapour [11, 17].

Error in the Estimation of D_e

The used measuring method requires a constant concentration of adsorptive on the outer surface of the adsorbent particles during the adsorption or desorption process. The fulfillment of this condition suggests that a zero resistance against mass transfer in the fluid phase has to be ensured. Moreover, the adsorbed amount must be negligible in comparison with the amount passing through the adsorber. It is obvi-

Table 1. Calculated Values of Effective Diffusion Coefficients

Measurement	Step	Temperature °C	$c_{A0} \cdot 10^3$ kg m^{-3}	q_∞ kg m^{-3}	$D_e \cdot 10^{10}$ m^2 s^{-1}	K	K'
1	1 – A	22.7	2.5	132.2	1.87	52 880	52 880
	1 – D	22.4	0		1.49		
2	1 – A	22.3	1.14	70.2	0.98	61 579	61 579
	2 – A	22.0	2.13	113.3	1.73	53 192	43 535
	1 – D	22.0	0		1.49	53 192	53 192
3	1 – A	23.0	1.47	84.5	1.42	57 469	57 469
	2 – A	22.5	2.92	145.4	1.56	49 794	42 014
	1 – D	22.2	0		1.56	49 794	49 794
4	1 – A	21.5	2.80	140.2	1.40	50 071	50 071
	2 – A	21.7	5.31	229.2	1.49	43 164	35 458
	3 – A	21.8	7.70	316.1	0.75	41 052	36 360
	1 – D	22.2	4.78	210.0	1.54	43 933	36 336
	2 – D	22.1	0		1.48	43 933	43 933
5	1 – A	21.5	4.47	209.9	1.82	46 957	46 957
	2 – A	22.0	8.47	320.0	1.12	37 780	27 525
	3 – A	22.0	12.0	394.5	2.87	32 875	21 105
	1 – D	22.0	5.22	226.5	1.51	43 390	24 779
	2 – D	22.0	0		1.54	43 390	43 390
6	1 – A	22.5	4.65	206.0	1.70	44 301	44 301
	2 – A	22.2	8.43	338.0	1.35	40 095	34 921
	3 – A	22.5	11.4	388.1	1.52	34 044	16 869
	1 – D	22.0	3.31	159.2	1.58	48 097	28 294
	2 – D	22.0	0		1.20	48 097	48 097

ous that with an increasing flow-rate of gas being saturated the resistance against mass transfer in the gas surrounding the adsorbent particle decreases. There exists, however, a certain minimal value of the resistance which varies only little with a further increase of turbulence. In our adsorbers, the respective value of flow-rate was 18 dm^3 min^{-1}. This can be documented on the dependence of D_e on the concentration of adsorptive, whereby the parameter is the flow-rate of gas [14]. The resistance against mass transfer in the fluid phase, however, only little affects the accuracy of the determined values D_e by the given method. In the described method, the flux of adsorptive from the gas into the particle $\tilde{n}_A$ can be expressed in accordance with the film theory

$$\tilde{n}_A = K_c\,(c_A - c_{As}) = k_{gc}\,(c_A - c_{Ai}) = k_{sq}\,(q_{Ai} - q_A) \tag{15}$$

where K_c is the overall mass-transfer coefficient and k_{gc}, k_{sq} are the individual mass transfer coefficients for the gas and solid phases, respectively.

The overall resistance against mass transfer could be expressed as follows

$$\frac{1}{K_c} = \frac{1}{k_{gc}} + \frac{1}{K k_{sq}} \tag{16}$$

or

$$R = R_g + R_s \tag{17}$$

The value of k_{gc} was calculated from dimensionless relations for the given arrangement of the system [18]. The value of k_{sq} was calculated from the relation [19]

$$k_{sq} = \frac{10 D_e}{d_p} \tag{18}$$

in which d_p is the diameter of the adsorbent particle.

The resistance against mass transfer in the fluid phase computed from eqn (*16*) did not exceed 12.5 % of the overall resistance. The relation used for the calculation of the individual mass-transfer coefficient in the fluid phase is valid for adsorbers with a fixed bed of adsorbent. The bed height of the adsorbers employed in our measurements was equal to the diameter of a single particle. The bed was covered from both sides by glass particles. The calculated values of Sherwood number and k_{gc} were then lower than the real values. Hence, the determined resistance against mass transfer in the fluid phase is maximal. The fact that the highest value of this resistance is 12.5 % of the overall resistance suggests that the value of the resistance against mass transfer in the porous solid phase can be determined with a maximal error of 12.5 %. By combining eqns (*16—18*) one gets the relationship

$$D_e = \frac{d_p}{10\,K R_s} \tag{19}$$

It follows that from the last equation the relative error in the estimation of D_e is given by the sum of relative errors of d_p, K, and R_s. The maximum relative error of the diameter of the particles is $[(4.00 - 3.46)/3.46] \times 100\ \% = 15.6\ \%$. The relative error of K estimated from the deviation of values K' calculated from experimental data and from the correlated dependence $q_{\mathrm{A}} = f(c_{\mathrm{A}})$ from equal concentration changes of water in air did not reach 10.5 %. Then, the overall error of the determined values of D_{e} does not exceed 40 %. For the second and further stages of successive adsorption the equilibrium constant of the system K' can be calculated from eqn (*5*).

The values of D_{e} obtained from the desorption data listed in Table 1 are ranging from 1.20×10^{-10} to 1.58×10^{-10} $\mathrm{m}^2\ \mathrm{s}^{-1}$. The major part of the values of the effective diffusion coefficients obtained from the adsorption data also lies in this interval. Supposing that D_{e} does not depend on adsorbate concentration, the arithmetic mean of all the calculated values is $D_{\mathrm{e}} = 1.52 \times 10^{-10}$. At a maximal error of 40 % the obtained values of D_{e} should be ranging from 0.91×10^{-10} to 2.13×10^{-10} $\mathrm{m}^2\ \mathrm{s}^{-1}$. Table 1 reveals that following two values lie outside of this interval: 0.75×10^{-10} $\mathrm{m}^2\ \mathrm{s}^{-1}$ (measurement 4) and 2.87×10^{-10} $\mathrm{m}^2\ \mathrm{s}^{-1}$(measurement 5). By excluding these values one gets the 95 % confidence interval of $D_{\mathrm{e}} = (1.5 \pm 0.6) \times 10^{-10}$ $\mathrm{m}^2\ \mathrm{s}^{-1}$. Thus, we can conclude that in the investigated interval of the adsorptive concentration, where the equilibrium equation can be considered linear, the mass transfer mechanisms during adsorption and desorption are the same. This conclusion is linked with the shape of the adsorption isotherm, in which no hysteresis is manifested in the investigated concentration interval, as can be seen in Fig. 5.

In the effort to judge the mechanism of water transport in the pores of silica gel for the measurement conditions, the diffusion coefficient of molecular diffusion of water in air was calculated. For these calculations, D_{AB} was 7.414×10^{-6} $\mathrm{m}^2\ \mathrm{s}^{-1}$ and the Knudsen diffusion coefficient in a straight capillary for silica gel of mean pore radius, $r_{\mathrm{k}} = 2V_{\mathrm{p}}/S_{\mathrm{p}} = 1.295 \times 10^{-9}$ m, was $D_{\mathrm{K}} = 5.179 \times 10^{-7}$ $\mathrm{m}^2\ \mathrm{s}^{-1}$. The mean value of the equilibrium constant K' was 39193. Thus, the effective diffusion coefficient was for the transport of water in silica gel pores by molecular diffusion in accordance with the model of waved pores

$$D_{\mathrm{e}} = \frac{D_{\mathrm{AB}}}{\beta + K'}\frac{\beta}{k^2} = \frac{7.414 \times 10^{-6} \times 0.778}{(0.778 + 39193) \times 1.2} = = 1.23 \times 10^{-10}\ \mathrm{m}^2\ \mathrm{s}^{-1} \qquad (20)$$

and for the transport by Knudsen diffusion

$$D_{\mathrm{e}} = \frac{D_{\mathrm{K}}}{\beta + K'}\frac{\beta}{k^2} = \frac{5.179 \times 10^{-7} \times 0.778}{(0.778 + 39193) \times 1.2} = = 8.57 \times 10^{-12}\ \mathrm{m}^2\ \mathrm{s}^{-1} \qquad (21)$$

The comparison of the calculated values of D_{e} with the values of D_{e} given in Table 1 demonstrates that only molecular diffusion and not Knudsen diffusion takes place during the transport of water in silica gel pores.

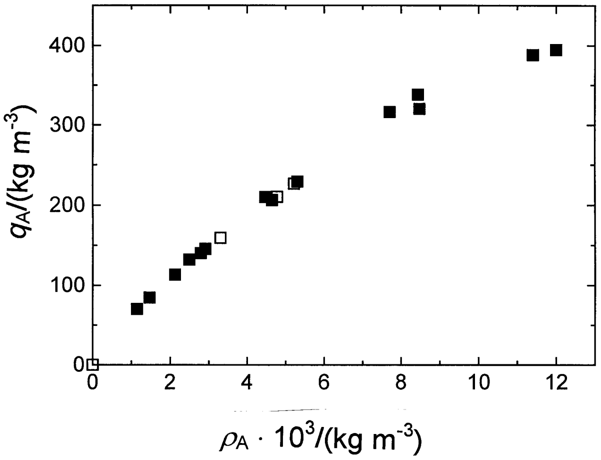

Fig. 5. Equilibrium points for the system water—silica gel obtained at adsorption and desorption steps: ■ adsorption, □ desorption.

CONCLUSION

The used mass transfer model in solid phase pores, in spite of numerous simplifications, allows a judgement of the transport mechanism. The employed laboratory apparatus enables to obtain values of the effective diffusion coefficients as a parameter necessary for the design of adsorption equipment. The relative error of the determined values of the effective diffusion coefficient does not exceed 40 %. The calculated value of the effective diffusivity of water in the presence of water in silica gel particles is $(1.5 \pm 0.6) \times 10^{-10}$ $\mathrm{m}^2\ \mathrm{s}^{-1}$ within the concentration range of adsorptive from 0 to 12 g m^{-3} at 20 ℃. Both the measurement conditions and the apparatus should be improved with the aim to decrease the errors of measurements.

SYMBOLS

c_{A}	concentration of component A in the gas phase	$\mathrm{kg\ m^{-3}}$
d_{p}	adsorbent particle diameter	m
D''	equivalent diffusion coefficient	$\mathrm{m^2\ s^{-1}}$
D_{AB}	molecular diffusion coefficient	$\mathrm{m^2\ s^{-1}}$
D_{e}	effective diffusion coefficient	$\mathrm{m^2\ s^{-1}}$
D_{K}	diffusion coefficient of Knudsen diffusion	$\mathrm{m^2\ s^{-1}}$
e	constant in eqn (*5*)	$\mathrm{kg\ m^{-3}}$
k	coefficient of wave of pores in eqn (*8*)	
k_{gc}	individual mass-transfer coefficient for the gas	$\mathrm{m\ s^{-1}}$
k_{sq}	individual mass-transfer coefficient for the solid phase	$\mathrm{m\ s^{-1}}$
K	equilibrium constant in eqn (*4*)	
K'	equilibrium constant in eqn (*5*)	
K_{c}	overall mass-transfer coefficient	$\mathrm{m\ s^{-1}}$

Δm_t	increment of the adsorbed amount at time t	kg
Δm_∞	increment of the adsorbed amount in equilibrium	kg
$\tilde{n}_\mathrm{A}$	adsorptive flux from the gas into the particle	kg m^{-2} s^{-1}
q_A	adsorbate concentration	kg m^{-3}
Δq_t	increment of the adsorbate concentration in time t	kg m^{-3}
Δq_∞	increment of the adsorbate concentration in equilibrium	kg m^{-3}
r	radial coordinate	m
r_o	the particle radius	m
R	overall resistance against mass transfer	s m^{-1}
R_g	individual resistance against mass transfer for the gas	s m^{-1}
R_s	individual resistance against mass transfer for the solid phase	s m^{-1}
R_A	rate of production of component A in the volume of the porous body	kg m^{-3} s^{-1}
β	porosity	m^3 m^{-3}
γ_t	dimensionless concentration defined by eqn (*12*) for adsorption or eqn (*13*) for desorption, respectively	
t	time	s

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