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THE SIMPLE PROCEDURE OF THE CALCULATION OF DIFFUSION COEFFICIENT FOR ADSORPTION ON SPHERICAL AND CYLINDRICAL ADSORBENT PARTICLES

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ABSTRACT

The simple method of the calculation of the diffusion coefficient of an adsorbed phase (D_e), based on the analytical solution of Fick's law of diffusion, for adsorption process is presented. Two shapes of the sorbent granules, i.e., spherical and cylindrical ones, are considered, and the adsorption process is assumed to take place in the Henry's region of the isotherm. Adopting the method proposed by Korta on the solution of Fick's law of diffusion, mathematically simple equations are developed to correlate the constant K of the analytical solution with the value of the relative adsorption as well as with the values of the geometrical parameters of the adsorbent granules. Using these simple equations, one can calculate the values of D_e and avoid, at the same time, mathematically advanced and time-consuming minimization procedure. Moreover, the procedure is likely to become complicated because of difficult math-

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ematical functions and operations occurring in the equations that describe the process, for example, Bessel function or the summation from unity to infinity. The presented simplified procedure is adopted for the results of paracetamol adsorption from water solution on two carbons with different shapes of granules, and it is shown that the obtained values of diffusion coefficients are practically the same as those calculated by means of the exact numerical procedure from the analytical solution.

INTRODUCTION

It is well known that the analytical solution of Fick's law of diffusion, in solids where internal diffusion plays a dominant role, is developed only for the Henry's region of an adsorption isotherm. This solution for the adsorption in solids possessing the spherical-shaped granules can be written as (1,2):

$$1 - \frac{a_t}{a_{\max}} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{10^{-\frac{0.4343 n^2 \pi^2 D_e t}{R^2}}}{n^2} \tag{1}$$

where a_t and a_{max} are the adsorption at the time t and the maximal adsorption (at infinite time), respectively, D_e is the effective diffusion coefficient of an adsorbed phase (valid for an adsorbed-phase concentration gradient driving force) and R is the radius of the sphere.

A similar solution, but for the adsorption process proceeding in solids possessing cylindrical-shaped granules (with the length of a cylinder equal to L), is given by (1,2):

$$1 - \frac{a_t}{a_{\max}} = \frac{32}{\pi^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{1}{\alpha_n^2 (2m-1)^2} \cdot 10^{-\frac{0.4343[\alpha_n^2 L^2 + (2m-1)^2 \pi^2 R^2] D_e t}{R^2 L^2}} \tag{2}$$

where R is the radius of the intersection of a cylinder, α_n are the roots of the equation $I_0(x) = 0$ and $I_0(x)$ is the Bessel function of the first kind 0 order.

The equations based on the analytical solution of Fick's law of diffusion (for example Eqs. 1 and/or 2) are widely used for the description of physisorption kinetics (3–5); however, their applicability can be limited due to the numerical fitting of Eqs. (1) and (2) to experimental results, which is a complicated and time-consuming procedure. To simplify the procedure of the solution the calculation of the average D_e value was postulated (2). However, in analyzing the kinetic data more thoroughly, it is interesting to know not only the average diffusion coefficient value but also to recognize how D_e values change while the adsorption process proceeds.

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THE METHOD OF THE CALCULATION OF D_E FROM THE ANALYTICAL SOLUTION OF FICK'S LAW

Korta (6), in his pioneering work, was the first to propose simplifying the method of the calculation of D_e values using Eqs. (1) and (2). He designed a simple procedure of the estimation of this coefficient for the cases where D_e changes during the adsorption process. Taking into account both the spherical and cylindrical adsorbent granules, Korta noticed that in Eqs. (1) and (2) one can denote:

$$D_e = KR^2/(\pi^2 t) \tag{3}$$

where K is a constant.

Equations (1) and (2) can be rewritten in the following form:

$$1 - \frac{a_t}{a_{\max}} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{10^{-0.4343 n^2 K}}{n^2} \tag{4}$$

$$1 - \frac{a_t}{a_{\max}} = \frac{32}{\pi^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{1}{\alpha_n^2 (2m-1)^2} \cdot 10^{-0.4343\left[\alpha_n^2 \frac{1}{\pi^2} + (2m-1)^2 \frac{R^2}{L^2}\right] K} \tag{5}$$

Korta calculated and tabulated the values of K for a/a_{max} from 0.1 up to 0.999 (14 points of $a/a_{\max}$) for spherical and cylindrical adsorbents (only two cases were considered, i.e., for L/R equal to 2 and 4). Because Korta's calculations were performed more than 30 years ago (6), they were restricted only to a relatively small number of sums in Eqs. (4) and (5) as well as to a small number of relative adsorption points and L/R values.

The aim of the current paper is to recalculate the data with greater precision (and for a larger number of cases) using the procedure developed by Korta. Besides, Korta's procedure was developed by us. It is shown that the results can be fitted by means of the equations describing the dependence of K on $a/a_{\max}$ (for spheres) and L/R (for cylinders). Moreover, it is postulated and proved in the current study that there exist general relationships between K and L/R, and the aim of this paper is to elaborate on them. Knowing these final equations, one can calculate D_e values for an arbitrarily chosen L/R value. Thus, the complicated and time-consuming fitting procedure is omitted.

THE SIMPLIFIED METHOD AND THE RESULTS

To develop the relationships $K = f(a/a_{\max}, L/R)$ with the closest possible accuracy, K values were calculated numerically (using Eqs. 4 and 5). The procedure of the numerical fitting was as follows: first the programs in Fortran 77 were

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created; then, the K values were found using the method of bisection for 255 of a/a_{max} points on the curve fitted by Eq. (4) (spheres); next, for the adsorption in cylinders (the method of bisection was also applied), the calculations were performed for 20 L/R values (shown in Table 1) (255 a/a_{max} points for each L/R), basing on the numerical recipes for the calculation of the roots of the Bessel functions of integer order (7). The accuracy of the fitting of the left sides of Eqs. (4) and (5) to the right ones was assumed as equal to $1\ 10^{-10}$. It means that the calculations of K values were performed until the difference between the left and the right sides of the both equations were smaller than 0.0000000001.

The minimum relative adsorption value was equal to 0.0025 whereas the maximum one was equal to 0.99. Because of the summation from unity to infinity, it was necessary to assume for each case (spheres and cylinders) the maximum (finite) number of sums in Eqs. (4) and (5). Thus, the elements of the summation

Table 1. The Parameters of *Eqs. (6–8)*. They were Obtained by the Simultaneous Fitting of All the Data Obtained from Numerical Calculations Using *Eqs. (4)* and *(5)* (the Data for Spheres and Some Data for Cylinders Are Shown in Figs. 1 and 2). DC is the Determination Coefficient Describing the Fit Between Calculated Numerically Data and These Obtained from *Eqs. (6–8)* Using the Parameters from the Table. Note That the Average DC Values Are Given in the Text

Sorbent		$0.0025 \le a/a_{max} \le 0.8$				$0.8 < a/a_{max} \le 0.99$				
		$K = a_{0s} b_s^{(a/a_{max})} (a/a_{max})^{cs}$				$K = ((A_s + B_s (a/a_{max}))/(1 + C_s(a/a_{max}) + Ds(a/a_{max})^2)$				
sphere		a_{0s}	bs	cs	DC	A_s	B_s	C_s	D_s	DC
		0.28572666	8.1511751	1.4528205	0.99999	0.28499707	−0.2839343	−1.9271993	0.9273312	0.99999
		$K = a_{0c} bc^{(a/a_{max})} (a/a_{max})^{cc}$				$K = (A_c + B_c (a/a_{max}))^{(-1/Cc)}$				
cylinder	L/R	a_0	b	c	DC	A_c	B_c	C_c	DC	
	0.5	0.106083	4.561839	1.671781	0.99987	18.359345	−17.380614	1.051021	0.99219	
	1.0	0.214901	6.448967	1.638375	0.99986	6.307463	−5.963633	1.015629	0.99519	
	1.25	0.258254	6.900238	1.622640	0.99982	4.854444	−4.586758	1.008609	0.99579	
	1.5	0.29346	7.213902	1.607512	0.99980	4.061067	−3.834716	1.004654	0.99617	
	1.75	0.321742	7.443262	1.592967	0.99981	3.579138	−3.377686	1.002415	0.99636	
	2.0	0.344461	7.617466	1.578981	0.99982	3.263210	−3.077899	1.001144	0.99643	
	2.5	0.377753	7.862821	1.552596	0.99986	2.884314	−2.717936	1.000010	0.99635	
	3.0	0.40021	8.025657	1.528194	0.99988	2.670328	−2.514181	0.999643	0.99614	
	3.5	0.415923	8.140335	1.505629	0.99989	2.534205	−2.384179	0.999524	0.99592	
	4.0	0.427296	8.224621	1.484774	0.99989	2.439582	−2.293488	0.999485	0.99572	
	4.5	0.435774	8.288591	1.465514	0.99989	2.369088	−2.225653	0.999473	0.99556	
	5.0	0.442257	8.338364	1.447748	0.99989	2.313575	−2.172008	0.999469	0.99542	
	5.5	0.447325	8.377862	1.431386	0.99989	2.267852	−2.127637	0.999467	0.99531	
	6.0	0.451363	8.40971	1.416348	0.99989	2.228796	−2.089581	0.999467	0.99521	
	6.5	0.454633	8.435726	1.402562	0.99989	2.194437	−2.055974	0.999467	0.99514	
	7.0	0.45732	8.457208	1.389964	0.99988	2.163483	−2.025592	0.999467	0.99506	
	7.5	0.459555	8.475103	1.378498	0.99988	2.135056	−1.997603	0.999467	0.99498	
	8.0	0.461436	8.490121	1.368114	0.99987	2.108544	−1.971430	0.999467	0.99487	
	10.0	0.466618	8.530555	1.336577	0.99984	2.014478	−1.878082	0.999467	0.99385	
	12.0	0.46965	8.552173	1.319643	0.99981	1.930718	−1.794498	0.999467	0.99102	

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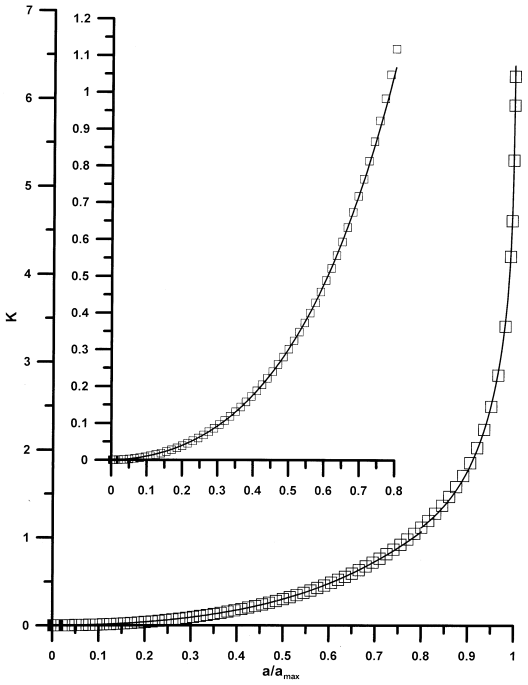

Figure 1. The fit between calculated, using Eq. (4), K values (squares—to make the figure more clear only ~100 among 255 calculated points are shown) for spherical adsorbent granules and the data calculated using Eqs. (6) and (7) (line) with the parameters for the sphere from Table 1.

in Eqs. (4) and (5) with the value of the exponents smaller than (–1000) were treated as negligible.

The results of that calculation for spherical and some cylindrical adsorbents are shown in Figs. 1 and 2 (the number of points shown in these figures is reduced due to the fact that displaying all 255 points leads to the complete covering of the fitted curve). To improve the accuracy of the approach presented in this paper, the data obtained in the calculations were divided into two groups.

Thus, in the range of $0.0025 \leq a/a_{\max} \leq 0.8$, the numerically calculated data for spheres (s) as well as for cylinders (c) can be successfully fitted, by the following relationship between K and $a/a_{\max}$:

$$K = a_{0c/s} b_{c/s}^{(a/a_{\max})} (a/a_{\max})^{c_{c/s}} \tag{6}$$

For the higher range of relative adsorption ($0.8 \leq a/a_{\max} \leq 0.99$), the numerically calculated data for spherical adsorbents can be fitted, with close accu-

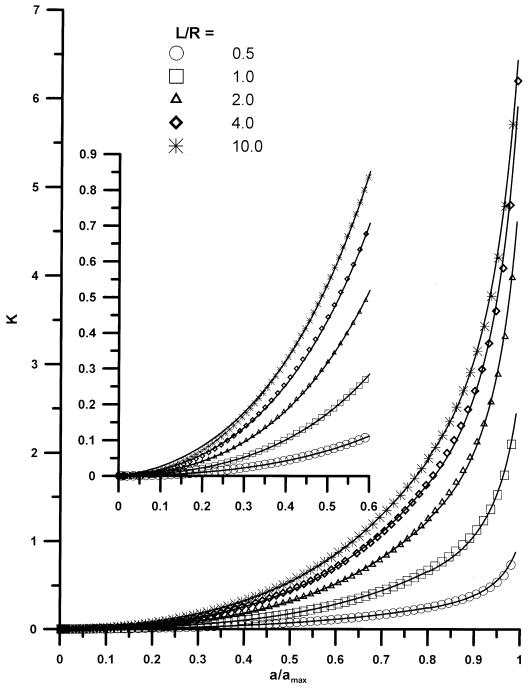

Figure 2. The fit between calculated, using Eq. (5), K values (symbols—to make the figure more clear only ~100 among 255 calculated points are shown) for cylindrical adsorbent granules (for arbitrarily chosen L/R values) and the data calculated using Eqs. (6) and (8) (line) with the parameters for the cylinders from Table1.

racy, by the following formula:

$$K = ((A_s + B_s(a/a_{\max}))/(1 + C_s(a/a_{\max}) + D_s(a/a_{\max})^2) \quad (7)$$

whereas for cylinders the following equation describes the obtained data with required accuracy:

$$K = (A_c + B_c(a/a_{\max}))^{(-1/C_c)} \quad (8)$$

The parameters of the fitting of the Eqs. (6) and (7) to the data calculated using Eqs. (4) and (5) for the both shapes of adsorbent granules (as well as for different L/R values) are presented in Table1. It is seen that the fit of the data calculated by the approximated procedure to those obtained by the exact numerical calculation is excellent.

Finally, an attempt was made to correlate all the parameters from Table 1 calculated for cylinders with the values of L/R. The purpose was to relate these pa-

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rameters into one equation (i.e., the dependence of the parameters a_0, b, c (Eq. 6) and A_c, B_c, C_c, D_c (Eq. 8) on L/R was evaluated). This was necessary because the knowledge of the dependence of these parameters on L/R makes it possible to calculate the K values not only for the cases tabulated in Table 1 but also for intermediate L/R (including fractional ones). The dependence of the parameters of Eqs. (6) and (7) on L/R is shown in Figs. 3 and 4.

It can be easily shown that the parameters of Eq. (6) depend on L/R in the following way:

$$a_{0c} = (a_1 b_1 + c_1 (L/R)^{d_1})/(b_1 + (L/R)^{d_1}) \tag{9}$$

where:

$a_1 = 2.520044\ 10^{-2}$
$b_1 = 1.387142$
$c_1 = 4.78042\ 10^{-1}$
$d_1 = 1.729134$

$$b_c = a_2 b_2^{(R/L)} (L/R)^{c_2} \tag{10}$$

where

$a_2 = 9.2374028$
$b_2 = 6.981364\ 10^{-1}$
$c_2 = -1.896661\ 10^{-2}$

$$c_c = 1/(a_3 + b_3 (L/R) + c_3 (L/R)^2) \tag{11}$$

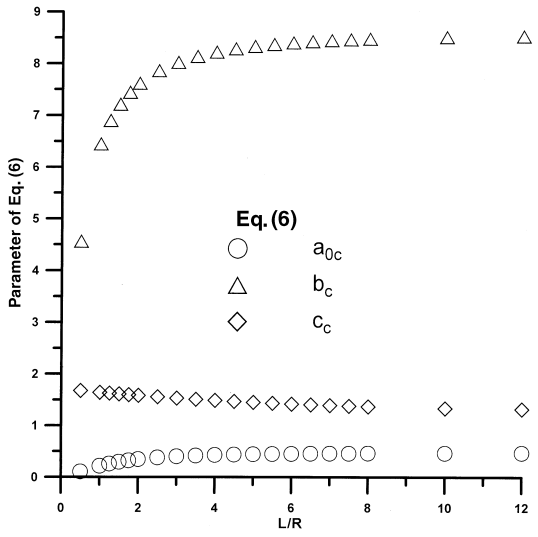

Figure 3. The dependence of the parameters a_{0c}, b_c, and c_c of Eq. (6) on L/R.

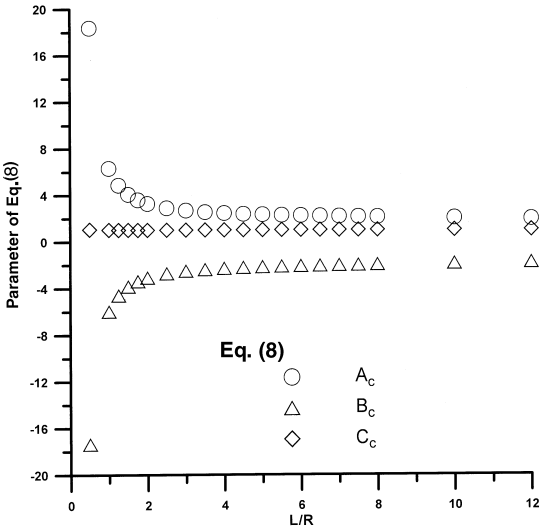

Figure 4. The dependence of the parameters A_c, B_c, and C_c of Eq. (8) on L/R.

where

$a_3 = 5.854904\ 10^{-1}$
$b_3 = 2.582611\ 10^{-2}$
$c_3 = -9.557168\ 10^{-4}$

The average determination coefficient calculated for all the 20 curves fitted by Eqs. (6), (9), (10), and (11) to the calculated numerically data is equal to 0.99986.

The same procedure applied for the parameters of Eq. (8) leads to:

$$A_c = A_1 + B_1(L/R) + C_1/(L/R)^2 \tag{12}$$

and

$A_1 = 2.331879$
$B_1 = -3.575147\ 10^{-2}$
$C_1 = 4.011335$

$$B_c = A_2 + B_2(L/R) + C_2/(L/R)^2 \tag{13}$$

where

$A_2 = -2.199958$
$B_2 = 3.59872110^{-2}$
$C_2 = -3.799663$

$$C_c = A_3/(1 + B_3 \exp(-C_3(L/R))) \tag{14}$$

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and

$A_3 = 9.994668\ 10^{-1}$
$B_3 = -1.511922\ 10^{-1}$
$C_3 = 2.251358$

The average determination coefficient calculated for all the 20 curves fitted by Eqs. (8), (12), (13), and (14) to the calculated numerically data is equal to 0.99513.

Due to the fact that the numerically calculated curves were fitted in two different ranges of the relative adsorption (up to a/a_{max} 0.8—Eq. 6 and $0.8 \leq a/a_{max} \leq 0.99$—Eqs. 7 and 8), at the point a/a_{max} is equal to 0.8, both sets of fitted curves give slightly different values of K. However, the differences in K values calculated from both models for this point are, for most of the cases, not larger than 1.5% (only for the extreme values of L/R the error is larger, but no more than 3.8%). Thus, the differences at this point are so small that they cannot lead to the creation of discontinuities on the curves of $D_e = f(a/a_{max})$.

VERIFICATION OF THE SIMPLIFIED METHOD USING EXPERIMENTAL DATA

To compare the results of the calculation of the diffusion coefficient using Eqs. (1) and (2) with those obtained as proposed in the current study the approximate procedure, two sets of paracetamol (4-hydroxyacetanilide) batch-reactor-test kinetic data published not long ago (8,9) were applied (they are presented in Fig. 5). The details of the kinetic measurements are as follows. The kinetics of adsorption was measured using a paddle agitator (60 rpm) at the initial solute concentration equal to 0.03 mol/l (125 cm^3 of paracetamol solution was applied). The sample preparation was the same as for the adsorption measurements (10–12). Each kinetic curve was measured at least twice. Each point on the kinetic curve was determined by pipetting 1 cm^3 of paracetamol solution and measuring absorbance using UV-VIS spectrophotometer; the procedure was described previously (8,9,11,12). Temperature was controlled with the same accuracy as during the adsorption measurements, which was described previously (11,12). The error of the kinetic measurement is not bigger than $\pm$ 0.03 (in the units of a/a_{max}).

As adsorbents, two de-ashed commercial activated carbons (characterized previously, (9–12)) with different shapes of granules were chosen. Therefore in the case of AHD carbon, the spherical fraction (fraction 0.75 mm $\leq$ 2R $<$1.2 mm) was separated by means of sieves. On the other hand, D43/1 carbon possesses only cylindrical granules. Some additional carbon characteristics (surface acidity, enthalpy of immersion in water, and others) were given previously (8,9).

The approximate procedure of the calculation of D_e is now as follows. For spherical adsorbent granules (AHD carbon), one can easily calculate K for an ar-

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bitrarily chosen a/a_{max} value using the parameters from Table 1 and Eqs. (6) and (7). Then, applying Eq. (3), the values of D_e (for AHD carbon $R = 0.04875$ cm) can be calculated. For cylinders (D43/1 carbon), the procedure is also simple. Knowing the value of L/R measured experimentally (for D43/1 carbon $L/R = 5.99512$), one can calculate K using the parameters from Table 1 and Eqs. (6) and/or (8). For the cases when L/R value is not included in Table 1 (it is in the range 0.5–12), the values of the parameters of Eqs. (6) and/or (8) should be calculated first using Eqs. (9–14) (such a procedure was applied for D43/1). Then, K values can be calculated from Eqs. (6) and (8). Finally, as in the case of spheres, the values of D_e can be calculated applying Eq. (3) where R (for D43/1 $R = 0.0615$ cm) is in this case the radius of the intersection of a cylinder.

The obtained values of D_e are compared in Fig. 6. From this figure it can be noticed that the proposed procedure (solid lines) leads to practically the same results as those obtained from the numerical fitting (symbols). Some differences occur at low a/a_{max} values (up to 0.2) where the simplified method leads to some slightly higher diffusion coefficients than calculated numerically ones; however, the differences are generally within the range of an experimental error and therefore they can be neglected. The verification of the simplified procedure for other experimental systems will be presented in the future.

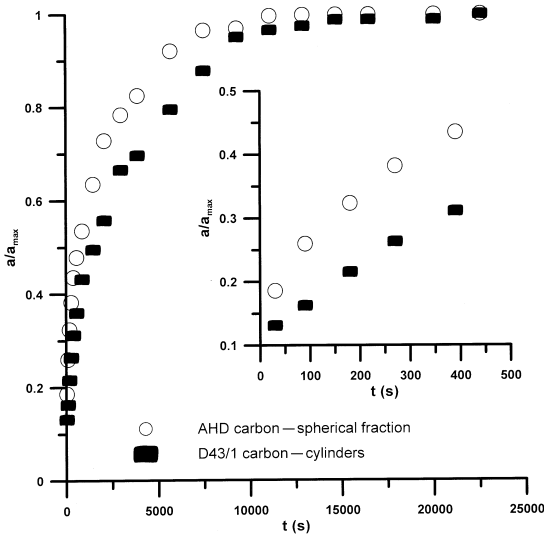

Figure 5. Measured experimentally data of paracetamol adsorption kinetics on carbons studied. The parameters are as follows: the temperature 320 K, paddle agitator was applied (60 rpm), the initial concentration of paracetamol solution was equal to 0.03 mole/l, the mass of carbon sample was equal to 0.5 g (correlated by mass loss after drying of carbon).

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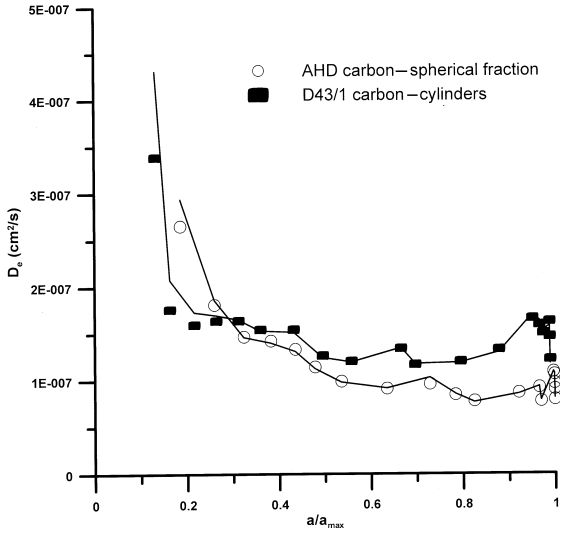

Figure 6. The comparison of the values of D_e calculated for the data presented in Fig. 5 by numerical fitting procedure (using Eqs.1 and 2—symbols) and using the simplified procedure described in the text (solid lines). For D43/1 carbon (cylindrical granules) $L/R = 5.99512$ and $R = 0.0615$ cm. For AHD carbon (spherical granules) $R = 0.04875$ cm.

COMPARISON WITH OTHER SIMPLIFIED METHODS

It should be pointed out that very good explicit approximations for the series solution in Eq. (1) (for adsorption on spherical adsorbents) are already available in the literature (13). However, for adsorption on cylinders the simplified equations presented in the current study are the first of this kind. The simplified approximations of Eq. (1) can be presented by the three equations. Thus, for $(a_t/a_{\max}) < 0.2$, the following equation can be used:

$$\frac{a_t}{a_{\max}} = \frac{6}{R}\left(\frac{D_e t}{\pi}\right)^{0.5} \tag{15}$$

in the range $0.2 < (a_t/a_{\max}) < 0.8$:

$$\frac{a_t}{a_{\max}} = \frac{6}{R}\left(\frac{D_e t}{\pi}\right)^{0.5} - \frac{3D_e t}{R^2} \tag{16}$$

and finally in the range $(a_t/a_{\max}) > 0.8$, the first term ($n = 1$) in Eq. (1) is generally sufficient for this range:

$$1 - \frac{a_t}{a_{\max}} = \frac{6}{\pi^2}\, 10^{-\frac{0.4343 n^2 \pi^2 D_e t}{R^2}} \tag{17}$$

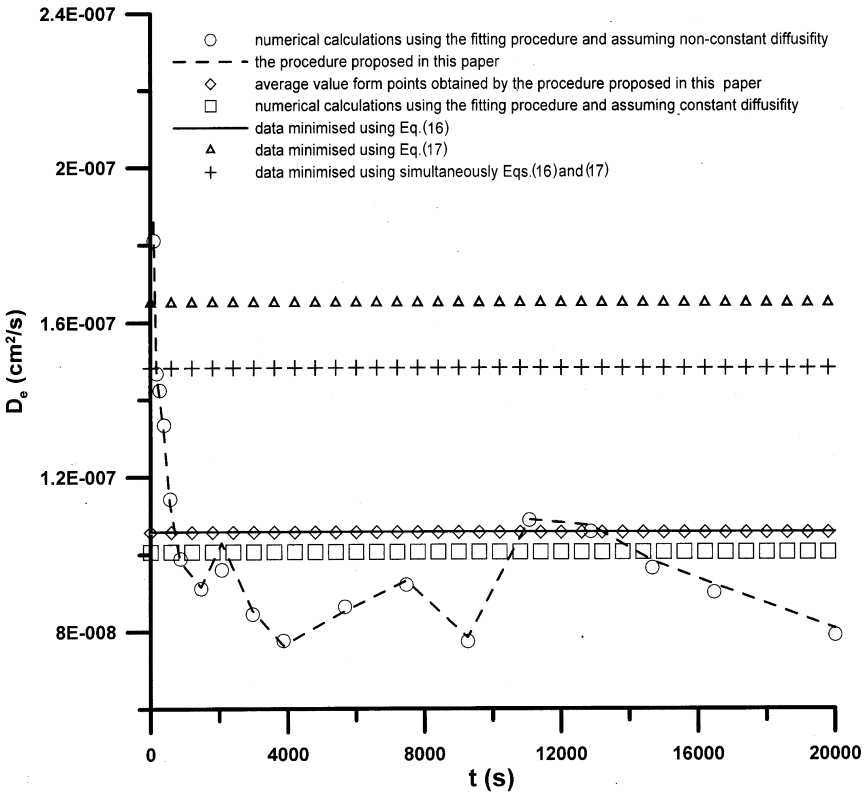

Figure 7. The comparison of the methods of D_e calculation for experimental data (the parameters are given in Fig. 6) of adsorption kinetics on AHD carbon.

Figure 7 shows the comparison of the effective diffusion coefficient values calculated from Eq. (1) (by the numerical fitting procedure described previously) with those obtained using the method from the current paper, as well as with those obtained using Eqs. (16) and (17) (Eq. 15 was not applied because only one experimental point was measured in this range). The two equations were applied as follows: They were fitted to the data independently, as well as simultaneously, assuming in this case that the same D_e values should be obtained from the both equations. Because it may seem to be mathematically incorrect to extract a concentration-dependent diffusivity by fitting experimental data to a model solution that assumes D_e constant in the first place, in this figure we also include the average values of those calculated by the method proposed in this study. Moreover, D_e value calculated by the numerical fitting procedure applying Eq. (1) but assuming that D_e is constant is also shown ($D_c = 0.9847$). From this figure it can be easily seen that if one does not agree that the presented procedure can lead to the determination of the concentration dependence of D_e, one can calculate the average

value of D_e, and the obtained result is almost the same as obtained from Eq. (1), but assuming constant diffusivity. Moreover, Eq. (16) leads also to satisfactory results ($D_c = 0.9459$), however Eq. (17) ($D_c = 0.9794$) as well as the simultaneous minimization of Eqs. (16) and (17) ($D_c = 0.8679$) leads to enormous differences between D_e values obtained and calculated from the exact numerical procedure. It should be also pointed out that that the substitution of D_e value obtained from the optimization of Eq. (16) into Eq. (17) leads to the extremely poor fit of the latter one to experimental data (negative value of D_c is obtained). On the other hand, D_e calculated by Eq. (17) and substituted into Eq. (16) leads to D_c equal to 0.7085 for this equation.

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