A Comparison of Experimental and Theoretical Adsorption Kinetics of Dichloromethane Vapour by Active Carbon under Non-isothermal Conditions

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The kinetics of the adsorption of dichloromethane vapour by a typical active carbon have been measured at 288 and 323 K and for spherical granules of 0.25 and 1.0 mm in diameter. The data have been analysed within the framework of a model for adsorption by bidispersed systems and under non-isothermal conditions. Micropore diffusion is the rate-controlling step, and direct measurements show that the process is not isothermal. However, thermal effects do not seem to influence significantly the kinetics of adsorption. Good agreement can be found between experimental and calculated data, and the model leads to micropore diffusivities and heat-transfer coefficients for the system under investigation.

From a theoretical point of view, the kinetics of vapour adsorption in bidispersed particles of activated carbon may be controlled by two important mechanisms: (*a*) heat transfer in the particle itself and towards the surroundings, which is reflected in the evolution of the temperature within the sample as adsorption proceeds, and (*b*) mass transfer in both the transport pores (macropores) and the micropores of the carbon. The influence of mass diffusion in the macropores can be assessed from the effect of the particle size: if the kinetics of adsorption are controlled by diffusion in the macropores, the timescale will be approximately an inverse function of the square of the average particle size. On the other hand, if diffusion in the micropores is the controlling factor, the timescale will be nearly independent of the particle size.

The relative influence of these factors on the kinetics of the adsorption of gases by zeolites has been examined in detail by Ruthven[1,2] and by the Soviet school of Dubinin.[3-6] On the other hand, in the case of active carbons, similar studies are still limited and deal mostly with isothermal models,[7] although it was pointed out some years ago that heat transfer may play a role.[8]

In the present paper we describe and analyse kinetic experiments carried out at 288 and 323 K for the adsorption of CH_2Cl_2 by a well defined active carbon.[9-11] The experimental data have been analysed using a mathematical model for adsorption by bidispersed adsorbents developed at Orsay.[12,13]

Experimental

The equipment used to determine the uptake of dichloromethane vapour by carbon F-02 as a function of time consisted of a microbalance (Mettler Ltd, Switzerland, type ME-21) containing *ca.* 200–300 mg of carbon in a glass bucket. The mass and its changes in the course of adsorption were determined with an accuracy of $\pm 10^{-5}$ g. The solid was

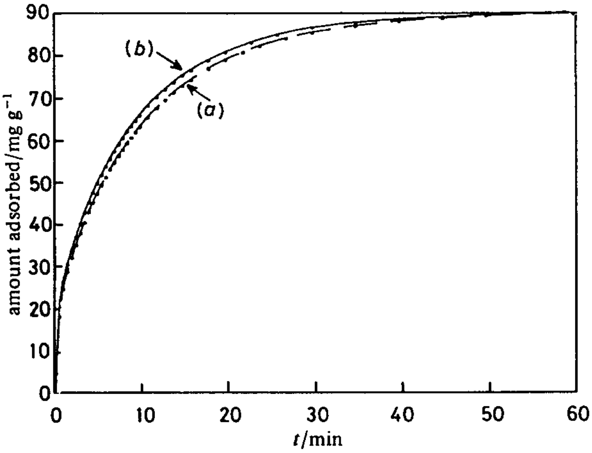

Fig. 1. Experimental uptake curves for pellets of two different sizes: (*a*) $R_p = 1$ mm; (*b*) $R_p = 0.25$ mm.

outgassed *in situ* for 20 h at 400 °C, under vacuum, by using a rotary pump and an oil diffusion pump. The pressures were measured with a capacitor gauge.

After outgassing, the lower part of the balance was surrounded by a thermostatted bath, kept to within ± 0.01 K at 288.15 or 323.15 K. Liquid CH_2Cl_2, outgassed and stored, provided the vapour to be fed into the system. The pressure of the vapour, to be kept constant during the adsorption experiment, was controlled manually by adjusting a needle valve. It was possible to maintain the pressure within $\pm 5.10^{-4}$ Torr† of the desired value.

Adsorption experiments began with a state of equilibrium, at pressures of 1–18 mmHg and $T = 288.15$ or 323.15 K. By opening and carefully adjusting the valve, the new constant pressure (2–30 Torr) was reached within seconds and the weight increase was recorded every 30 s over a period of 1 h. Typical plots of the amount adsorbed as a function of time are shown in fig. 1.

In order to determine the temperature profile in the sample as a function of time, adsorption experiments were repeated under the same conditions but without weighing. Three copper–constantan thermocouples were wired in series in the bucket containing the carbon. The reference junctions of the thermocouples were located in the vapour phase *ca.* 15–20 cm above the bucket. This device had a sensitivity of 121 μV K^{-1} near 298 K, and temperature differences between the gas phase and the carbon bed could be measured with an accuracy of ± 0.01 K. In fig. 2 are shown temperature deviations for adsorption and desorption experiments with differential pressure steps. The temperature deviations depend on the pressure steps, but do not exceed 4–6 K. It was found experimentally that the kinetics and the temperature curves were identical in absolute values for adsorption and desorption, and therefore the diffusion coefficients have been calculated for adsorption data only.

The static adsorption isotherm for the system CH_2Cl_2–carbon F-02 is the Dubinin–Radushkevich equation:

$$W = W_0 \exp\left[-\left(\frac{RT}{\beta E_0}\ln\frac{p}{p_s}\right)^2\right] \tag{1}$$

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† 1 Torr = 1 mmHg = 101 325/760 Pa.

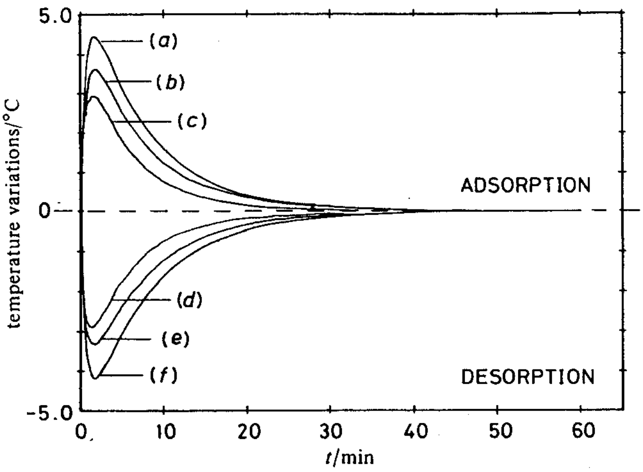

Fig. 2. Experimental temperature measurements for $R_p = 1$ mm and $T_0 = 15$ °C. Δp/mmHg = (*a*) 3.0–6.0, (*b*) 6.0–10.5, (*c*) 18.0–30.0, (*d*) 30.0–18.0, (*e*) 10.5–6.0 and (*f*) 6.0–3.0.

where W is the volume of adsorbate condensed in the micropores of the carbon at a temperature T and relative pressure p/p_s, W_0 is the total volume of the micropores accessible to the vapour, and β and E_0 are specific parameters of the system under investigation; β depends on the adsorbate, whereas E_0 is a characteristic of the micropore system.[1,10]

The adsorbent used in the present study had a micropore volume $W_0 = 0.64$ cm^3 g^{-1} and a characteristic adsorption energy $E_0 = 18.7$ kJ mol^{-1}. The external surface area was 36 m^2 g^{-1}, corresponding to the walls of the macropores, and the total micropore surface was close to 1000 m^2 g^{-1}. The average width of the ideally slit-shaped micropores derived from E_0 was *ca.* 1.2 nm, with a relatively large spectrum.[10] The solid was sieved and particles with diameters of either 1.0 ± 0.2 or 0.25 ± 0.05 mm were used, in order to assess the possible influence of the macropore diffusion on adsorption kinetics.

The main characteristics of the adsorbate,[14] CH_2Cl_2, are $\beta = 0.66$, a molar volume $V_m(298\ K) = 64.02$ cm^3 mol^{-1}, and a thermal expansion coefficient $\alpha = 1.339 \times 10^{-3}$ K^{-1}. The saturation pressures p_s at 288.15 and 323.15 K are, respectively, 281.5 and 1069.9 Torr.

Theory

The kinetics of adsorption in a bidispersed adsorbent pellet have been studied extensively and the literature provides either isothermal models, as proposed by Ruckenstein *et al.*,[15] Dubinin,[7] Ma[16] and Lee,[17] or non-isothermal models.[1-5,12,13] (These models have been applied mostly to adsorption by zeolites.)

In these models, the following assumptions are usually made: (*a*) the pellet is spherical and consists of small microporous particles; (*b*) the microparticles have simple shapes and are of uniform size; (*c*) the adsorption equilibrium is kept on the surface of the pellet and on the surface of the microporous particles; (*d*) the diffusion coefficients are constant and the adsorption isotherms are either linear or can be linearized.

The recent model of Sun and Meunier[13] contains the following additional assumptions and conditions: (*e*) adsorption on the walls of the macropores is small, relative to the sorptive capacity of the micropores, and can therefore be neglected; (*f*) the temperature is only considered at the pellet's scale and is uniform within the microparticles; (*g*) the gradient of μ/T is taken as the driving force for mass diffusion, where μ and T are the chemical potential and temperature, respectively. The diffusion flux is given by the relation:

$$J_{\mathrm{m}} = -BqT\nabla\frac{\mu}{T} \tag{2}$$

where B is the mobility and q is the amount adsorbed.

Experimental results (fig. 1) show that the kinetics of adsorption do not depend on the pellet size. This implies that the macropore diffusion is very rapid with respect to the micropore diffusion and can be reasonably considered as instantaneous. Consequently a simplified model which takes into account heat transfer and micropore diffusion is used for the present system.

Derivation of the Simplified Model

Supposing that the mass of vapour is very small relative to the adsorbed mass, the diffusion in the spherical microparticles of radius R_{i} can be described by

$$\frac{\partial q}{\partial t} = \frac{1}{r_{\mathrm{i}}^2}\frac{\partial}{\partial r_{\mathrm{i}}}\left(r_{\mathrm{i}}^2\, BqT\frac{\partial \mu_{\mathrm{i}}/T}{\partial r_{\mathrm{i}}}\right). \tag{3}$$

The temperature being independent of the radial coordinate r_{i}, the above equation can be reduced to a Fickian equation:

$$\frac{\partial q}{\partial t} = \frac{1}{r_{\mathrm{i}}^2}\frac{\partial}{\partial r_{\mathrm{i}}}\left(r_{\mathrm{i}}^2 D\frac{\partial q}{\partial r_{\mathrm{i}}}\right) \tag{4}$$

where D is the effective diffusivity based on the amount adsorbed and is defined by

$$D = Bq\left(\frac{\partial \mu_{\mathrm{i}}}{\partial q}\right)_T = D_0\frac{\partial \ln p}{\partial \ln q}. \tag{5}$$

D_0 is termed the corrected diffusivity.[1]

The diffusion in the macropores is instantaneous (the corresponding diffusivity is infinite):

$$\frac{\mu_{\mathrm{p}}}{T} = \frac{\mu(p_{\infty}, T_0)}{T_0}. \tag{6}$$

The heat balance in the pellet is given by Fourier's law, with a heat source generated by the adsorption in the micropores:

$$\rho c_{\mathrm{s}}\frac{\partial T}{\partial t} - \Delta H n 4\pi R_{\mathrm{i}}^2 D\left.\frac{\partial q}{\partial r_{\mathrm{i}}}\right|_{r_{\mathrm{i}}=R_{\mathrm{i}}} = \frac{\lambda}{r_{\mathrm{p}}^2}\frac{\partial}{\partial r_{\mathrm{p}}}\left(r_{\mathrm{p}}^2\frac{\partial T}{\partial r_{\mathrm{p}}}\right) \tag{7}$$

where ρ is the apparent pellet density, c_{s} is the heat capacity of the pellet, ΔH is the heat of adsorption, n is the number of microparticles per unit pellet volume, λ is the effective thermal conductivity and r_{p} is the radial coordinate at the pellet's scale.

The appropriate initial conditions are written as

$$q = q_0 \tag{8}$$

$$T = T_0 \tag{9}$$

Table 1. Experimental data and values of transfer coefficients determined by moments method

(*a*) Pellets of $R_p = 1$ mm at $T_0 = 15$ °C

pressure step /mmHg	S_m^0 /s	S_T^0 /s	h /W K^{-1} m^{-2}	(D/R_i^2) /10^{-4} s^{-1}	(D_0/R_i^2) /10^{-4} s^{-1}
1–2	420.87	7.25	6.57	1.932	0.774
2–3	438.84	4.74	6.21	1.813	0.666
3–6	443.80	8.55	6.15	1.813	0.578
6–10.5	407.50	6.86	6.05	1.967	0.533
10.5–18	354.74	5.89	6.56	2.226	0.510
18–30	282.74	4.71	6.71	2.768	0.524

(*b*) Pellets of $R_p = 0.25$ mm at $T_0 = 15$ °C ($h = 1.6$ W K^{-1} m^{-2})

pressure step /mmHg	S_m^0 /s	(D/R_i^2) /10^{-4} s^{-1}	(D_0/R_i^2) /10^{-4} s^{-1}
1–2	471.29	1.686	0.675
2–3	487.97	1.632	0.586
3–6	489.68	1.612	0.512
6–10.5	461.34	1.698	0.459
10.5–18	411.48	1.872	0.428
18–30	331.16	2.305	0.436

(*c*) Pellets of $R_p = 1$ mm at $T_0 = 50$ °C ($h = 6$ W K^{-1} m^{-2})

pressure step /mmHg	S_m^0 /s	(D/R_i^2) /10^{-4} s^{-1}	(D_0/R_i^2) /10^{-4} s^{-1}
1–3	159.04	5.757	3.508
3–7	210.06	4.201	2.172
7–13	254.14	3.313	1.482
13–21	269.98	3.042	1.202
21–30	268.43	3.014	1.071
30–45	263.75	3.017	0.963
45–70	243.47	3.222	0.899

while for the boundary conditions we have the following:

symmetry at sphere centres:

$$\left.\frac{\partial q}{\partial r_i}\right|_{r_i=0} = 0 \tag{10}$$

$$\left.\frac{\partial T}{\partial r_p}\right|_{r_p=0} = 0 \tag{11}$$

thermodynamic equilibrium at the microparticle surface:

$$\left.\frac{\mu_i}{T}\right|_{r_i=R_i} = \frac{\mu_p}{T} = \frac{\mu(p_\infty, T_0)}{T_0} \tag{12}$$

heat exchange between the pellet and its surroundings:

$$-\lambda \frac{\partial T}{\partial r_p}\bigg|_{r_p=R_p} = h(T\,|_{r_p=R_p} - T_0) \tag{13}$$

h being the overall heat transfer coefficient and R_p the pellet radius.

This set of partial differential equations holds for both linear and non-linear adsorption isotherms. For the sake of simplicity, we will assume that the experimental pressure steps are so small that a linear system can be used. Since adsorption isotherms are non-linear, they must be linearized. According to our study on the influence of non-linear effects (the results of which will be presented elsewhere), the reference point about which the non-linear relations are linearized should be $[p^* = (p_0 p_\infty)^{\frac{1}{2}}, T_0]$ in order to attenuate non-linear effects.

After linearization eqn (12) becomes

$$q\,|_{r_i=R_i} - q^* + \delta_{\mu i}(T - T_0) = q_\infty - q_0 \tag{14}$$

with

$$\delta_{\mu i} = \left(\frac{\partial \mu_i/T}{\partial T}\right)_q^* \bigg/ \left(\frac{\partial \mu_i/T}{\partial q}\right)_T^*.$$

At this stage it is convenient to define the following normalized variables and dimensionless parameters in order to obtain a normalized system of equations.

Normalized variables:

$$\omega = \frac{q - q_0}{q_\infty - q_0} \quad \theta = \frac{T}{T_0} \quad \tau = \frac{Dt}{R_i^2} \quad \psi = \frac{r_i}{R_i} \quad \eta = \frac{r_p}{R_p}. \tag{15}$$

Dimensionless parameters:

$$K_0 = (1 - \varepsilon_p)\frac{\Delta H(q_\infty - q_0)}{pc_s T_0} \quad Le = \frac{a_1 R_i^2}{DR_p^2} \quad Bi = \frac{hR_p}{\lambda} \quad P_{nt} = \frac{\delta_{\mu i} T_0}{q_\infty - q_0}. \tag{16}$$

Here, K_0 is the ratio of heat generation due to the adsorption and the heat capacity; the Lewis number, *Le*, stands for the ratio of mass diffusion resistance within the microparticles and heat conduction resistance in the pellet, and the Biot number *Bi* represents the ratio of the internal and external heat transfer resistances.

The system is rewritten as

$$\frac{\partial \omega}{\partial \tau} = \frac{1}{\psi^2}\frac{\partial}{\partial \psi}\left(\psi^2 \frac{\partial \omega}{\partial \psi}\right) \tag{17}$$

$$\omega(\psi, 0) = 0 \tag{18}$$

$$\frac{\partial \omega}{\partial \psi}\bigg|_{\psi=0} = 0 \tag{19}$$

$$\omega\,|_{\psi=1} + P_{ni}(\theta - 1) = 1 \tag{20}$$

$$\frac{\partial \theta}{\partial \tau} - 3K_0 \frac{\partial \omega}{\partial \psi}\bigg|_{\psi=1} = \frac{Le}{\eta^2}\frac{\partial}{\partial \eta}\left(\eta^2 \frac{\partial \theta}{\partial \eta}\right) \tag{21}$$

$$\theta(\eta, 0) = 1 \tag{22}$$

$$\frac{\partial \theta}{\partial \eta}\bigg|_{\eta=0} = 0 \tag{23}$$

$$-\frac{\partial \theta}{\partial \eta}\bigg|_{\eta=1} = Bi(\theta_s - 1) \tag{24}$$

where θ_s is the normalized surface temperature.

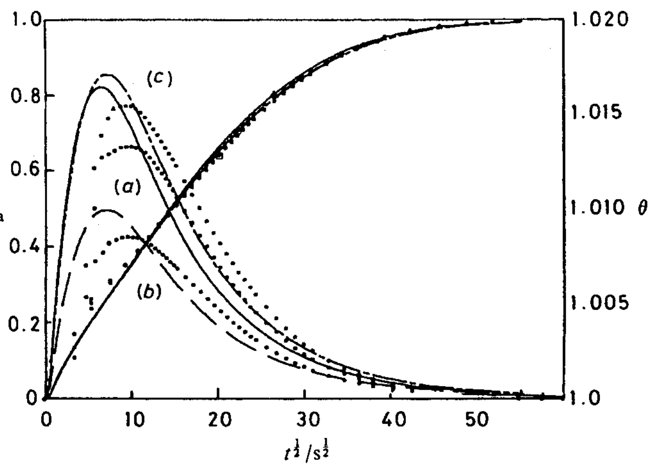

Fig. 3. Comparison of experimental (points) and calculated (best fit) (lines) uptake ω_a and surface temperature θ_s for $R_p = 1$ mm and $T_0 = 15$ °C. Δp/mmHg = (*a*) 1–2, (*b*) 2–3 and (*c*) 3–6.

Results and Discussion

In order to obtain the solution of the above system, we must know values of all the parameters of the system. Some parameters can be derived from the adsorption isotherms (as P_{ni} and K_0), or from data found in the literature (as λ). The two transfer coefficients D and h have to be determined from the comparison between experimental measurements and theoretical results of this model.

Owing to its simplicity, the statistical moments method will be used here. Moments of the system presented above [eqn (17)–(24)] can be determined by using the following relation between moments in the time domain and derivatives in the Laplace domain:

$$S_m^n = (-1)^n \lim_{s\to 0}\left[\frac{d^n}{ds^n}\left(\frac{1}{s}-\tilde{\omega}_a\right)\right] \tag{25}$$

$$S_T^n = (-1)^n \lim_{s\to 0}\left[\frac{d^n}{ds^n}\left(\tilde{\theta}_s-\frac{1}{s}\right)\right] \tag{26}$$

where ω_a is the normalized adsorbate concentration averaged over the pellet volume, S_m^n and S_T^n are nth moments of the variables $(1-\omega_a)$ and (θ_s-1), s is the Laplace transform variable and a tilde over ω_a and θ_s denotes the Laplace transform of the variable. Then, the zeroth moments can be easily obtained after transformation of the system in the Laplace domain:

$$S_m^0 = \int_0^\infty (1-\omega_a)\,dt = \lim_{s\to 0}\left(\frac{1}{s}-\tilde{\omega}\right) = \frac{1}{15}\frac{R_i^2}{D} + P_{ni}K_0\left(\frac{R_p^2}{15a_q}+\frac{R_p}{3h/\rho c_s}\right) \tag{27}$$

$$S_T^0 = \int_0^\infty (\theta_s-1)\,dt = \lim_{s\to 0}\left(\tilde{\theta}_s-\frac{1}{s}\right) = \frac{K_0}{3}\frac{R_p}{h/\rho c_s}. \tag{28}$$

Consequently, experimental values of S_m^0 and S_T^0 give directly D and h. With these coefficients, the solution of the system can be easily obtained using finite-difference methods.

Several experimental runs were made for three different conditions: (1) large pellets

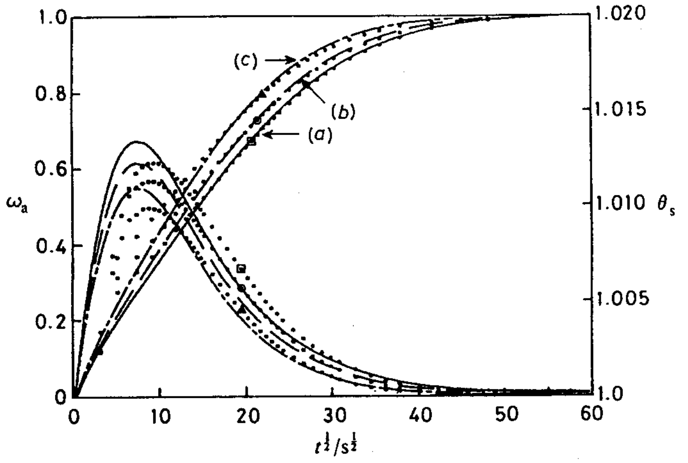

Fig. 4. Comparison of experimental (points) and calculated (best fit) (lines) uptake ω_a and surface temperature θ_s for $R_p = 1$ mm and $T_0 = 15$ °C. Δp/mmHg = (*a*) 6–10.5, (*b*) 10.5–18 and (*c*) 18–30.

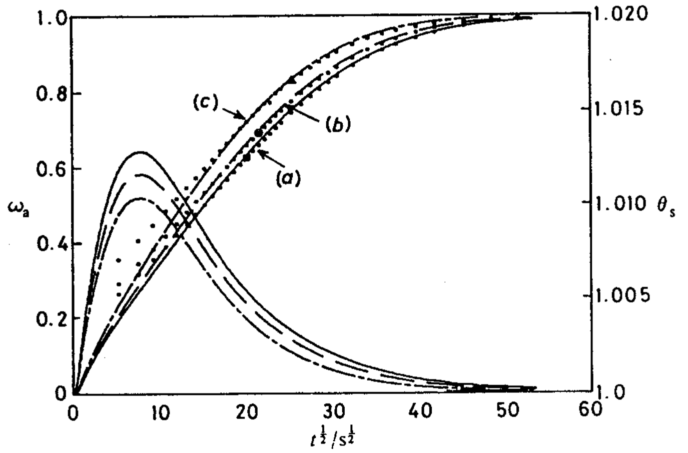

Fig. 5. Experimental (points) and calculated (lines) uptake ω_a and surface temperature θ_s for $R_p = 0.25$ mm, $T_0 = 15$ °C. Δp/mmHg = (*a*) 6–10.5, (*b*) 10.5–18 and (*c*) 18–30.

($R_p = 1$ mm) with $T_0 = 15$ °C; (2) small pellets ($R_p = 0.25$ mm) with $T_0 = 15$ °C; and (3) large pellets ($R_p = 1$ mm) with $T_0 = 50$ °C. Small differential pressure steps were applied for each condition. Table 1 gives experimental and calculated values of the transfer coefficients D (as well as D_0) and h. Since no temperature measurements were obtainable for the last two cases, the heat transfer coefficient h had to be estimated.

Table 1 shows that the values of the effective diffusivity D and the corrected diffusivity D_0 exhibit a slight dependence on the concentration and an increase with temperature. Furthermore, the diffusivities determined for pellets of two different sizes are consistent in value: they are nearly independent of the pellet size R_p.

The validity of the calculated transfer coefficients given in table 1 can be confirmed from the satisfactory agreement between experimental uptake and temperature measurements and predicted results, as can be observed in fig. 3–6.

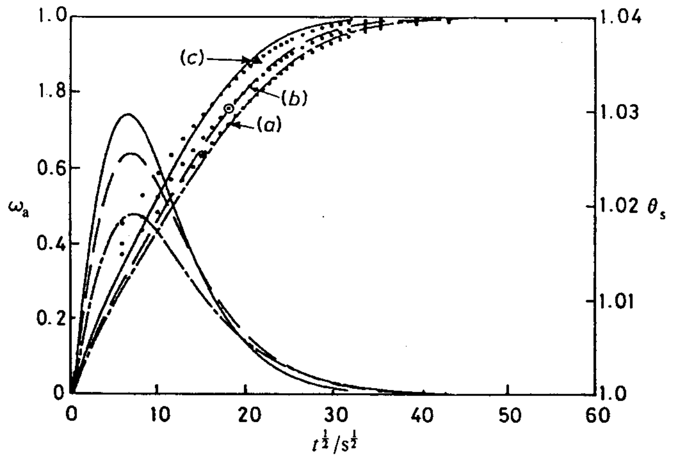

Fig. 6. Experimental (points) and calculated (lines) uptake ω_a and surface temperature θ_s for $R_p = 1$ mm, $T_0 = 50$ °C. Δp/mmHg = (*a*) 1–3, (*b*) 3–7 and (*c*) 7–13.

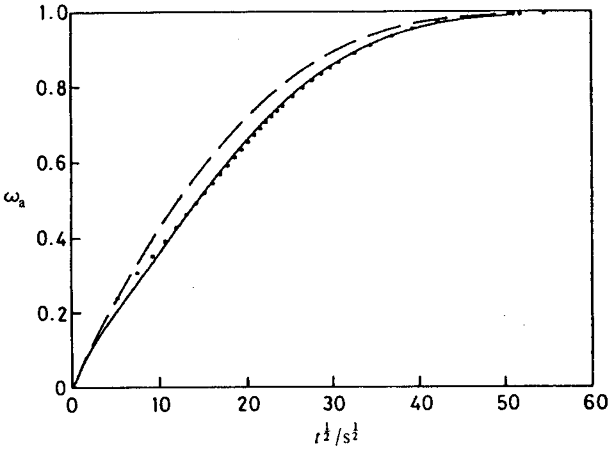

Fig. 7. Influence of heat transfer on the uptake history ($R_p = 1$ mm, $T_0 = 15$ °C, $\Delta p = 1$–2 mmHg): (·····) experimental, (——) theory with $h = 6.37$ and (– – – –) theory with $h = \infty$.

Discrepancies between experimental and calculated temperature curves are observed in fig. 3 and 4; a possible explanation is that the temperature is actually measured between the grains, whereas the computed values correspond to their surface.

Non-isothermal effects are illustrated in fig. 7, where a non-isothermal uptake curve is compared with its corresponding isothermal curve ($h = \infty$). The two curves do have some differences, but these are not notable. Consequently, in this particular case adsorption can be roughly considered as isothermal, although temperature variations are observed during sorption.

Glossary

a_q	thermal diffusivity	$m^2\ s^{-1}$
B	mobility	s
Bi	Biot number defined by eqn (16)	
c_p	gas concentration in the macropores	$kg\ m^{-3}$
c_s	heat capacity	$J\ kg^{-1}\ K^{-1}$
D	microporous diffusivity	$m^2\ s^{-1}$
D_0	corrected diffusivity defined by eqn (5)	$m^2\ s^{-1}$
E_0	characteristic constant of microporous systems	$J\ kg^{-1}$
h	overall heat transfer coefficient	$W\ m^{-2}\ K^{-1}$
J_m	mass flow density	$kg\ m^{-2}\ s^{-1}$
K_0	dimensionless parameter defined by eqn (16)	
Le	Lewis number [see eqn (16)]	
n	number of microparticles per unit pellet volume	m^{-3}
p	pressure	Pa
P_{n1}	dimensionless parameter defined by eqn (16)	
P_s	saturated pressure	Pa
q	amount adsorbed in the micropores	$kg\ m^{-3}$
r	radial coordinate	m
$\mathbf{R}$	gas constant	$J\ kg^{-1}\ K^{-1}$
R	radius	m
s	Laplace transform variable	s^{-1}
S_m^n	nth moment of the normalized adsorbed mass $(1-\omega_a)$	s
S_T^n	nth moment of the normalized surface temperature (θ_s-1)	s
t	time	s
T	temperature	K
W	volume of adsorbate condensed in the micropores	$m^3\ kg^{-1}$
W_0	total volume of the micropores accessible to the vapour	$m^3\ kg^{-1}$
α	thermal expansion coefficient	K^{-1}
β	constant in the equation of adsorption isotherms	
ε_p	pellet porosity	
η	normalized radial coordinate at the pellet's scale	
θ	normalized temperature	
θ_s	normalized surface temperature	
λ	effective thermal conductivity	$W\ K^{-1}\ m^{-1}$
μ	chemical potential	$J\ kg^{-1}$
ρ	apparent density of the pellet	$kg\ m^{-3}$
τ	normalized time	
ψ	normalized radial coordinate at the microparticles' scale	
ω	normalized adsorbate concentration	
ω_a	averaged normalized adsorbate concentration	
ΔH	heat of adsorption	$J\ kg^{-1}$
Δp	pressure step	Pa

subscripts

0	initial state
∞	final state
i	microparticle scale
p	pellet scale

Superscript * denotes the reference state. A tilde over a variable denotes the Laplace transform of the variable.

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