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Contribution of Concentration-Dependent Surface Diffusion in Ternary Adsorption Kinetics of Ethane, Propane and *n*-Butane in Activated Carbon

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Abstract. The role of concentration-dependent surface diffusion in the adsorption kinetics of a multicomponent system is investigated in this paper. Ethane, propane and *n*-butane are selected as the model adsorbates and Ajax activated carbon as the model adsorbent. Adsorption equilibrium isotherm and dynamic parameters extracted from single-component systems are used to predict the ternary adsorption equilibria and kinetics. The effect of concentration-dependent surface diffusion on the adsorption kinetics predictions is studied by comparing the results of two mathematical models with the experimental data. Three diffusion mechanisms, macropore, surface and micropore diffusions are incorporated in both models. The distinction between these two models is the use of the chemical potential gradient as the driving force for the diffusion of the adsorbed species in one model and the concentration gradient in the other. It was found that the model using the chemical potential gradient provides a better prediction of the ternary adsorption kinetics data, suggesting the importance of the concentration dependency of the surface diffusion, which is implicitly reflected in the chemical potential gradient. The kinetic model predictions are also affected by the way how single-component adsorption equilibrium isotherm data are fitted.

Keywords:

Introduction

Although the concentration-dependent surface diffusion has been widely recognized for adsorption processes of gases in porous media for single-component systems (Sladek et al., 1974; Seidel and Carl, 1989; Kapoor and Yang, 1991) and recently studied for binary kinetics (Karger and Bulow, 1975; Krishna, 1990; Hu and Do, 1992, 1993), its effect on a ternary adsorption rate process is rarely shown in the literature, probably due to both the complexity of the model equations and the procedure in obtaining experimental data. In this paper, experimental ternary adsorption dynamics data of ethane, propane and *n*-butane in Ajax activated carbon are collected using a differential adsorption bed, which has been proved in our laboratory to be a very powerful tool in obtaining multicomponent sorption data. The contribution of concentration-dependent adsorbed-species diffusion is studied by comparing two mathematical models with the obtained experimental data. One of the models assumes constant diffusivities, while the other requires the chemical potential gradient as the driving force for the diffusion of adsorbed species so that the concentration dependency of the surface and micropore diffusivities can be accounted for.

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Theory

The macropore, surface and micropore diffusion (MSMD) model recently proposed by Hu and Do (1993) is used in this paper to describe the adsorption dynamics of ethane, propane and n-butane in Ajax activated carbon. In this model the activated carbon particle is assumed to be bidispersed in pore structure with the free species diffusing along the macropores and the adsorbed species diffusing in the micropores and along the particle coordinate. The driving force for the diffusion of the adsorbed species is their chemical potential gradients. Because of the high flow rate used in experiments, there are two benefits resulting from this. The bulk concentrations surrounding the particle are constant and the particle system is isothermal due to the quick dissipation of adsorption heat. The MSMD model equations are restated below for completeness.

Because the particle structure is bidispersed, there are networks of macropores and micropores. Separated by macropores are small grains (microparticles) which accommodate the micropores. If C_μ denotes the adsorbed phase concentration inside the microparticle, r_μ the microparticle coordinate with a maximum of R_μ, when the driving force for the diffusion of adsorbed species is the chemical potential gradient, the diffusion flux of adsorbed species along the microparticle coordinate can be expressed as follows:

$$[J_\mu(k)]_{r_\mu} = -L(k)C_\mu(k)\frac{\partial \mu(k)}{\partial r_\mu} \tag{1}$$

where $L(k)$ and $\mu(k)$ are the mobility constant and chemical potential of species k.

However, if the concentration gradient is assumed to be the driving force for the diffusion of adsorbed species, its diffusion flux is determined as:

$$[J_\mu(k)]_{r_\mu} = -D_\mu^*(k)\frac{\partial C_\mu(k)}{\partial r_\mu} \tag{2}$$

where D_μ^* is the adsorbed species diffusivity in the micropore and assumed constant.

In Eq. (1) it is required to calculate the chemical potential of the adsorbed phase. This chemical potential is the same as that of the gas phase if both phases are in local equilibrium. However, this is not true inside the microparticle because in the presence of micropore diffusion resistance, the adsorbed concentration inside the microparticle is not in equilibrium with the local gas phase environment surrounding that microparticle. To overcome this difficulty we introduce an imaginary gas phase where the gas species are in equilibrium with the adsorbed species inside the microparticle. Hence, the chemical potential in the adsorbed phase can be calculated as that of the imaginary gas phase. Now we can write the chemical potential as:

$$\mu(k) = \mu^\circ(k) + R_g T \ln[C_{\text{im}}(k)] \tag{3}$$

where R_g is the gas constant and T is temperature in Kelvin, C_{im} is the imaginary gas phase concentration, which is reversely calculated via the equilibrium equation from the adsorbed phase concentration inside the microparticle. Therefore, the diffusion flux of adsorbed species along the microparticle coordinate can be written in terms of the imaginary concentration gradient:

$$\begin{aligned}[J_\mu(k)]_{r_\mu} &= -D_{\mu 0}^*(k)\frac{C_\mu(k)}{C_{\text{im}}(k)}\frac{\partial C_{\text{im}}(k)}{\partial r_\mu};\\ D_{\mu 0}^*(k) &= L(k)R_g T\end{aligned} \tag{4}$$

where $D_{\mu 0}^*$ is the zero coverage diffusivity of the adsorbed species along the microparticle direction.

Similarly, by using the notion of imaginary gas phase concentrations, the diffusion flux of the adsorbed species along the particle coordinate can be expressed as:

$$[J_\mu(k)]_r = -D_{\mu 0}(k)\frac{C_\mu(k)}{C_{\text{im}}(k)}\frac{\partial C_{\text{im}}(k)}{\partial r} \tag{5}$$

Because the tortuosity might be different in different directions, the zero coverage diffusivity of the adsorbed species along the particle direction, $D_{\mu 0}$, may be different from that along the microparticle coordinate, $D_{\mu 0}^*$, by a factor of β^2, i.e.,

$$D_{\mu 0}^*(k) = \beta^2(k)D_{\mu 0}(k) \tag{6}$$

When the concentration gradient is assumed the driving force, the diffusion flux of the adsorbed species along the particle coordinate is:

$$[J_\mu(k)]_r = -D_\mu(k)\frac{\partial C_\mu(k)}{\partial r} \tag{7}$$

and D_μ also differs D_μ^* by a factor of β^2.

The microparticle is usually assumed to be spherical although other geometries such as slab or cylinder can also be used. The mass balance equation of species k

in a spherical microparticle is:

$$\frac{\partial C_\mu(k)}{\partial t} = -\frac{1}{r_\mu^2}\frac{\partial}{\partial r_\mu}\{r_\mu^2[J_\mu(k)]_{r_\mu}\} \tag{8}$$

One boundary condition for this mass balance equation is the symmetry condition at the center of the microparticle:

$$r_\mu = 0; \qquad \frac{\partial C_\mu(k)}{\partial r_\mu} = 0 \tag{9}$$

The other boundary condition is at the exterior surface of the microsphere, where the adsorbed species is in local equilibrium with the gas species in the macropore. The imaginary and macropore concentrations are the same at this boundary.

$$\begin{aligned} &r_\mu = R_\mu; \\ &C_\mu(k) = f_k[C_p(1), C_p(2), \ldots, C_p(\mathrm{NC})] \end{aligned} \tag{10}$$

where f is the multicomponent equilibrium function, such as the ideal adsorbed solution theory (IAST).

The particle can have a geometry of slab, cylinder or sphere, characterized by a parameter s of 0, 1 or 2, respectively. Let r be the particle coordinate with a length scale of R, ε_M the macropore porosity, the mass balance equation for species k in the particle is:

$$\begin{aligned} &\varepsilon_M\frac{\partial C_p(k)}{\partial t} + (1-\varepsilon_M)\frac{\int_0^{R_\mu}\left[\frac{\partial C_\mu(k)}{\partial t}\right]r_\mu^2 dr_\mu}{\int_0^{R_\mu} r_\mu^2 dr_\mu} \\ &\quad = -\varepsilon_M\frac{1}{r^s}\frac{\partial}{\partial r}\left[r^s J_p(k)\right] - (1-\varepsilon_M)\frac{1}{r^s}\frac{\partial}{\partial r} \\ &\qquad \times\left[r^s\frac{\int_0^{R_\mu}[J_\mu(k)]_r r_\mu^2 dr_\mu}{\int_0^{R_\mu} r_\mu^2 dr_\mu}\right] \end{aligned} \tag{11}$$

where $J_p(k)$ is the molar flux of species k in the pore phase and defined as:

$$J_p(k) = -D_p(k)\frac{\partial C_p(k)}{\partial r} \tag{12}$$

with D_p being the pore diffusivity.

The corresponding boundary conditions for Eq. (11) are:

$$\begin{aligned} &r = 0; \\ &\varepsilon_M J_p(k) + (1-\varepsilon_M)\frac{\int_0^{R_\mu}[J_\mu(k)]_r r_\mu^2 dr_\mu}{\int_0^{R_\mu} r_\mu^2 dr_\mu} = 0 \end{aligned} \tag{13}$$

$$\begin{aligned} &r = R; \\ &\varepsilon_M J_p(k) + (1-\varepsilon_M)\frac{\int_0^{R_\mu}[J_\mu(k)]_r r_\mu^2 dr_\mu}{\int_0^{R_\mu} r_\mu^2 dr_\mu} \\ &\quad = k_m[C_p(k) - C_b(k)] \end{aligned} \tag{14}$$

where k_m is the film mass transfer coefficient and C_b is the gas concentration in the bulk phase.

The initial condition of the system is:

$$t = 0; \quad C_p(k) = C_{pi}(k); \quad C_\mu(k) = C_{\mu i}(k) \tag{15}$$

Equations (8) to (15) represent a model describing the multicomponent sorption kinetics of gases in activated carbon. All three diffusion mechanisms, macropore, surface and micropore, are taken into account in the model. If a chemical potential gradient is used in the calculation of adsorbed phase diffusion (Eqs. (4) and (5)), the model is called MSMD model which can explain the concentration dependency of the adsorbed species diffusivities. While if a concentration gradient instead of a chemical potential gradient is utilized in the expression of adsorbed phase diffusion (Eqs. (2) and (7)), the model will have a constant adsorbed phase diffusivity and therefore is called CMSMD model. When an extended multicomponent Langmuir equation instead of IAST is used, the CMSMD is the same as that of Mayfield (1990). The model equations are cast into non-dimensional forms (Hu and Do, 1993) and then solved by using an orthogonal collocation method (to transform the PDEs to ODEs) and a standard ODE integration package. The detailed computation procedures, including the implicit multicomponent equilibria of IAST, are available in Hu and Do (1992, 1993).

In this article an isotherm equation proposed by O'Brien and Myers (1985) is used to describe the single-component adsorption equilibrium isotherm:

$$C_\mu = C_{\mu s}\left[\frac{bC_p}{1+bC_p} + \frac{\sigma^2 bC_p(1-bC_p)}{2(1+bC_p)^3}\right] \tag{16}$$

The reason for this isotherm will be discussed later.

Results and Discussions

In this paper, ethane, propane and n-butane were chosen as the adsorbates and Ajax activated carbon as the adsorbent particle. The single component adsorption equilibrium isotherm was measured by using a volumetric technique and the experimental data were fitted

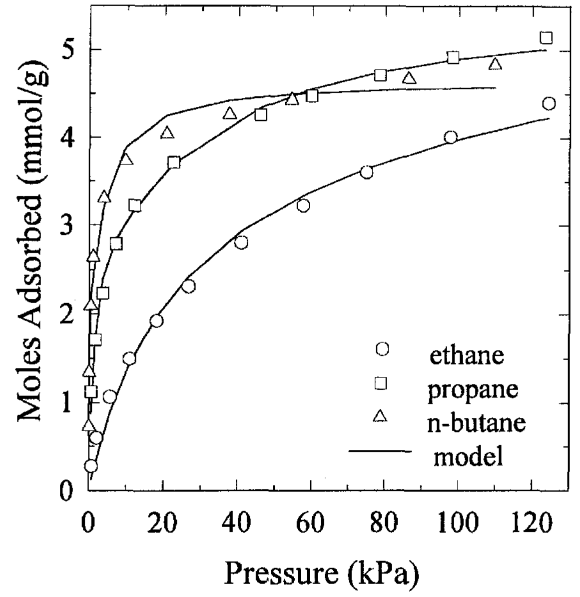

Figure 1. Adsorption equilibrium isotherm of ethane, propane and *n*-butane in Ajax activated carbon at 30°C.

to the O'Brien-Myers equation. Figure 1 shows the adsorption equilibrium of ethane, propane and *n*-butane in Ajax activated carbon at 30°C. *n*-butane has the highest adsorption affinity, followed by propane, with ethane as the weakest adsorbate. The O'Brien-Myers equation is in good agreement with the experimental data. The extracted isotherm parameters for the O'Brien-Myers equation were tabulated in Table 1.

In order to predict the multicomponent adsorption kinetics, single-component information of both adsorption equilibrium and mass transfer is required. Figure 2 shows the single-component adsorption kinetics of ethane, propane and *n*-butane in Ajax activated carbon at 30°C and 1 atm. The experimental data were collected using a differential adsorption bed (DAB). Three bulk concentrations and three particle sizes (shapes) were conducted for each species. These data were then used to extract the dynamic parameters of MSMD and CMSMD models. The macropore diffusivity was calculated from the combined molecular and Knudsen diffusivities and a tortuosity of three (Hu et al., 1993). Therefore, only the adsorbed species diffusivity (D_μ) and the effective diffusion length in the microparticle (R_μ/β) need to be found from the data fitting. The model fits from the MSMD model were presented in Fig. 2 as solid lines and those from the CMSMD model as dashed lines. The dynamic parameters were listed in Table 2 for these two models. Both models are in good agreement with the experimental data, but the CMSMD model is required to use different values of surface diffusivities for different bulk concentrations in order to match the data, which is inconsistent to its assumption of constant surface diffusivity.

Table 1. Isotherm parameters of ethane, propane and *n*-butane in Ajax activated carbon at 30°C.

Species	$C_{\mu s}$ (kmol/m^3)	b (kPa^{-1})	σ
Ethane	5.92	0.0212	0.956
Propane	5.32	0.152	1.517
n-butane	4.45	1.290	1.778

Table 2. Dynamic parameters of ethane, propane and *n*-butane in Ajax activated carbon at 30°C.

Species	D_p (10^{-6} m^2/s)	$D_{\mu 0}$ (10^{-10} m^2/s) (MSMD)	D_μ (10^{-10} m^2/s) (CMSMD)	R_μ/β (10^{-4} m)
Ethane	4.48	13.96	16.32 (5%) 21.91 (10%) 19.39 (20%)	3.43
Propane	3.47	3.23	6.76 (5%) 8.77 (10%) 8.03 (20%)	3.63
n-butane	3.04	1.99	3.88 (2%) 8.36 (5%) 9.91 (10%)	6.00

Having obtained the single-component parameters of adsorption equilibrium and kinetics, we can now predict the ternary adsorption kinetics of ethane, propane and *n*-butane in Ajax activated carbon. The ternary equilibrium was calculated using the IAST and the single component isotherm equation. The kinetic prediction were obtained by using the MSMD or CMSMD model and the single-component parameters. In this article we used the O'Brien-Myers equation to describe the single-component adsorption equilibrium. The agreement between the isotherm model and the experimental data is good for the whole pressure range (Fig. 1). Other isotherms such as the Unilan equation can also be used to describe the single-component data; but the numerical integration of MSMD model could not converge for the ternary kinetics. This is the limitation of the MSMD model because it relies on the iterative IAST to compute the multicomponent adsorption equilibrium.

Figure 3 shows the ternary adsorption kinetics of ethane, propane and *n*-butane in Ajax activated carbon.

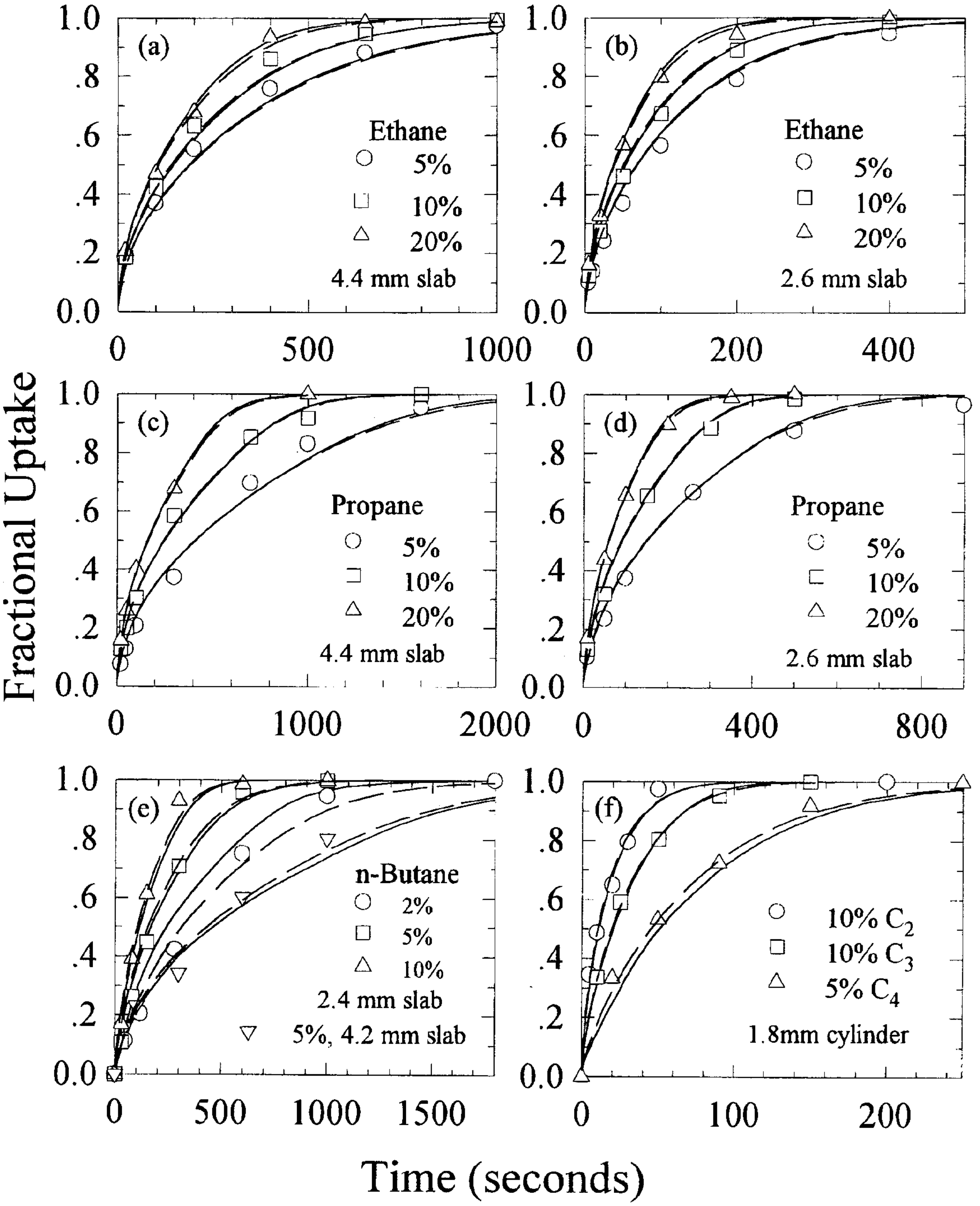

Figure 2. Adsorption kinetics of gases in Ajax activated carbon at 30°C, 1 atm (——) MSMD; (___) CMSMD.

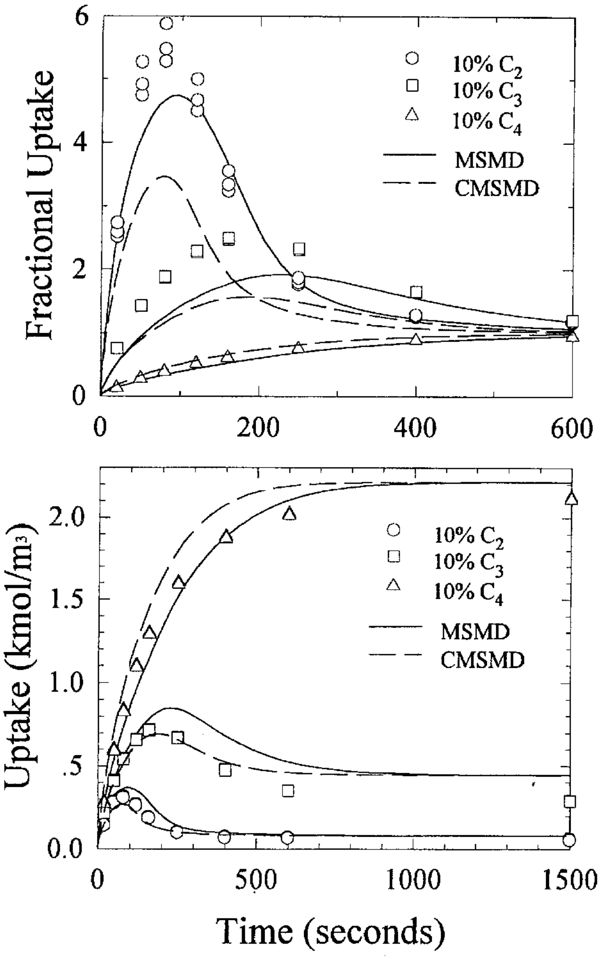

Figure 3. Ternary adsorption kinetics of gases in Ajax activated carbon of 2.4 mm full length slab at 30°C, 1 atm.

The bulk concentration of each adsorbate was 10% at a total pressure of 1 atm. The carbon particle had a slab geometry and a full length of 2.4 mm. The data were presented in the form of fractional uptake versus time as well as the amount adsorbed versus time. The fractional uptake was defined as the total amount in the particle (macropore + solid) divided by its value at final equilibrium. The fractional uptakes of ethane and propane are greater than unity during the course of adsorption, because ethane and propane diffuse faster than *n*-butane but they are less-strongly adsorbed and eventually displaced partially by the incoming *n*-butane. Both MSMD and CMSMD models reasonably predict the adsorption kinetics of ethane and *n*-butane. Since the adsorbed amount of ethane at final equilibrium is very small, a slightly overprediction on the ethane observed in the plot of the fractional uptake versus time by the MSMD model becomes negligible if the result is presented in the form of the amount adsorbed versus time. It is noted that the IAST overpredicts the adsorption amount of propane, which causes the underprediction on its fractional uptake. The CMSMD model at first glance seems to better predict the real uptake of propane, but this is based on the overprediction of the IAST on propane adsorbed amount. If this factor is corrected, the CMSMD will fail to predict the uptake of propane, just like the case of fractional uptake. Both MSMD and CMSMD models reasonably predict the (fractional) uptake of *n*-butane. However, the CMSMD model significantly underpredicts the overshoot degree of ethane fractional uptake.

The ternary adsorption kinetics of the same system as in Fig. 3 but having a different bulk concentration combination was shown in Fig. 4. The bulk concentration was 10% for ethane, 5% for both propane and *n*-butane.

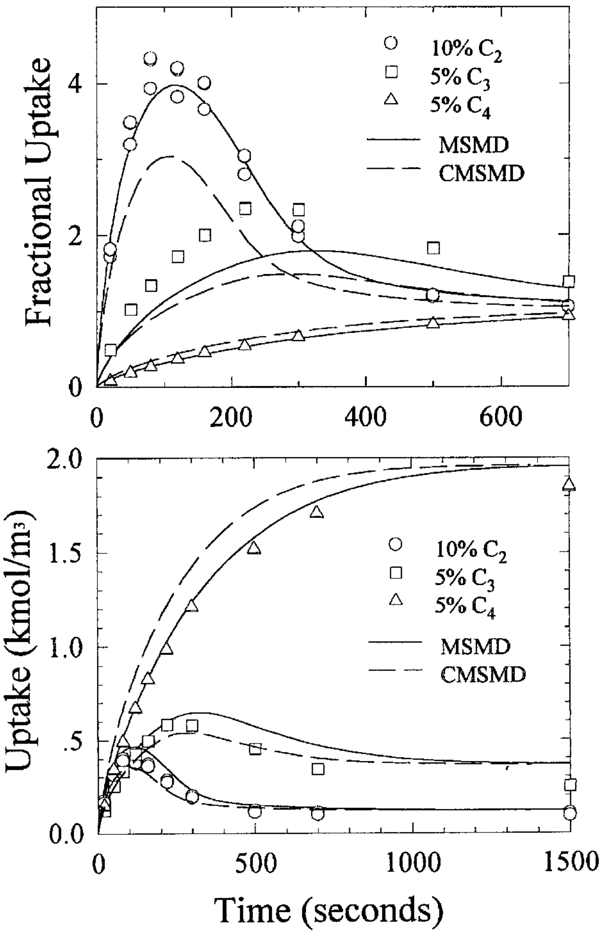

Figure 4. Ternary adsorption kinetics of gases in Ajax activated carbon of 2.4 mm full length slab at 30°C, 1 atm.

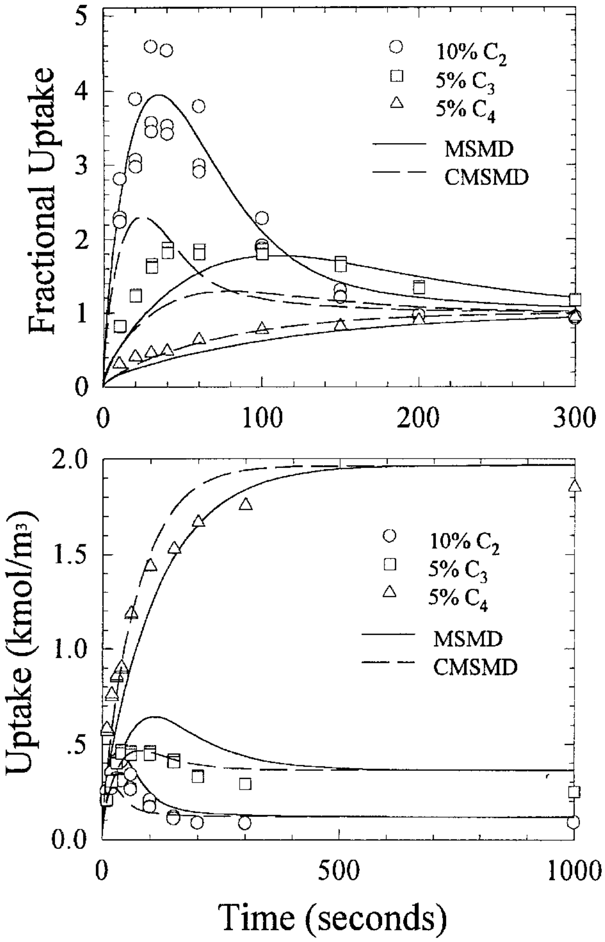

Figure 5. Ternary adsorption kinetics of gases in Ajax activated carbon of 1.8 mm diameter at 30°C, 1 atm.

The CMSMD model underpredicts the fractional uptakes of both ethane and propane and overpredicts the uptake of *n*-butane. Again the MSMD model is in good agreement with the experimental data, except for propane fractional uptake where some deviations are observed due to the incorrect prediction by IAST on the final adsorbed equilibrium concentration of propane.

The same adsorption system as in Fig. 4 but having different particle size and shape was plotted in Fig. 5. Here the Ajax activated carbon had a cylindrical shape and its diameter was 1.8 mm. Since the particle is smaller, the contribution from the micropore resistance is more significant. Therefore, the diffusion of adsorbed species is expected to play a more important role in this case, which is indeed reflected in Fig. 5 where a large difference, between the MSMD and CMSMD model predictions is observed. The CMSMD model significantly underpredicts the overshoot degrees of ethane and propane fractional uptakes. While the agreement between the MSMD model prediction and the experimental data is within experimental error.

In the above discussions we have seen that the ternary adsorption equilibrium plays an important role in the prediction of the fractional uptake overshoot degree of ethane and propane. To further study this effect, we refitted the isotherm equilibrium data of ethane by the same O'Brien-Myers equation but using only the experimental data having pressure less than 30 kPa. The reason for this cut-off value was that the total pressure of hydrocarbons in our dynamic study was always less than 30 kPa. One may argue that the fitting is irrelevant beyond the total pressure of the system, provided a good fit is obtained for the pressure range considered. However, this is incorrect when the IAST is used to predict multicomponent adsorption equilibria, because the isotherm information at the higher pressure range is also required in the calculation of spreading pressure of pure component. Therefore, the isotherm model must be in good agreement with the experimental data for both low and high pressure regions in order to correctly predict the multicomponent adsorption equilibrium using IAST (Hu and Do, 1995). Now we further investigate this effect on the adsorption kinetics prediction. The result by fitting the O'Brien-Myers equation to the ethane isotherm data of less than 30 kPa was shown in Fig. 6 as a dashed line. The extracted

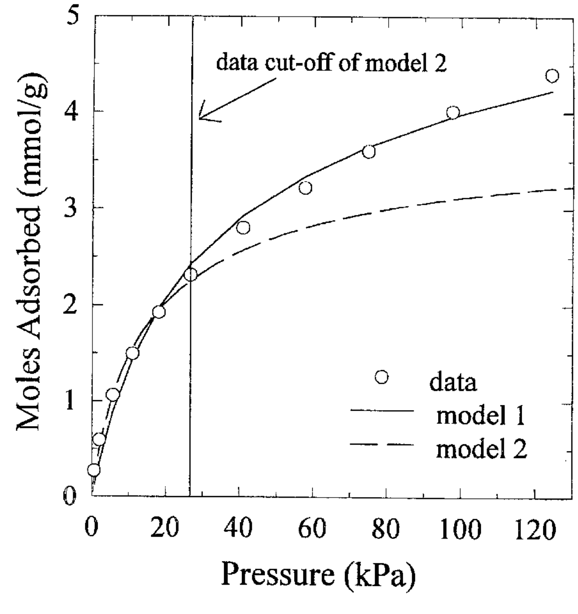

Figure 6. Adsorption equilibrium isotherm of ethane in Ajax activated carbon at 30°C. (——) O'Brien-Myres, fitting up to 41 kPa. (— — —) O'Brien-Myres, fitting up to 26 kPa.

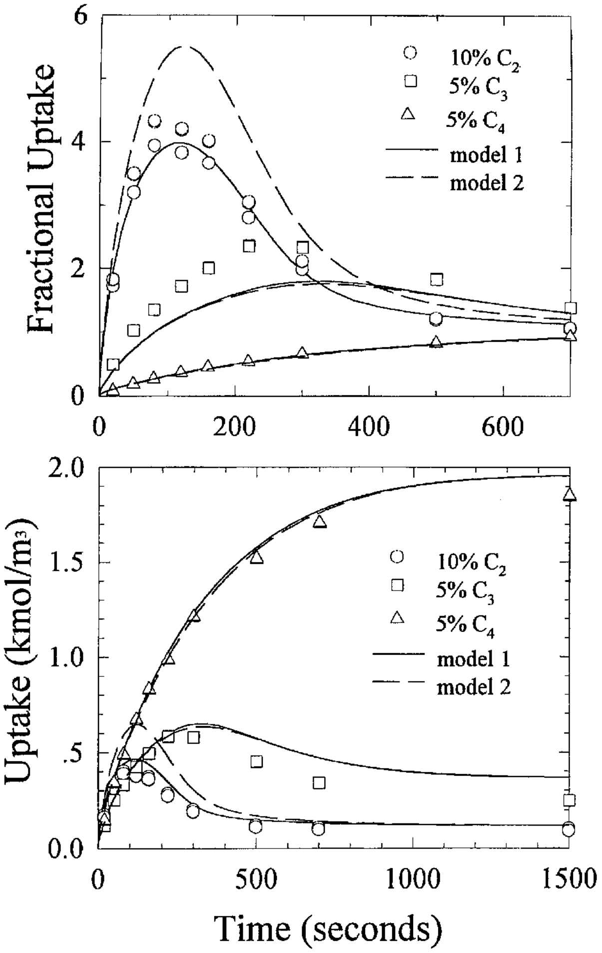

Figure 7. Ternary adsorption kinetics of gases in Ajax activated carbon of 2.4 mm full length slab at 30°C, 1 atm.

parameters were: $C_{\mu s} = 3.60$ kmol/m^3; $b = 0.0599$ kPa^{-1}; $\sigma = 0.809$. We called this model 2. Although this fitting method gives a better fit in the low pressure range, it significantly differs from the experimental data when extrapolated to the high pressure region. What was called model 1 in Fig. 6 (solid line) was referred to the result in Fig. 1, i.e., all data points were used in the model fitting. These new isotherm parameters were then used in the single-component adsorption kinetics fitting of ethane in Ajax activated carbon using the MSMD model. The pore diffusivity and the effective diffusion length in the microparticle were kept the same as in model 1. The zero coverage surface diffusivity was then found to be 5.57×10^{-10} m^2/s, a value smaller than that in model 1. The goodness of the model fits was similar to that of model 1 which is shown in Fig. 2.

The new isotherm and dynamic parameters of ethane were then used to predict the ternary adsorption kinetics of 10% ethane, 5% propane and 5% *n*-butane in Ajax activated carbon of 2.4 mm full length slab, the same system as in Fig. 4. The isotherm and dynamic parameters of propane and *n*-butane were kept unchanged. The result was plotted in Fig. 7 as dashed lines, compared with those predictions from model 1 as solid lines. It is obvious that the fitting method using only ethane isotherm data of less than 30 kPa (model 2) significantly overpredicts the overshoot degree of both fractional and real uptakes of ethane. Since the affinities of propane and *n*-butane are stronger than that of ethane, their uptake is little affected by the choice of ethane parameters, either in dimensional or non-dimensional form. Therefore one needs to be very careful in the choice of the isotherm parameters for the weakest species in the mutlicomponent adsorption kinetics calculations.

Conclusions

Experimental data were used to study the effect of concentration-dependent surface diffusion on ternary adsorption kinetics of ethane, propane and *n*-butane in Ajax activated carbon. Although a model assuming a constant surface diffusivity can fit the single-component kinetics data well, it requires different values of surface diffusivities when different bulk concentrations are used. This is obviously in conflict with its assumption of constant surface diffusivity. The CMSMD model also fails to correctly predict the fractional uptake overshoot degree of ethane and propane in ternary adsorption kinetics in Ajax activated carbon. On the other hand, the model allowing the concentration-dependent surface diffusion (MSMD) can adequately fit the single-component adsorption dynamics with a unique surface diffusivity. The agreement between the MSMD model and the experimental data of ternary adsorption kinetics is generally good. However, there are difficulties for MSMD model to numerically converge in the ternary kinetics when a Unilan equation is used in the IAST to calculate the ternary adsorption equilibrium. It is also noted that predictions on the fractional uptake overshoot degree of ethane in the ternary adsorption kinetics is strongly affected by the choice of ethane equilibrium parameters.

Nomenclature

CMSMD	Constant surface diffusivity macropore,

	surface and micropore diffusion	
C_b	Adsorbate concentration in the bulk	kmol/m^3
C_{im}	Imaginary adsorbate concentration inside the microparticle	kmol/m^3
C_p	Adsorbate concentration in the macropore	kmol/m^3
C_{pi}	Initial adsorbate concentration in the macropore	kmol/m^3
C_μ	Adsorbed concentration in the particle	kmol/m^3
$C_{\mu i}$	Initial adsorbed concentration in the particle	kmol/m^3
D_p	Macropore diffusivity	m^2/s
$D_{\mu 0}$	Diffusivity of adsorbed species at zero coverage in the particle coordinate	m^2/s
$D^*_{\mu 0}$	Diffusivity of adsorbed species at zero coverage in the microparticle coordinate	m^2/s
f	Multicomponent equilibrium isotherm function	
IAST	Ideal adsorbed solution theory	
J_p	Flux through the macropore	kmol/m^2/s
J_μ	Flux through the solid	kmol/m^2/s
L	Mobility constant	kmol/m^2/kJ/s
MSMD	Macropore, surface and micropore diffusion	
NC	Number of component	
ODE	Ordinary differential equation	
PDE	Partial differential equation	
r	Particle radial position	m
r_μ	Microparticle coordinate	m
R	Particle radius	m
R_μ	Diffusion path length of microparticle	m
s	Geometric factor of particle	
t	Real time	sec

Greek Symbol

β^2	Ratio of zero coverage surface diffusivity in the microparticle coordinate to that in the particle coordinate	
ε_M	Particle macropore porosity	
μ	Chemical potential	kJ/kmol
μ°	Chemical potential at standard state	kJ/kmol

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