# Capillary Transport in Adsorption from Liquid Phase on Activated Carbon

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*Two models presented here predict the kinetics of adsorption from solution. The adsorbate is considered to form a capillary separated phase inside the micropores. The transport in the adsorbed phase is modeled as capillary transport under the influences of gradients in capillary pressure. In model I the capillary separated phase is assumed to be continuous, while in model II it is assumed to be discontinuous. The effective adsorbed phase diffusion coefficient for this transport is directly related to the pore-size distribution, which can be found from the sorption isotherm. In both models the overall apparent diffusion coefficients are compared with experimental data. Both models satisfactorily predict the dependency of the diffusion coefficient on carbon loading. Model I slightly underestimates the temperature dependency of the effective diffusion coefficient, while model II gives a good prediction of the influence of temperature on the rate of adsorption.*

## Introduction

Adsorption of organic compounds from an aqueous solution on activated carbon is a separation process widely used in industry and environmental technology. The descriptions of adsorption kinetics for activated carbon are considerably less developed than are those for microcrystalline solids like zeolites (e.g., Kärger and Ruthven, 1992).

In general, the transport of adsorbate can be described by means of an effective diffusivity, $D'_a$, defined by the Fickian equation:

$$J_a = -D'_a \rho_{app} \frac{\partial Q}{\partial x}, \qquad (1)$$

where $J_a$ is the adsorbate flux, $\rho_{app}$ the apparent density of the adsorbent, $Q$ the carbon loading, and $x$ the position coordinate. The effective diffusion coefficient will, in general, depend on the carbon structure, loading, and temperature.

The transport of some components in activated carbon takes place at a much higher rate than predicted by molecular diffusion transport. The adsorption of nitrobenzene from an aqueous solution is often reported in this context (van Lier, 1989; Kouyoumdjiev, 1992; Miyahara and Okazaki, 1993). An often proposed mechanism for this is the transport by surface diffusion, parallel to the molecular diffusion inside the (macro)pore liquid. Surface diffusion is imagined as the hopping or sliding of molecules along the pore walls. Local equilibrium between pore solution and adsorbed phase is assumed. This is illustrated in Figure 1. Formally, we can write Eq. 1 for this parallel transport as:

$$J = J_w + J_a = -(D'_w + D'_a)\rho_{app}\frac{\partial Q}{\partial x} \qquad (2)$$

in which the subscripts $w$ and $a$ denote the water (solution) phase and adsorbed phase, respectively. For the contribution of the aqueous-phase diffusion in the larger pores, we can write

$$J_w = -\frac{D_w \epsilon}{\tau}\left(1-\frac{V_a}{V_p}\right)\frac{\partial C}{\partial x} = -\frac{D_w \epsilon}{\tau}\left(1-\frac{V_a}{V_p}\right)\frac{\partial Q}{\partial x}\bigg/\frac{\partial Q}{\partial C}, \qquad (3)$$

and so we obtain:

$$D'_w = \frac{D_w \epsilon}{\tau}\left(1-\frac{V_a}{V_p}\right)\bigg/\left(\rho_{app}\frac{\partial Q}{\partial C}\right), \qquad (4)$$

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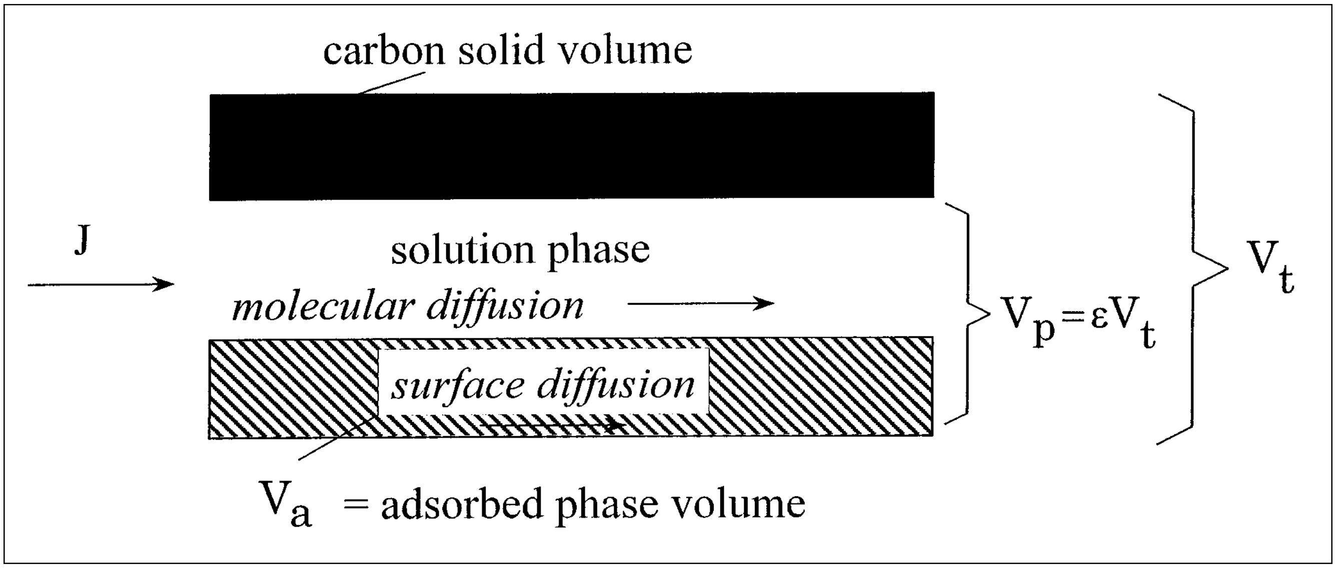

**Figure 1. A model for parallel surface and molecular diffusion.**

where $C$ is the adsorbate concentration in the water phase, $D_w$ is the diffusion coefficient of the adsorbate in water, $\tau$ is the tortuosity, $V_p$ is the total pore volume, $V_a$ is the pore volume filled with adsorbate, and $\epsilon$ is the porosity. The term $\epsilon(1 - V_a/V_p)$ is the fraction of the volume that is available for transport by molecular diffusion. For a known porosity $\epsilon$, this term and the derivative $\partial Q/\partial C$ follow from the sorption isotherm.

In Figure 2 we compare the value of $D'_w$ with the experimental data of Miyahara and Okazaki for the effective diffusion coefficient of nitrobenzene in Takeda-X-7000-activated carbon at three temperatures. We estimated the diffusion coefficient $D_w$ of nitrobenzene in water from the experimental value of $9.25\times10^{-10}$ m²/s at 21.5°C (Kouyoumdjiev, 1992) and the Stokes–Einstein relation; the results are given in Table 1. Even for the minimal tortuosity factor $\tau = 1$ we see that the contribution of the aqueous-phase diffusion is very low.

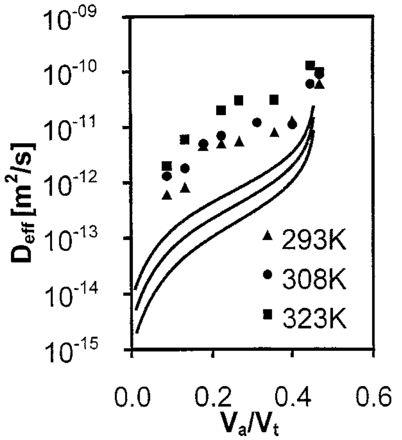

**Figure 2. Dependency of the diffusivity of nitrobenzene on volume fraction filled with nitrobenzene at different temperatures.**

Comparison of the experimental data of Miyahara and Okazaki (1993) with $D'_w$, the contribution of molecular diffusion in the aqueous phase. Assume that minimum tortuosity $\tau = 1$.

Various models have been presented in the literature for the surface diffusion in the adsorbed phase. Van Lier (1989) and Miyahara and Okazaki took a Fickian type of equation like Eq. 1, in which the gradient in loading is the driving force for surface diffusion. In our laboratory we developed a Maxwell–Stefan model, in which the driving force for surface diffusion is the gradient in chemical potential (Kouyoumdjiev, 1992), starting from the model of Krishna (1990).

Although the results macroscopically can agree reasonably well with experimental data, the problem that the diffusion coefficients found depend on external concentration, particle size, and so on, often arises (van Lier, 1989), indicating that the modeling is incomplete. Also, from the physical point of view, the parallel model has a disadvantage: hardly any sur-

**Table 1. Physical Data of Nitrobenzene and Benzonitrile as a Function of Temperature**

| $T$ [K] | Solubility [kg/m³] | Viscosity [mPa·s] | Density [kg/m³] | Interfacial Tension [N·m$^{-1}$] | Diffusion Coefficient in Water* $10^{-9}$m²/s |
|---|---|---|---|---|---|
| *Nitrobenzene* | | | | | |
| 293 | 1.84 | 2.06 | 1200 | 0.0257 | 0.89 |
| 308 | 2.2 | 1.6 | 1190 | — | 1.29 |
| 323 | 2.6 | 1.27 | 1170 | — | 1.81 |
| *Benzonitrile* | | | | | |
| 293 | 4.0 | 1.36 | 1000 | 0.028 | 0.89 |
| 308 | 4.8 | 1.09 | 990 | — | 1.29 |

*Source:* Miyahara and Okazaki, 1993; Lide, 1996.

*For nitrobenzene derived from Kouyoumdjiev and the application of the Stokes–Einstein relation. The same values were taken for benzonitrile as for nitrobenzene.

face is present in the macropores, while hardly any liquid diffusion can be expected in the micropores.

More extended models differentiate between macropore and microparticle diffusion, which lead to coupled models on the particle scale (Ma and Lee, 1976; van Lier, 1989). Recent overviews have been given by Yang (1987) and Kärger and Ruthven (1992). Van Lier used the biporous model to explain the influence of the initial concentration of the adsorbate and the carbon dose on adsorption kinetics, which cannot be explained with homogeneous models. The biporous model, however, contains parameters that can hardly be obtained independently by experiments.

In their study of the transport of gases Tamon et al. (1981) showed that, for the region in which capillary condensation occurs, the diffusion in the adsorbed phase could well be described by the additivity of surface hopping and capillary flow. Rajniak and Yang (1996) used a network model for predicting effective Fickian diffusivities of condensable vapors in porous media where capillary condensation and adsorption/desorption hysteresis occur. The network Rajniak and Yang used was a Bethe Tree. Using a combination of surface diffusion and capillary transport caused by a gradient of capillary pressure as transport mechanisms, they were able to successfully predict the concentration dependency of the effective diffusivity in the whole range of relative pressures for different adsorbent/adsorbate systems. For drying of porous materials van der Zanden et al. (1996) developed a model for the partially saturated region in which capillary flow was the mechanism for liquid water transport. Their results agreed well with the experimental data on the effective water diffusion coefficient in clay as a function of water content. They explicitly formulated the effective diffusivity in terms of the pore-size distribution.

For the adsorption equilibrium between activated carbon and aqueous solutions, a theory based on capillary phase separation was recently presented by Miyahara and Okazaki (1992, 1993, 1994) and Okazaki (1993). This can be considered an analogy of the capillary condensation for adsorption from the vapor phase such as that developed by Okazaki et al. (1978) for the study of binary equilibria between vapor and activated carbon. The basic phenomenon was described decades earlier by Patrick and Jones (1925) and Patrick and Ebermann (1925). As shown by Miyahara and Okazaki (1992, 1993, 1994), according to this concept, the adsorption equilibrium isotherms for various organic compounds between water and activated carbon could be described well with the theory.

For the case of adsorption from the liquid phase, the transport of capillary separated phase under the influence of a gradient in capillary pressure has to our knowledge not been investigated in the literature. Since the equilibrium descriptions based on this model are promising, it was thought worthwhile to also investigate a transport model based on this concept. In the following we will present two capillary flow models that lead to expressions for the effective diffusion coefficient, along the lines of van der Zanden et al. (1996). In the first model a flow of capillary separated adsorbate phase influenced by a gradient in capillary pressure is assumed, assuming the possibility of continuous percolation throughout the particle. In the second model capillary flow is assumed to occur in small regions, in series with solution-phase molecular diffusion in larger pores. Predictions of the effective diffusivities as a function of concentration and temperature are compared to experimental data from the literature (Miyahara and Okazaki, 1992, 1993).

## Theory

### *Model I: Continuous capillary transport*

The system considered here is shown in Figure 3. The adsorbed phase is thought to be present as a separate phase in a fraction of the pores, separated from the aqueous solution

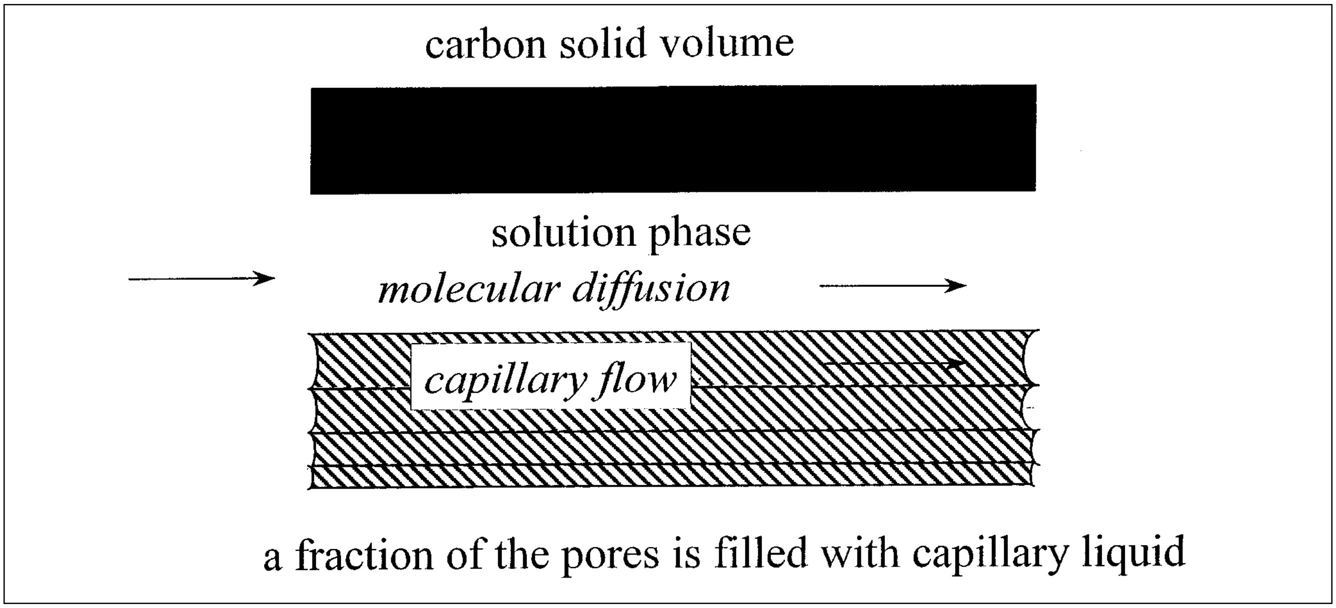

**Figure 3. Parallel capillary flow in the adsorbate phase and molecular diffusion in the aqueous phase, for continuous capillary flow (model I).**

by a curved interface. At a given concentration $C$ in the aqueous phase the equilibrium meniscus radius $r^*$ is given by the Kelvin equation (Miyahara and Okazaki, 1994):

$$RT \ln \frac{C_s}{C} = \frac{2\sigma_w M_a \cos \vartheta}{\rho_a r^*}, \tag{5}$$

where $C_s$ is the adsorbate solubility, $\rho_a$ is the adsorbate density, $M_a$ is the molar mass of the adsorbate, $\sigma_w$ is the interfacial tension at the water/adsorbate interface, and $\theta$ is the contact angle of the adsorbate with the adsorbent. This means that approximately at the given activity $C/C_s$ the pores with radius $r_m = r^*$ will be filled, as well as the smaller pores, of which the meniscus will have a smaller radius of curvature. For the load onto the carbon, we can write:

$$\frac{V_a}{V_p} = \int_0^{r_m} f(r)\, dr, \tag{6}$$

with

$$\frac{V_a}{V_p} = \frac{\rho_{app}}{\epsilon \rho_a} Q, \tag{7}$$

and $f(r)$ is the volumetric pore-size distribution, normalized at the total pore volume. The pore-size distribution is calculated from the experimental sorption isotherm using Eqs. 5 and 6.

As stated before, the capillary separated adsorbate phase is assumed to flow under the influence of a gradient in capillary pressure. Pressure gradients in the aqueous phase, caused by viscous forces, are neglected because the adsorbate flow takes place in much smaller pores than the aqueous flow. Another assumption is that the pore structure consists of parallel cylindrical pores with a different diameter. Although the properties of the adsorbate, such as viscosity, surface tension, and density, will change close to the pore walls, in this approximate model we will not take such effects into account.

Darcy's law describes the superficial velocity $u_c$ of the capillary separated adsorbate through a porous material under a pressure gradient $dP/dx$ for a Newtonian fluid (Quintard and Todorovic, 1992)

$$u_c = -\frac{k_r k_\infty}{\mu_a} \frac{\partial P}{\partial x}, \tag{8}$$

where $P$ is the pressure in the adsorbate phase. The expression $k_r k_\infty/\mu_a$, in which $k_r k_\infty$ is the effective permeability and $\mu_a$ the adsorbate viscosity, may be regarded as the reciprocal of the resistance against transport of the capillary separated phase.

For the flux of adsorbate per unit adsorbent cross section we then have the capillary flow contribution as

$$J_a = -D'_a \rho_{app} \frac{\partial Q}{\partial x} = \rho_a u_c = -\rho_a \frac{k_r k_\infty}{\mu_a} \frac{\partial P}{\partial x}, \tag{9}$$

from which

$$D'_a = \frac{1}{\epsilon} \frac{k_r k_\infty}{\mu_a} \frac{\partial P}{\partial (V_a/V_p)} \tag{10}$$

follows.

If the pores are assumed to have cylindrical geometry, the pressure difference over the water/adsorbate interface is calculated with the Laplace equation

$$P - P_w = -\frac{2\sigma_w \cos \theta}{r_m}, \tag{11}$$

in which $P_w$ is the pressure in the aqueous phase.

Differentiating Eq. 11 gives

$$\frac{\partial P}{\partial r_m} = \frac{2\sigma_w \cos \theta}{r_m^2}. \tag{12}$$

From Eq. 6 we have

$$\frac{\partial (V_a/V_p)}{\partial r_m} = f(r_m), \tag{13}$$

and so

$$\frac{\partial P}{\partial (V_a/V_p)} = \frac{2\sigma_w \cos \theta}{r_m^2 f(r_m)}. \tag{14}$$

The linear velocity $u$, which is caused by a pressure gradient in the adsorbate, is calculated as a Poiseuille flow

$$u = -\frac{r^2}{8\mu_a} \frac{\partial P}{\partial x} \tag{15}$$

in one cylindrical pore, filled with adsorbate.

For the average linear velocity $\langle u \rangle$ over all pores, we have

$$\langle u \rangle = \int_0^{r_m} u(r) f(r)\, dr = \int_0^{r_m} \left( -\frac{r^2}{8\mu_a} \frac{\partial P}{\partial x} \right) f(r)\, dr. \tag{16}$$

Relating the superficial velocity of the capillary transport to the linear velocity:

$$u_c = \epsilon \langle u \rangle \tag{17}$$

gives

$$k_r k_\infty = \frac{\epsilon}{8} \int_0^{r_m} f(r) r^2\, dr. \tag{18}$$

Substitution of Eqs. 14 and 18 in Eq. 10 then gives

$$D'_{a,pc} = \frac{1}{4\mu_a} \frac{\sigma_w \cos \theta}{r_m^2 f(r_m)} \int_0^{r_m} r^2 f(r)\, dr, \tag{19}$$

in which the subscript *pc* stands for "parallel cylinders." To also include tortuosity effects and other deviations from the idealized parallel cylinder model here, we include a matching factor, $F$, analogous to van der Zanden et al. (1996):

$$D'_a = FD'_{a,pc} = \frac{F}{4\mu_a}\,\frac{\sigma_w \cos\theta}{r_m^2 f(r_m)}\int_0^{r_m} r^2 f(r)\,dr, \tag{20}$$

and from Eqs. 1 and 2 we obtain

$$D_{eff,I} = D'_w + D'_a = \frac{D_w \epsilon}{\tau}\left(1-\frac{V_a}{V_p}\right)\Bigg/\left(\rho_{app}\frac{\partial Q}{\partial C}\right) + \frac{F}{4\mu_a}\,\frac{\sigma_w \cos\theta}{r_m^2 f(r_m)}\int_0^{r_m} r^2 f(r)\,dr \tag{21}$$

for the effective diffusion coefficient.

### *Model II: Discontinuous capillary transport*

We now take within a representative element the capillary phase as continuous over a distance $d_1$, in series with an aqueous phase region over a distance $d_2 = d_1/\delta$, as represented in Figure 4. For the transport in the lefthand part of the system, the same transport equations hold as were derived for model I. Since all adsorbate is now thought to be present over the distance $d_1$, the cross sections for water and adsorbate transport should be corrected for this at a given load onto the carbon. Thus in the lefthand part we have

$$J = -\left(D'_{w,II} + D'_{a,II}\right)\rho_{app}\frac{\partial Q}{\partial x} \qquad 0 < x < d_1, \tag{22}$$

with

$$D'_{w,II} = \frac{D_w \epsilon}{\tau}\left(1-\frac{V_a}{V_p}\,\frac{1+\delta}{\delta}\right)\Bigg/\left(\rho_{app}\frac{\partial Q}{\partial C}\right) \tag{23}$$

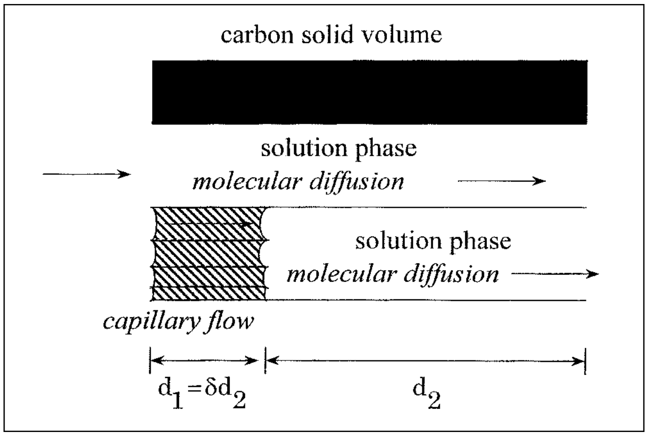

**Figure 4. Parallel capillary flow in adsorbate phase and molecular diffusion in aqueous phase, for discontinuous capillary flow (model II).**

and

$$D'_{a,II} = \frac{F}{4\mu_a}\left(\frac{1+\delta}{\delta}\right)\frac{\sigma_w \cos\theta}{r_m^2 f(r_m)}\int_0^{r_m} r^2 f(r)\,dr. \tag{24}$$

For the aqueous phase part on the righthand side, we can write

$$J = -D''_{w,II}\,\rho_{app}\frac{\partial Q}{\partial x} \qquad d_1 < x < d_2, \tag{25}$$

with

$$D''_{w,II} = \frac{D_w \epsilon}{\tau}\Bigg/\left(\rho_{app}\frac{\partial Q}{\partial C}\right). \tag{26}$$

Thus, we obtain

$$J = -D_{eff,II}\,\rho_{app}\frac{\partial Q}{\partial x} \tag{27}$$

from the series arrangement of the two regions, with

$$\frac{1}{D_{eff,II}} = \frac{\delta}{1+\delta}\,\frac{1}{D'_{w,II} + D'_{a,II}} + \frac{1}{1+\delta}\,\frac{1}{D''_{w,II}}. \tag{28}$$

There are two limiting situations for $\delta$. If $\delta \to \infty$, a situation is considered in which a continuous capillary separated adsorbate phase exists through the whole porous material. The adsorbate flux will then be given by model I. The minimum value for $\delta$ follows from Eq. 23:

$$1 - \frac{V_a}{V_p}\,\frac{1+\delta}{\delta} = 0, \tag{29}$$

and thus,

$$\delta_{\min} = \frac{1}{\left(\dfrac{V_a}{V_p}\right)^{-1} - 1}. \tag{30}$$

In this case there is only serial transport by molecular diffusion and capillary flow.

## Results and Discussion

Miyahara and Okazaki (1992, 1993) measured the adsorption isotherms of nitrobenzene at 293 K, 308 K, and 323 K, and benzonitrile at 293 K and 308 K, both from an aqueous solution on activated carbon X-7000. They also measured the diffusion coefficients for both compounds as a function of carbon loading at the different temperatures.

The adsorption equilibria data of one component at different temperatures can be expressed by a single characteristic curve using the potential theory (Miyahara and Okazaki, 1993). In such a plot the amount adsorbed is given as a func-

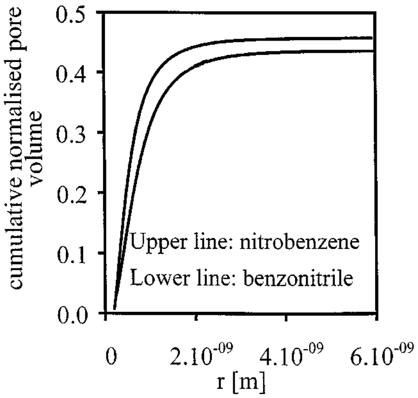

**Figure 5. Normalized cumulative pore-size distributions of activated carbon X-7000, obtained with Eqs. 5 and 6 from the sorption isotherms of nitrobenzene and benzonitrile.**

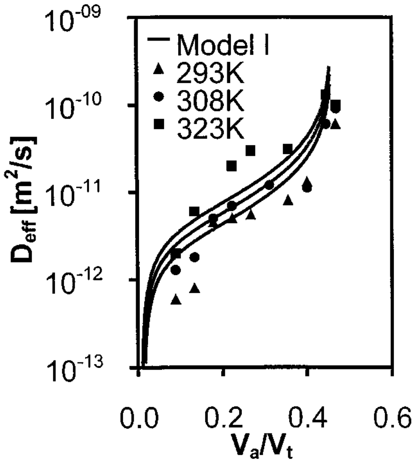

**Figure 6. Dependency of diffusivity of nitrobenzene on volume fraction filled with nitrobenzene at different temperatures.**

Comparison of the experimental data of Miyahara and Okazaki (1993) with $D_{eff,I}$, calculated by model I. $F = 0.0125$.

tion of $RT\ \ln(C_s/C)$. Potential plots were used to calculate the pore size distribution from the isotherms of nitrobenzene and benzonitrile via Eqs. 5 and 6. The result is shown in Figure 5. The validity of the potential theory indicates that the temperature dependency of $\sigma_w \cos\theta$ on temperature in the Kelvin equation (Eq. 5) is negligible.

The measured sorption isotherms gave no information about the pore volume fraction from zero to 0.05. To obtain this information, the isotherms were extrapolated to the lower carbon loading. Also, when using the sorption isotherms in calculating the pore-size distribution, the distribution of pores with a radius larger than 6 nm remains unknown.

The diffusivity for continuous capillary transport, model I, was calculated from Eq. 21. Viscosity data at different temperatures were obtained from the *Handbook of Chemistry and Physics* (Lide, 1996). The temperature dependency of the diffusivity via the temperature-dependent surface tension is about one order of magnitude smaller than the temperature dependency of the diffusivity via the temperature-dependent viscosity. That is why the dependency of surface tension on temperature was neglected in the calculation of the effective diffusivity. Some properties of nitrobenzene and benzonitrile are shown in Table 1.

In Figures 6 and 7 the effective diffusion coefficient $D_{eff}$, calculated with model I, is compared to the experimental data of Miyahara and Okazaki for nitrobenzene and benzonitrile, respectively. The pore-size distribution, used in the calculation, was the pore-size distribution calculated from the isotherm of the corresponding compound. The correction factor $F$ in Eq. 21 equals 0.0125 in both cases. The correction factor is expected to be a constant for a particular porous material, because it is determined mainly by the pore structure. Translated to the model in this work, van der Zanden would have found a correction factor in the same order of magnitude for the drying behavior of clay ($F = 0.01$) (van der Zanden et al., 1996; van der Zanden, 1995).

The model underestimates the temperature dependency of the diffusivity. Miyahara and Okazaki used Eyring's rate theory in combination with the potential theory of adsorption to model the same experimental data as were used here (Miyahara and Okazaki, 1993). If their model is compared to the capillary transport model used here, it is found that the two models are quite similar as regards the temperature dependency. Miyahara and Okazaki found a free-energy activation for the hole-making step that is comparable to the activation free energy activation for the viscosity of liquids.

The large difference between the models is that the capillary models account for the pore structure, as related to the sorption isotherm, and the surface diffusion model does not.

In Figure 8 some results, calculated with model II, are given for the adsorption of nitrobenzene. The tortuosity and the

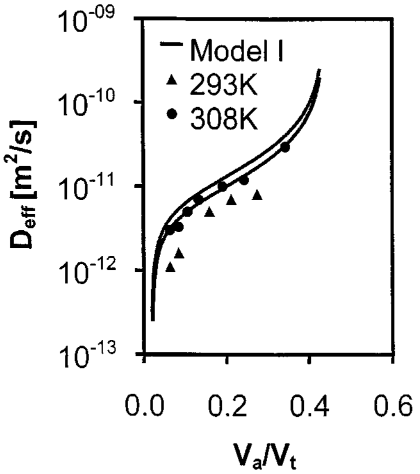

**Figure 7. Dependency of diffusivity of benzonitrile on volume fraction filled with benzonitrile at different temperatures.**

Comparison of the experimental data of Miyahara and Okazaki (1993) with $D_{eff,I}$, calculated by model I. $F = 0.0125$.

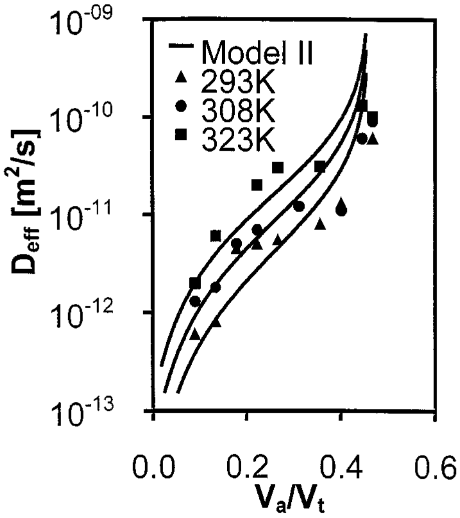

**Figure 8. Dependency of diffusivity of nitrobenzene on volume fraction filled with nitrobenzene at different temperatures.**

Comparison of the experimental data of Miyahara and Okazaki (1993) with $D_{eff,II}$, calculated by model II. $F = 1$, $\delta = \delta_{\min} + 15$.

correction factor $F$ both equal 1. Good coverage of experimental data was found for $\delta = \delta_{\min} + 15$.

The discontinuity of the capillary separated phase results in a calculated effective diffusivity, which is smaller at the same correction factor. Model II gives a good estimation of the temperature dependency of the effective diffusion coefficient. This is caused by the transport by molecular diffusion, which plays a limiting role in model II. This transport has a larger temperature dependency than does transport by capillary flow.

In this model $\delta$ is fitted to obtain good agreement with the experimental data. One way to avoid a fitting parameter like $\delta$ is to model the pore structure as a network of connected cylinders (Koplik, 1982; Matthews, 1993). In that case, the continuity of the adsorbate phase will be determined by the geometry of the network.

## Conclusions

It has been shown that the assumption of solely molecular diffusion of adsorbate through the porous system underestimates the experimental effective diffusion coefficient.

Two models, based on capillary transport, were used to predict the diffusivities of the adsorption of nitrobenzene and benzonitrile on activated carbon from an aqueous solution. The influence of carbon loading on the diffusivity is included in the pore structure, which is obtained from the adsorption isotherm of the adsorbate. Model I assumes a continuous capillary separated adsorbate phase. One eligible parameter is needed to correct for the pore structure and other deviations from reality. The fit parameter is the same for the two adsorbates, and thus characteristic for the adsorbent. The eligible parameter is also found to be on the order of magnitude as the same eligible parameter found in modeling the drying behavior of clay. The temperature dependency of the diffusivity is slightly underestimated by the model, but the dependency of carbon loading on the diffusivity fits reasonably well.

Model II, in which capillary transport and molecular diffusion partly occur in series, gives a good estimation of the temperature and carbon loading dependency of the apparent diffusivity.

Although many physical phenomena will deviate on the molecular level from the simplified models used here, it is remarkable that a relatively simple transport theory, such as capillary flow, is able to give such a good description of such complex phenomena. It is therefore of interest, on the one hand, to investigate this type of theory further for the purpose of engineering, and, on other hand, to try to relate molecular science to the effective parameters used here.

## Notation

$D_{eff}$ = effective diffusivity [m$^2$ s$^{-1}$]
$D'_w$, $D''_w$ = effective diffusivity of adsorbate in water phase [m$^2$ s$^{-1}$]
$J_c$ = volumetric adsorbate flux [m$^3$ m$^{-2}$ s$^{-1}$]
$J$ = mass adsorbate flux [kg m$^{-2}$ s$^{-1}$]
$J_w$ = adsorbate flux in aqueous phase [kg m$^{-2}$ s$^{-1}$]
$k_\infty$ = permeability [m$^2$]
$k_r$ = relative permeability
$R$ = gas constant [J mole$^{-1}$ K$^{-1}$]
$T$ = absolute temperature [K]
$V_t$ = volume of adsorbate at total saturation [m$^3$]

### Subscripts

$I$, $II$ = model I, model II, respectively

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