

THE ABSORPTION OF GASES IN AQUEOUS ACTIVATED CARBON SLURRIES ENHANCED BY
ADSORBING OR CATALYTIC PARTICLES.

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ABSTRACT

The enhanced absorption of gases in aqueous activated carbon slurries of fine particles is studied with an instationary absorption model taking into account the finite adsorption capacity of the carbon particles, and with a stationary geometrical model, which describes the absorption into a highly reactive and concentrated slurry. These models are applied to explain literature results on activated carbon slurries ("grazing" effect) and results obtained on hydroxylamine production in a slurry reactor. With the results of the two presented models it can be understood that even in intensively agitated reactors enhanced absorption is possible.

KEYWORDS

gas absorption enhancement; activated carbon slurries; finite adsorption capacity; penetration model; stationary geometrical model

INTRODUCTION

During process development work on hydroxylamine production at DSM (The Netherlands) it was observed that enhancement of gas-liquid mass transfer occurred with 1% Pt on activated carbon catalyst. The particle size was roughly equal to the estimated hydrodynamic film thickness. Taken into account the relatively low bulk solid concentration (5-24 kg/m³) the observed enhancement (~3) could not be explained by any available consistent theory. This prompted us to investigate the enhancement in slurry systems thoroughly. We concluded that three separate factors could be involved:

- 1. Increase of particle concentration close to the g/l-interface.
- 2. Particle finite adsorption capacity effects.
- 3. Geometrical factors at the elevated particle concentration close to the interface.

It is known from relatively recent work, that in catalytic slurry reactors small particles may enhance the gas absorption by forming a sink for dissolved gas in the diffusion film at the g/l-interface (Alper, 1980; Alper & Deckwer, 1983). In that case the particles should be small enough to be accommodated in the boundary layer. Alper and Deckwer (1983) have given an extensive review of this subject and classified roughly two categories of particles:

- 1. Particles that affect the mass transfer rate by their presence in the film, because of their large adsorption capacity for the absorbed gas, e.g. finely powdered activated carbon (Alper, 1980).
- 2. Particles that enhance the reaction in the film by heterogeneous catalysis: e.g. Pt, Pd, or immobilized enzymes on fine activated carbon particles (Alper, 1980; Wimmers, 1984, 1987).

In fact also a combined action of these two processes is possible.

For the first category of Alper and Deckwer due to the particle saturation effect, a film model cannot be used, and therefore we proposed a pseudo-homogeneous instationary penetration model, which describes the mass transfer of a gas in a slurry containing particles with limited capacity for adsorption or reaction (Holstvoogd, 1986). It can be demonstrated with this model that the partial conversion/saturation of the activated carbon particles within the mass transfer zone may strongly reduce the possible enhancement factor. The purpose of the present study is to demonstrate with literature data on the physical absorption of gases (O₂ and CO₂) into aqueous activated carbon slurries, Alper's first category, the three factors involved. The previously mentioned absorption of H₂ and NO in an aqueous activated carbon slurry in order to produce hydroxylamine is taken as an example of the second category.

LITERATURE

Generally it has been observed that the absorption rate, when enhanced by fine activated carbon

particles, increases with increasing activated carbon concentration, until a certain concentration is reached after which the rate remains constant. Moreover studying many different gas activated

Table 1 Minimum activated carbon concentration for maximum enhancement, $m_{E^{\infty}}$ of gas absorption in several systems.

Gas - slurry system	$m_{E^{\infty}}$ kg/m ³
1. O ₂ in aq. AcC-slurry (Alper, 1980): $d_p \leq 5 \mu m$, T=50°C	1.0
2. CO ₂ in aq. AcC-slurry (Alper, 1980): $d_p \leq 5 \mu m$, T=25°C	2.0 - 3.0
3. propane in aq. AcC-slurry (Kars, 1979): $d_p = 34 \mu m$	no $m_{E^{\infty}}$ known, but if $m_s = 4.9$: E = 1.2
4. CO ₂ in aq. Na ₂ CO ₃ -buffer/AcC-slurry (Alper, 1980): $d_p \leq 5 \mu m$	2.0 - 3.0
5. O ₂ in aq. Na ₂ SO ₃ /AcC-slurry (Alper, 1983, 1986b): $d_p \leq 5 \mu m$	at 5°C : 1.5 at 25°C : 1.0
6. O ₂ in aq. Na ₂ S/AcC-slurry - (Pal, 1982): $d_p \leq 5 \mu m$, T=80-150°C - (Alper, 1986a): $d_p < 10 \mu m$, T=15, 25, 35°C - (Chandrasekaran, 1977): $d_p < 100 \mu m$, T=70°C	0.6 - 16 ~ 1.0 1.0
7. O ₂ in aq. glucose/Pt on AcC-slurry - (Lee, 1972): according to Alper no enhancement could be possible - (Alper, 1980): $d_p \leq 5 \mu m$	0.4 10
8. CO ₂ in aq. Na ₂ SO ₃ -buffer/Pt or imm. enzyme on AcC-slurry (Alper, 1980; Alper, 1983, 1986b; Sada, 1982): $d_p < 20 \mu m$	2 - 3

carbon slurry systems on the enhanced absorption, it is remarkable that the maximum enhancement is nearly always reached at the same activated carbon concentration level in spite of the large differences in reaction rate constants, which are to be expected. In Table 1 the minimum carbon concentrations necessary for reaching the limiting enhancement, $m_{E^{\infty}}$, are summarized for several systems studied experimentally. The range of values for $m_{E^{\infty}}$ is quite narrow in spite of the large differences in the investigated systems. Only the data of Pal (1982), which are criticized by Alper (1986a), form an exception.

The limiting enhancement is usually explained (Alper, 1981; Sada, 1982) as a consequence of reaching the minimum effective film thickness, $(\delta_{l,eff})_{min} = \delta_l / E^{\infty} = d_p$, which approaches the particle diameter. A smaller effective film thickness cannot be reached, because in a layer of about the size of the particle diameter no particles are assumed to be present, so no heterogeneous reaction can take place there. However, from this model it cannot be easily understood that for different physical and chemical systems the maximum enhancement (E^{∞}) is

reached at approximately the same $m_{E^{\infty}}$ -values. Moreover in case of reaction of dissolved O₂ with a bulk reactant, S²⁻ (O₂ absorption in aq. Na₂S solution with activated carbon particles), increasing the latter concentration increased the maximum enhancement (Alper, 1986a), which is not consistent with this model (Holstvoogd, 1988a, 1988b).

THE INFLUENCE OF THE ADSORPTION CAPACITY ON THE ENHANCEMENT FACTOR

Increased particle concentration at the interface

Several authors have suggested that the interfacial activated carbon concentration could be larger than the bulk concentration. Lee and Tsao (1972) visually observed that there were carbon particles floating on the liquid surface, and concluded that this tendency causes a higher local concentration and thus accounts through the higher local reaction rate for the extra enhancement. Alper and Ozturk (1986a) agree with the suggestion of Sada and Kumazawa (1982), who evaluated experimental results of Alper (1980, 1981) on glucose oxidation on Pt/activated carbon and hydrolysis of CO₂ in the presence of immobilized carbonic anhydrase, that the catalyst particle concentration in the range of $\lambda < x < D/k_1$ should have been much higher than in the bulk liquid phase. However they still adhered to the idea of a layer λ , which does not contain particles. Another indication of the higher interfacial concentration is given by Wimmers (1984). They measured enhanced absorption of gas from a gas bubble into a stagnant liquid, in which on the bubble surface catalytic activated carbon particles are attached by their surface properties with a volume fraction of about 16%, corresponding to 140 kg/m³ activated carbon ($\rho_p = 850 \text{ kg/m}^3$). So although there seems to be general agreement, that the particle concentration is increased at the interface it is mainly based on indirect evidence: enhancement interpreted by the film model or on measurements with stagnant bubbles. Another argument in favour of increased solid concentration can be derived from experiments on physical absorption of gases in activated carbon suspensions. Even in those cases that the carbon concentration in the bulk would be sufficient for providing enhancement in the film, particle saturation in the film could prevent it, as will be demonstrated below.

Physical gas absorption

Model. It is possible to simulate the absorption of a gas in aqueous activated carbon slurries with the aid of the penetration model developed earlier (Holstvoogd, 1986) taking into account the limited adsorption capacity of the activated carbon particles. In this model the gas phase component is considered to penetrate into the slurry by stationary diffusion, while as a first approximation mass transfer and chemical reaction and/or physical adsorption are treated as pseudo-homogeneous phenomena in the mass transfer film.

The absorption of gas A in the liquid followed by the adsorption on the carbon particle is treated as follows. Assuming the activated carbon/water system has an almost linear adsorption isotherm for adsorption of adsorbed component A, the concentration of A in the liquid, which is in equilibrium with the current amount of adsorbed gas on the particle, is:

$$C_{A,c} = n_A / (m_s K_{ads}) \tag{1}$$

If, as a simplification of a much more complex process, the mass transfer to the particles and the adsorption on the particles are considered to be processes in series, the flux to the particles can be represented by

$$J \cdot a_s = k_s a_s (C_A - C_{A,s}) = k_{ad} a_s (C_{A,s} - C_{A,c}) \tag{2}$$

where $C_{A,s}$ is the concentration of A in the liquid at the liquid-solid interface. After defining k_p as the overall particle rate constant for mass transfer to the particle with or without adsorption on the particle,

$$\frac{1}{k_p} = \frac{1}{k_s} + \frac{1}{k_{ad}} \tag{3}$$

the total flux to the particles becomes $J \cdot a_s = k_p a_s (C_A - C_{A,c})$ (4)

Now assuming, that a stagnant slurry package with a zero initial adsorbed gas loading (to start with a higher loading is also possible) stays for a contact time τ at the g/l-interface, and that no corrections for the effective diffusion coefficient have to be made, the basic equations of the penetration model are:

- mass balance of A:

$$\frac{\partial C_A}{\partial t} = D_A \frac{\partial^2 C_A}{\partial x^2} - k_p a_s (C_A - C_{A,c}) \tag{5}$$

- mass balance of adsorbed A:

$$\frac{\partial n_A}{\partial t} = k_p a_s (C_A - C_{A,c}) \tag{6}$$

- boundary conditions: $t = 0; x \geq 0$, and $t \neq 0; x = \infty : C_A = C_{A,c} = n_A = 0$
 $x = 0; t \geq 0 : C_A = mC_{A,g}$ (7)

This set of parabolic differential equations (Eqns. (5)-(7)) has been solved numerically by a method published previously (Holstvoogd, 1988a). The enhancement factor E is defined as the average flux of A as compared to the situation with inert particles

$$E = \frac{1}{\tau} \int_0^\infty -D_A \frac{\partial C_A}{\partial x} \Big|_{x=0} dt / \left[2 \sqrt{\frac{D_A}{\pi t}} (C_{A,x=0} - C_{A,x=\infty}) \right] \tag{8}$$

where τ is the Higbie contact time of the stagnant slurry element with the gas phase.

We will define a Hatta number for pseudo-homogeneous and heterogeneous applications:

$$Ha_h = \sqrt{k_p a_s D_A} / k_l \tag{9}$$

Although the saturation of the particles will take place more gradually, we may calculate an equivalent layer of slurry in which the particles are completely saturated:

from the mean flux to the particles, $J_{ad} = J_{enh} - J_{phys} = (E-1)k_l m C_{A,g}$ follows that the equivalent saturated layer thickness relative to the hydrodynamic film layer becomes

$$\frac{\delta_{sl}}{\delta_l} = \frac{J_{ad} \tau}{n_{A,max}} / \delta_l = \frac{4(E-1)}{\pi m_s K_{ads}} \tag{10}$$

The simulations show that at a given solid concentration varying the overall particle rate constant, k_p , that at low rates the enhancement factor can be calculated by the simple penetration theory solution: $E = Ha_h / \tanh(Ha_h)$. We found that an equivalent saturation level of lower than 5% had no effect on the enhancement. Now a minimum adsorption capacity for avoiding capacity effects on the enhancement factor can be deduced (Holstvoogd, 1988a, 1988b):

$$m_s K_{ads} > \frac{Ha_h / \tanh(Ha_h)}{0.05} \frac{4}{\pi} \tag{11}$$

for instance to achieve $E = Ha_h = 2$ then $m_s K_{ads} \geq 27.4$

However increasing the particle rate constant lower enhancement factors compared to the simple penetration theory solution are being obtained due to partial saturation of the activated carbon particles in the boundary layer. Then in a transition region also the adsorption capacity determines the enhancement factor. A limiting situation occurs when the adsorption capacity is low and the adsorption rate is high. Then this capacity is completely determining the enhancement and E becomes an unique function of the adsorption capacity, $m_s K_{ads}$ (see Fig. 1). Simulations of literature results on enhanced absorption with finite adsorption capacity will be discussed hereafter.

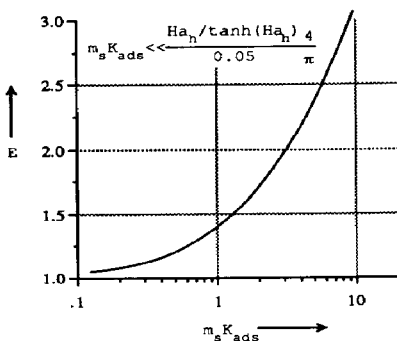


Fig. 1 The enhancement factor as an unique function of the adsorption capacity.

O₂ absorption. Alper (1980) has studied the absorption of O₂ in aqueous activated carbon slurries. In a stirred cell with flat g/l-interface the absorption rate, $k_1 a$ has been measured as a function of the stirrer speed and the carbon concentration. In absence of exact data, normal flat surface stirred cell dimensions have been assumed, where the liquid height is roughly equal to the cell diameter. In Table 2, for a stirrer speed of 250 r.p.m., the enhancement factor caused by activated carbon particles is listed as a function of the carbon concentration. Alper could only

explain qualitatively the enhancement caused by the presence of carbon particles by postulating that the particles adsorb the gas in the film and transport it to the bulk of the liquid, the so called "shuttle" or "grazing" mechanism (Alper & Deckwer, 1983; Kars, 1979).

Table 2 Data on O₂ absorption in activated carbon slurries estimated from Alper's experiments (Alper, 1980). Conditions: N=250 r.p.m., T=50°C, D_{O₂}=4.3×10⁻⁹m²/s, m=0.012, d_p≤5 μm, V_l=3×10⁻⁴m³, k_l≈3.6×10⁻⁵m/s, E=k_l(withAcC)/k_l(withoutAcC).

m _s kg/m ³	E
< 0.1	1.00
0.1	1.46
≥ 1.0	3.38

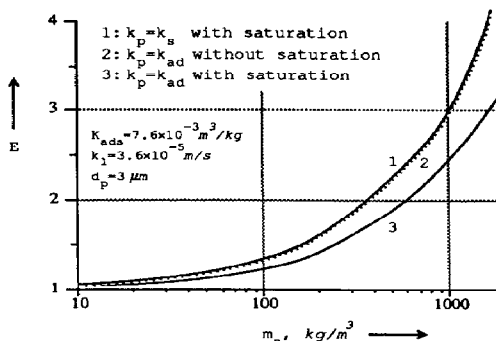


Fig. 2. Enhancement factor vs. solid concentration during O₂ absorption in carbon slurries showing the influence of possible regimes in local rates.

The parameters given in Fig. 2 refer to the experiments described above. The more difficult parameters the adsorption equilibrium constant, K_{ads} , and the adsorption rate constant, k_{ad} , were obtained in the following way. From different sources (Li & Deckwer, 1982; Recasens, 1984) it was concluded that $K_{ads}=7.6 \times 10^{-3} \text{ m}^3/\text{kg}$ is a reasonable value. The adsorption rate constant was estimated to be $k_{ad}=1.2 \times 10^{-6} \text{ m/s}$ (Recasens, 1984). The liquid to particle mass transfer coefficient can be calculated assuming $Sh=2$ to be valid for the average particle diameter, resulting in k_s for $d_p=3 \text{ μm}$: $k_s=2.9 \times 10^{-3} \text{ m/s}$. On basis of a k_p -value varying between 1.2×10^{-6} for adsorption limitation and 2.9×10^{-3} for mass transfer limitation to the particle, we have performed our simulations to investigate the activated carbon concentrations in the film (see Table 2) needed to reach the enhancement factors for O₂ absorption obtained by Alper. The model enhancement factors are calculated and plotted against the solid concentration for the two extreme k_p -values in Fig. 2. If k_p is determined by the adsorption rate, line 3 represents the simulations. Here capacity and Ha_H determine the enhancement. The influence of the capacity can be observed by a comparison with line 2, where saturation is excluded. At the high k_p -value, $k_p=k_s$ (see line 1), the relation between E and the solid concentration is independent of the Ha_H -number (relation of Fig. 2 for $K_{ads}=7.6 \times 10^{-3} \text{ m}^3/\text{kg}$).

A similar phenomenon can be observed while evaluating the physical absorption enhancement of CO₂ in water by activated carbon particles. Alper (1980) measured maximum enhancement at bulk solids concentrations larger than 2 to 3 kg/m³. With an adsorption equilibrium constant of $K_{ads}(25^\circ\text{C})=3.3 \times 10^{-3} \text{ m}^3/\text{kg}$ (Li & Deckwer, 1982), the adsorption capacity is much too low to allow enhancement. Alper measured for an average bulk solid concentration of 0.1 kg/m³, an enhancement factor of 1.46. In our simulations this enhancement could only be reached at solid concentrations of 240 and 140 kg/m³ for resp. $k_p=k_{ad}$ and $k_p=k_s$, for $K_{ads}=7.6 \times 10^{-3} \text{ m}^3/\text{kg}$. These concentrations are far higher than the actual bulk concentrations. Experimentally the limiting enhancement factor, $E^\infty (=3.38)$ is reached at bulk concentrations of higher than 1.0 kg/m³ however the simulations have pointed out that very high carbon concentrations (>500 kg/m³) are needed to calculate such an enhancement. On the other hand the adsorption capacities needed to avoid any saturation can be estimated with Eqn. (11). For an enhancement factor of 1.46 an adsorption capacity, $m_s K_{ads}$ would be needed larger than 11.7. Unrealistic high solid concentrations (>1500 kg/m³) would be necessary to reach this capacity. Even higher concentrations are needed for the limiting enhancement factor. Our simulations show clearly that during the experiments of Alper the saturation effect had a considerable influence on the O₂ absorption, and that the carbon concentration in the boundary layer at the g/l-interface should have been much larger than the bulk concentration. Therefore the limiting value of E in Alper's experiments is probably caused by a maximum solid concentration at the interface rather than by a solid free zone at the interface. Saturation was not taken into account by Alper as in the stationary film model used by them it cannot be incorporated in a meaningful way.

GEOMETRICAL FACTORS

Model

Given the conclusion that the carbon particles are concentrating to almost packed bed densities at the interface, especially at extremely high adsorption/conversion rates it can be expected that the description on basis of a pseudo-homogeneous model will become inaccurate. Therefore the gas absorption into a slurry has been modelled taking into account geometrical factors. The stationary diffusion flux through a regular lattice of reacting particles was mathematically solved by a finite-element method (Holstvoogd, 1988a). In the near future the instationary diffusion will be studied to allow introduction of the geometrical factors into the

penetration model, but at present only stationary results are available.

The essential part of this heterogeneous model is the definition of the regular packing of the cubic cells, which constitute the slurry in the mass transfer zone. Assuming the spherical solid particle is placed in the centre of such a cubic cell, then the particles volume fraction is equal the ratio of the sphere's volume and the cube's volume (edge is d_c) resulting in the following relationship

$$\epsilon_s = [\pi d_p^3 / 6] / d_c^3 \quad (12)$$

For a given solid concentration the dimensions of the cubic cell follow from this relationship. Only the most simple geometry will be considered, where the particles, seen from the interface, are situated in line with each other. Therefore the hydrodynamic film thickness, δ_1 , will be built up by a number of cubic cells lined-up in a row.

For diffusion through the liquid the following stationary diffusion flux equation holds

$$D \left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right) = 0 \quad (13) \quad \text{with at } x=0: C=C_i \quad \text{and at } x=\infty: C=C_b^i$$

and at particle's surface, the diffusion flux is equal to the local reaction rate:

$$-D \frac{\partial C}{\partial r} \Big|_{r=r_p} = k_r'' (C - C_e) \quad (14)$$

Due to the complex spatial dimensions this differential Eqn. (13) with boundary conditions were solved by a finite-element method. The enhancement factor is defined as the ratio between the flux with heterogeneous reaction and the flux with no reaction and only diffusion through the lattice of particles.

Implications of the geometrical model

Model simulations (Holstvoogd, 1988a) have been carried out for a wide variety of conditions. An interesting feature was that the enhancement factor becomes limited at very high reaction rates. This effect is caused by the diffusion rate limitation of dissolved gas between the g/l-interface at the first layer of fast reacting particles. The thickness of this layer depends on the solid fraction as can be deduced from Eqn. (12). Therefore the maximum enhancement factor will be higher for higher solid fractions. At maximum enhancement conditions and high solid concentrations the diffusion process is confined to the first layer of reactive particles. This situation occurs when particles with an extremely fast (external) surface reaction rate and an infinite capacity are attracted to the interface forming a reactive monolayer.

A comparison of the enhancement factors calculated by the pseudo-homogeneous model with infinite capacity of the particles and by this heterogeneous model in Fig. 3 demonstrates, that at low solid concentrations the enhancement factors are of nearly equal value, however at higher concentrations the heterogeneous model calculated higher enhancement factors. The maximum enhancement factor according the pseudo-homogeneous film absorption model, assuming that the adsorption capacity is infinitely large, can be calculated when the mass transfer to the particles can be described by the Sh=2 relation. Then according to Eqn. (3) the overall particle rate constant, k_p is equal to k_s .

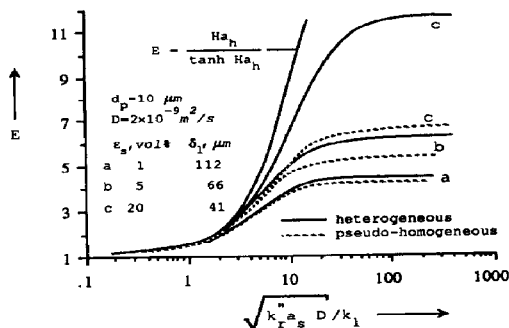


Fig. 3. Comparison of the enhancement factor according to the pseudo-homogeneous model with infinite capacity and the heterogeneous model as function of the solid volume fraction. Heterogeneous model calculated for 3 cubic cells.

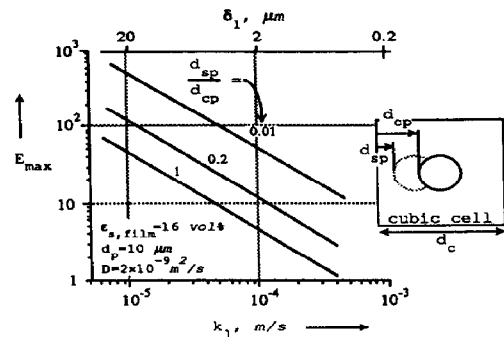


Fig. 4. The effect of particle attraction to the g/l-interface for extremely fast surface reaction. Capacity effects of the particles neglected.

What the practical consequences are of a possible strong adhesion effect on the maximum reachable enhancement factor is revealed by Fig. 4. Here, assuming an average solid concentration of 16 vol%, observed by Wimmers (1984) as the activated carbon concentration around a stagnant bubble, the maximum enhancement factor for three relative particle to g/l-interface distances of the first layer of particles (d_{sp}/d_{cp}) are calculated for different k_1 -values. From this Figure it is evident that, assuming a high film solid concentration, the maximum attainable enhancement can become several factors larger than the enhancement calculated for the central particle position, and that

even at high k_1 -values considerable enhancement can be obtained, when the particles are attracted close to the g/l-interface. Moreover it was clear from the simulations that at high solid concentrations ($\geq 5\%$) the whole phenomenon is dominated by the first particle layer. Although for high k_1 -values the film thickness is small compared to the cube's length, the model's maximum enhancement factor is in general correct, since then only in the front part of the cell diffusion and reaction take place. It should be realized that the particle capacity factor has been neglected, and that exact calculations are only possible if local geometry is known.

EVALUATION

During the production of hydroxylamine by the heterogeneous reaction of H_2 and NO in an aqueous slurry of 1% Pt on activated carbon particles (in a high-intensity stirred reactor, bulk solid concentration = 5 to 24 kg/m^3 , $d_p(50\%) < 10 \mu m$) mentioned before the gas absorption was enhanced. Overall volumetric mass transfer coefficients of about 2.6–2.9 s^{-1} was measured, while under similar conditions a physical volumetric mass transfer coefficients of 0.9 s^{-1} was determined. The difference obtained could only be explained by chemical enhancement by a monolayer of catalytic particles ($d_p/\delta_1=1$). Pseudo-homogeneous model calculations, taking into account the mass transfer from the liquid to the solids and simultaneous diffusion and adsorption/reaction steps in the particle, have shown that only catalyst concentrations of 20 times the bulk solid concentrations could explain the observed enhancement factors.

The evaluation of the physical absorption in activated carbon slurries by the instationary model has shown that the film solid concentration has to be much higher than the bulk concentration due to attraction of activated carbon particles to the interface, although the required film concentrations were unrealistically high. However the results of the geometrical model have demonstrated, that at much lower film solid concentrations enhancement is possible, and that in case of strong attraction of activated carbon to the interface at even lower concentrations the found enhancement factors can be reached. Considering the results of the simulations of two models proposed the reported enhancement of gas absorption during hydroxylamine production can be understood.

Although in this evaluation the facilitation of mass transfer by gas adsorption on the particles has not been considered, this effect can also play a role in case the actual surface reaction rate is slow compared to the adsorption rate. Then the reaction will progress in the bulk of the slurry and the gas absorption rate can be increased by the adsorption effect. However to model this phenomenon implicates the incorporation of the instationary saturation process in the heterogeneous model, which has not yet been realized. Moreover much more parameters involved, like internal diffusion coefficients, local geometry, should be known exactly, and these data cannot be obtained easily.

CONCLUSIONS

Gas absorption in activated carbon slurries can be enhanced by adsorption and/or by chemical reaction on the particles. In case of adsorption, partial saturation of the particles may strongly influence enhancement and this effect can be described by the pseudo-homogeneous penetration model. For higher (local) slurry concentrations and particle surface reaction rates, due to geometrical effects, still higher enhancement factors are possible than foreseen from the pseudo-homogeneous film model. The maximum enhancement may be quite sensitive to the local geometry. In these cases a monolayer of particles can already lead to high enhancement factors if particle capacity factors can be neglected.

From evaluation of literature data it follows that carbon particles are attracted to the g/l-interface. Specially with physical absorption in slurries, due to the saturation of the particles during the contact time, much higher concentration at the interface should be present than in the bulk to explain the experimental results. Also direct observation and observed chemical enhancement reported in literature indicate increased interface particle concentrations.

From the analysis of the hydroxylamine production in slurries it was found that a monolayer of relatively large particles could provoke an enhancement. Apart from the chemical reaction also adsorption of limiting reactants by the particles followed by conversion in the bulk may contribute to the observed overall enhancement.

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NOTATION

a_s specific interface of solids in liquid, $=6m_s/(\rho_p d_p)$, $m^2/m^3 liq.$
 C concentration of a gas component in the liquid phase, $mol/m^3 liq.$
 d_p particle diameter, m

ϵ_s solid volume fraction, -
 λ particle-free liquid zone at g/l-interface, m
 τ Higbie contact time, $=4D_A/(\pi k_1^2)$, s
 ρ_p dry particle density, $kg/m^3 solid$

d_{cp}	distance between g/l-interface and particle for central position in cubic cell, m	δ_1	film thickness in terms of film model, $=D/k_1$, m
d_{sp}	distance between g/l-interface and particle, m	δ_{s1}	equivalent saturated film layer, Eqn. (10), m
J	molflux, $mol/(m^2s)$		
K_{ads}	adsorption equilibrium constant, Eqn. (1), $m^3 liq./kg solid$		
k_{ad}	adsorption rate constant, m/s		
k_1	liquid-side mass transfer coefficient, m/s		
k_r	surface reaction rate constant, m/s		
k_s	liquid to solid mass transfer coefficient, m/s		
m	gas solubility, $=C_1/C_g$, -		
m_s	solid concentration, $kg solid/m^3 liq.$		
n	adsorbed gas concentration, $mol/m^3 liq.$		
n_{max}	maximum adsorbed gas concentration, $=m.C_g.K_{ads}.m_s$, $mol/m^3 liq.$		
r_p	particle radius, m		
		Indices	
		A	component A
		ad	adsorption
		b	bulk value
		e	equilibrium value
		eff	effective
		i	at interface
		∞	maximum

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