Three-Phase Mass Transfer: Improved Pseudo-Homogeneous Model

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This new pseudo-homogeneous model describes the absorption rate in the presence of dispersed second liquid phase or solid catalyst particles. In contrast to previous models, it considers the diffusion process and chemical reaction inside the dispersed phase. The absorption rate is defined in the case of first- and zero-order reactions for both continuous and dispersed phases. The concentration of particles from the bulk phase to the gas-liquid boundary layer replaces the initial concentration to the balance equation for the liquid boundary layer at the gas-liquid interface. For mass transport in the bulk phase, steady-state film theory, while in the gas-liquid boundary layer, unsteady-state film-penetration theory is applied. The effect of mass-transfer and reaction kinetic parameters, as well as the particle size on the absorption rate, is described as well as the simulated data verified by previous experimental studies.

Introduction

The absorption rate into slurries containing fine solid particles (catalysts or reactants) has been investigated intensively from both theoretical and experimental points of view in the last decade (Alper et al., 1981; Pal et al., 1982; Janakiraman and Shama, 1985; Nagy et al., 1986; Holstvoogd et al., 1986, 1988). Recent investigations have shown that the gas absorption rate can be substantially increased using a dispersed second liquid phase with permeability and/or solubility higher than that of the continuous phase. Bruining et al. (1986) investigated oxygen absorption into the aqueous phase in the presence of fine organic droplets, while Mehra et al. (1988) carried out a very rigorous study in two different three-phase systems. They investigated the absorption of organic components (isobutylene, propylene) in aqueous solutions of sulfuric acid and the dissolution rate of solid esters during alkaline hydrolysis in aqueous solutions of potassium hydroxide in the presence of an emulsified organic phase (chlorobenzene). The essential point of their work was the assumption that the mass transport inside the drops should be practically instantaneous. Therefore, the internal mass-transfer resistance could be neglected, and thus the concentration in the whole drop could be regarded as constant. In the drop-size regime up to 10 μm, the characteristic diffusion time, t_D ($t_D = R^2/D$ with $D = 2\times10^{-9}$ m^2/s, which is in the order of magnitude of diffusivity of oxygen in water), was less than 0.05 s. This is several times less than the average residence time of fluid elements in the gas-liquid boundary layer (with $\beta^o = \sqrt{Ds} = 1\times10^{-4}$ m/s, the lifetime is about 0.2 s). In the case of a three-phase slurry reactor where the dispersed phase consists of solid particles, however, the diffusion coefficient [$D_{\text{eff}} = D\theta/\tau$, with $\theta = 0.3$ to 0.5 and $\tau = 2$ to 6 (Satterfield et al., 1969; Kenney and Sedriks, 1972) can be one order of magnitude lower than that in a drop. Thus, the diffusion time for fine solid catalyst particles could even be higher than 0.2 s.

If a chemical reaction takes place in the dispersed phase, the problem is essentially different. This can be the case in both chemical (Mehra et al., 1988) and biochemical systems (Junker et al., 1990). The chemical reaction can considerably alter the concentration distribution of the absorbed component in the droplets and, consequently, the mass-transfer rate into the particles. Thus, this concentration should not be considered constant, as it was by Bruining et al. (1986) and Mehra (1988). To give the exact effect of reaction on the mass-transfer rate between the continuous and dispersed phases, the internal con-

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centration distribution in the dispersed phase has to be quantified. Junker et al. (1990) investigated the oxygen absorption rate during fermentation in two cases, namely, when perfluorocarbon was emulsified in the aqueous phase and when the aqueous phase was emulsified in the continuous perfluorocarbon phase. In their theoretical model, a zero-order reaction in the drops was taken into account to calculate the bulk phase concentration only.

There is no exact mathematical equation available from the literature to calculate the effect of an internal chemical reaction in the dispersed phase on the absorption rate (or on the dissolution rate of solid particles). A general mathematical model has been developed to quantify it. Because of its great importance in biochemical systems, the effect of first- and zero-order reactions is discussed in this article. The method used can easily be extended to other types of reaction kinetics, although an analytical solution for the mass-transfer rate cannot be obtained in this case. To solve the problem mentioned, the concentration distribution in the particles (drops or solid catalyst particles) of the bulk phase has to be defined first. Particles containing fluid elements come from the bulk phase into the gas-liquid (or solid-liquid) boundary layer with this concentration distribution. The mass transfer for the particles in the continuous bulk phase is regarded as a steady-state process. For mass transport in the gas-liquid boundary layer, the most general film-penetration theory (Toor and Marcello, 1958) has been applied, which contains the film ($s \to 0$) and the surface renewal theories ($\delta \to \infty$) as limiting cases. The external mass-transfer coefficient is an important factor for the pseudo-homogeneous model. In this case the hydrodynamic effect (β_h) has also been taken into account, thus $\beta_{cd}^o = DS/\delta_d + \beta_h$ where $\beta_h = D/\delta_d$ for the bulk phase or $\beta_h = (D/\delta_d)s_{cd}^o/th(s_{cd}^o)$ for the boundary layer at the gas-liquid interface. The value of β_h cannot be neglected for larger particles (Nagy et al., 1988). A more exact value for β_{cd}^o cannot be obtained by the pseudo-homogeneous model. The so-called heterogeneous model discussed by Nagy (1995) overcomes this problem.

The mass-transfer rate has also been defined for this case, if the gas bubbles (or solid particles) are small compared to the thickness of the boundary layer around them, $d_b < 200\ \mu$m, and thus the spherical effect is no longer negligible. This case can easily occur especially with a solid phase as a third phase.

Due to its lesser importance (Mehra et al., 1988), the Bovian diffusion of particles in the boundary layer has been neglected. This type of particle movement has significant effect on mass-transfer rate only in special cases.

The pseudo-homogeneous model presented enables calculation of the rate of absorption into liquids containing dispersed drops or solid catalyst particles under all important mass-transfer and reaction kinetic conditions.

Theory

The concentration profile of a dissolved gas (or solid) component is illustrated in Figure 1a. It is assumed that the particles in the gas-liquid boundary layer are motionless, and thus their residence time is the same as that for the surface element in the boundary. The concentration distribution of the droplets in the boundary layer differs from that of drops in the bulk phase. Both values have to be determined.

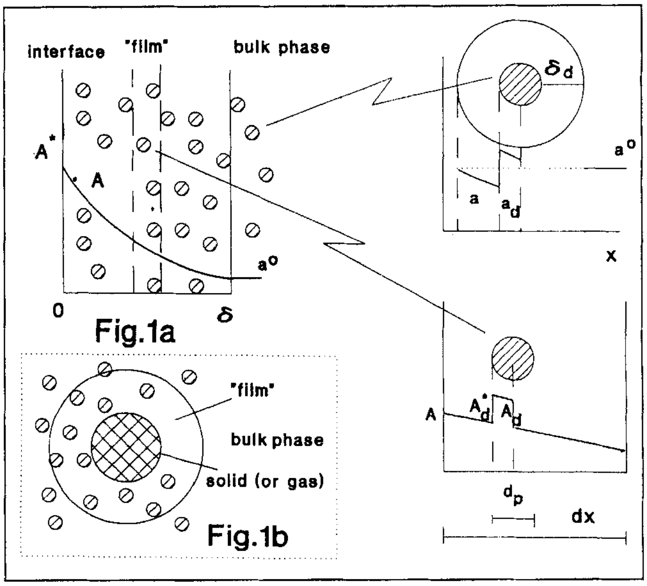

Figure 1. Concentration of the absorbed component *A* in the presence of a dispersed phase (1a) and the spherical boundary layer for small bubble (1b).

The differential mass balance equation for boundary layers at a planar gas-liquid (or solid-liquid) interface can be:

$$D\frac{\partial^2 A}{\partial x^2} - Q - J_d\frac{\omega}{1-\epsilon} = \frac{\partial A}{\partial t} \tag{1}$$

with initial and boundary conditions:

$$\begin{aligned} &\text{if } t=0, \quad x>0 \quad \text{then} \quad A=a^o \\ &\text{if } t>0, \quad x=0 \quad \text{then} \quad A=A^* \\ &\text{if } t>0, \quad x=\delta \quad \text{then} \quad A=a^o \end{aligned} \tag{2}$$

The function J_d provides the specific mass-transfer rate into the droplets at the gas-liquid boundary layer. To define its value, first the mass-transfer rate into particles in the bulk liquid phase, j, has to be given. Then, the mass-transfer rate J_d, as well as the absorption rate J, are given for the following cases: (1) in absence of chemical reactions in the phases; (2) the accompanying reaction is a first-order one; (3) the accompanying reaction is a zero-order one. These are the most important reactions for both chemical and biochemical processes and represent limiting cases for the Michaelis-Menten kinetics.

Mass-transfer rate into particles in the bulk-liquid phase, j

To obtain the value of j, the concentration distribution of a dissolved gas component in the boundary layer between particles and the bulk phase as well as in the drops must be given. If there is no reaction in the dispersed phase, the concentration can be regarded uniform because of the long residence time in the bulk phase ($a_d^o = \mathrm{H}a^o$). Due to the large residence time of drops in the bulk, the mass-transfer process in the dispersed phase is regarded as a steady-state one. Similarly, the mass transport in the boundary layer is described by the film theory. Thus, the mass balance for the inside of drops or for the spherical boundary layer around the drops can be given as follows, with boundary conditions for drops and boundary layer given by Eqs. 4 and 5, respectively:

$$D\left(\frac{\partial^2 a}{\partial r^2}+\frac{2}{r}\frac{\partial a}{\partial r}\right)-Q=0 \tag{3}$$

$$\begin{aligned} &\text{if } r=R \quad a=a_d^* \\ &\text{if } r=0 \quad \frac{da}{dr}=0 \end{aligned} \tag{4}$$

and

$$\begin{aligned} &\text{if } r=R \qquad a=a^* \\ &\text{if } r=R+\delta_d \quad a=a^o \end{aligned} \tag{5}$$

First-Order Reaction ($Q=k_{1d}a_d$ *and/or* $Q=k_1a$). Solving differential Eq. 3 with boundary conditions 4, the concentration for drops can be:

$$a_d(r)=\frac{a_d^*R}{r}\,\frac{sh\left(r\sqrt{\frac{k_{1d}}{D_d}}\right)}{sh\;k_d} \tag{6}$$

where

$$k_d=\sqrt{\frac{k_{1d}R^2}{D_d}}$$

The concentration of dissolved species on the particle surface (a_d^*) is also affected by the external mass-transfer rate, that is, by the mass transfer between the continuous and bulk phases. To quantify this effect Eq. 3 has to be solved with boundary conditions 5. The mass-transfer rate from the bulk into the particles at the interface of the particles (j) can be provided by Eq. 7. Note that this mass-transfer rate is given for the inner edge of boundary layer around dispersed particles (Nagy and Ujhidy, 1989). Therefore, the values of β_{cd} and j decrease rapidly with increasing reaction rate or with increasing film thickness (δ). While the value of T_1 increases exponentially with reaction rate and δ, the product of T_1a^* increases as well.

$$j=D\frac{da}{dr}\bigg|_{r=R}=\beta_{cd}(a^o-T_1a^*) \tag{7}$$

where

$$\beta_{cd}=\frac{D}{\delta_d}\,(1+S)\,\frac{k_{cd}}{sh(k_{cd})} \tag{8}$$

$$T_1=\frac{D}{\delta_d}\left(\frac{k_{cd}}{sh(k_{cd})}+S\right)\frac{1}{\beta_{cd}} \tag{9}$$

and

$$k_{cd}=\sqrt{\frac{k_1\delta_d^2}{D}},\quad \delta_r=\frac{\delta}{\delta_d},\quad S=\frac{\delta_d}{R}$$

From Eq. 6 the value of j at the drop interface can also be given as a function of the internal concentration gradient:

$$j=D_d\frac{da_d(r)}{dr}\bigg|_{r=R}=\beta_d a_d^* \tag{10}$$

where

$$\beta_d=\frac{D}{\delta_d}\frac{S}{D_r}\left(\frac{k_d}{th\;k_d}-1\right) \tag{11}$$

Substituting Eq. 10 into Eq. 7, the interfacial concentrations (a^*, a_d^*, $a_d^*=Ha^*$) can be eliminated, and the value of j can be formulated as a function of a measurable concentration (a^o):

$$j=\beta_1Ha^o \tag{12}$$

where

$$\beta_1=\frac{1}{\frac{H}{\beta_{cd}}+\frac{T_1}{\beta_d}}=\frac{1}{H}\frac{1}{\left(\frac{1}{\beta_{cd}}+\frac{T_1}{\beta_dH}\right)} \tag{13}$$

The value of a_d^* is also needed for quantifying the initial concentration distribution of drops coming from the bulk liquid phase into the gas-liquid boundary layer. From Eqs. 7, 10 and 13 a_d^* can be expressed as:

$$a_d^*=\frac{\beta_1Ha^o}{\beta_d} \tag{14}$$

Equations 6 to 13 provide the mass-transfer rate between drops and the continuous bulk phase in a general case, that is, when a chemical reaction can take place in both phases. Particular cases can easily be obtained from this, as shown later.

Zero-Order Reaction ($Q=k_{0d}$ *and/or* $Q=k_0$). Solving differential Eq. 3 with conditions 4, the concentration of dissolved species in the droplets can be formulated as:

$$a_d(r)=a_d^*+\frac{k_{0d}r^2}{6D_d}-\frac{k_{0d}R^2}{6D_d} \tag{15}$$

The mass-transfer rate through the particle-liquid boundary layer, after solving Eq. 3 with conditions 5, is given by Eq. 16. The same mass-transfer rate is defined by Eq. 17 expressed by the internal concentration of drops. As can be seen, its value increases linearly with particle size and, as expected, it is independent of the internal concentration.

$$\begin{aligned} j=D\frac{da}{dr}\bigg|_{r=R}&=\frac{D}{\delta_d}\,(a^o-a^*)\,(1+S)+\frac{k_0R}{6}\,S(3+S) \\ &=\beta_{cd}^o(a^o-a^*)+\frac{D}{\delta_d}\,k_{cd}(3+S)A^* \end{aligned} \tag{16}$$

where

$$k_{cd}=\sqrt{\frac{k_0\delta_d^2}{DA^*}}$$

and

$$j=\frac{k_{0d}R}{3}=\frac{D}{\delta_d}k_d^2\frac{SA^*}{3D_r} \tag{17}$$

where

$$k_d=\sqrt{\frac{k_{0d}R^2}{D_dA^*}}$$

The value of a_d^* can be expressed from Eqs. 16 and 17 as:

$$a_d^*=Ha^*=Ha^o+T_2HA^* \tag{18}$$

where

$$T_2=\frac{S}{3(1+S)}\left\langle\frac{1+3S+S^2}{S^2}k_{cd}^2-\frac{k_d^2}{D_r}\right\rangle. \tag{19}$$

Mass-transfer rate into particles, J_d, in the gas-liquid boundary layer

According to Eq. 1, the mass transport in the gas- (or solid-) liquid boundary layer is unsteady. The differential mass balance equation for the drops in this layer, as well as the initial and boundary conditions are:

$$D_d\left(\frac{\partial^2A_d}{\partial r^2}+\frac{2}{r}\frac{\partial A_d}{\partial r}\right)-Q=\frac{\partial A_d}{\partial t} \tag{20}$$

$$\begin{array}{llll} \text{if } t=0, & \text{at all } r & \text{then} & A_d=a_d(r) \\ \text{if } t>0, & r=R & \text{then} & A_d=A_d^* \\ \text{if } t>0, & r=0 & \text{then} & \dfrac{\partial A_d}{\partial r}=0 \end{array} \tag{21}$$

The concentration drop across the boundary layer in this particle-size range could be neglected because $d_p \ll \delta$. Thus, the value of A_d^* could be considered as a constant around the particle. Equation 20 is usually solved by means of Laplace transform. Transforming it with respect to time the equation obtained is:

$$\frac{d^2\overline{A}_d}{dr^2}+\frac{2}{r}\frac{d\overline{A}_d}{dr}-\frac{\overline{Q}}{D_d}=\frac{s\overline{A}_d}{D_d}-\frac{a_d(r)}{D_d} \tag{22}$$

In the case of both first- and zero-order reactions, Eq. 22 can be solved analytically, and the time-averaged value of the mass-transfer rate (J_d) can be obtained by Eq. 23 (Froment and Bischoff, 1979):

$$J_d=J_d(t)se^{-st}dt=D_ds\left.\frac{d\overline{A}_d}{dr}\right|_{r=R} \tag{23}$$

First-Order Reaction ($Q=k_{1d}A_d$ and/or $Q=k_1A$). The Laplace transform of the differential equation for the drops in the boundary layer given by Eq. 24 can be solved with known mathematical methods. Solving it with boundary conditions

$$\frac{d^2\overline{A}_d}{dr^2}+\frac{2}{r}\frac{d\overline{A}_d}{dr}-\frac{k_{1d}}{D_d}\overline{A}_d=\frac{s\overline{A}_d}{D_d}-\frac{a_d^*R}{D_dr}\frac{sh\left(r\sqrt{\dfrac{k_{1d}}{D_d}}\right)}{sh\ k_d} \tag{24}$$

$$\begin{array}{lll} \text{if } r=R & \text{then} & \overline{A}_d=\dfrac{A_d^*}{s} \\ \text{if } r=0 & \text{then} & \dfrac{d\overline{A}_d}{dr}=0 \end{array} \tag{25}$$

the mass transfer rate at the surface of the drops can be:

$$J_d=B_d(A_d^*-a_d^*)+\beta_da_d^*=B_d(A_d^*-a_d^*)+j \tag{26}$$

where

$$B_d=\frac{D}{\delta_d}\frac{S}{D_r}\left(\frac{\lambda_d}{th\ \lambda_d}-1\right) \tag{27}$$

$$\lambda_d=R\sqrt{\frac{k_{1d}+s}{D_d}}=s_d^o\sqrt{1+\left(\frac{k_d}{s_d^o}\right)^2} \tag{28}$$

and

$$s_d^o=\frac{\sqrt{sR^2}}{D_d}=s_{cd}^o\frac{\sqrt{D_r}}{S} \tag{29}$$

If there is no reaction in the drops, the value of the physical mass-transfer rate can be:

$$J_d^o=B_d^oA_d^* \tag{30}$$

with

$$B_d^o=\frac{D}{\delta_d}\frac{S}{D_r}\left(\frac{s_d^o}{th\ s_d^o}-1\right) \tag{31}$$

Taking into account also the external mass-transfer resistance

$$B_{cd}^o=\frac{D}{R}+\beta_h=\frac{D}{\delta_d}\left(S+\frac{s_{cd}^o}{th\ s_{cd}^o}\right) \tag{32}$$

around the droplets in the boundary layer (Eq. 32), the value of J_d is:

$$J_d=B_1HA-B_1\left(a_d^*-\frac{j}{B_d}\right)=B_1HA-B_1\left(\frac{\beta_1}{\beta_d}-\frac{\beta_d}{B_d}\right)Ha^o \tag{33}$$

where

$$B_1=\frac{1}{\frac{H}{B^o_{cd}}+\frac{1}{B_d}} \tag{34}$$

The value of J_d given by Eq. 33 is a time-averaged one. It is zero in the stagnant film ($s=0$) and it increases with increasing surface renewal frequency. Substituting it into Eq. 1, the development of the concentration profile of the continuous phase of the layer is taken into account by this average value. The pseudo-homogeneous model does not allow a more exact solution; only the heterogeneous model could make it (Nagy, 1995).

Zero-Order Reaction ($Q=k_{0d}$ and/or $Q=k_0$). The Laplace transform of the differential mass balance equation is:

$$\frac{d^2\overline{A}_d}{dr^2}+\frac{2}{r}\frac{d\overline{A}_d}{dr}-\frac{k_{0d}}{s}=s\overline{A}_d-\left(a^*_d-\frac{k_{0d}}{3D_d}\langle R^2-r^2\rangle\right) \tag{35}$$

Solving Eq. 35 with boundary conditions 25, the mass-transfer rate at the droplet surface can be:

$$J_d=B^o_d(A^*_d-a^*_d)+\frac{k_{0d}R}{3}=B^o_d(A^*_d-a^*_d)+j \tag{36}$$

It is interesting to note that the value of J_d is not influenced by the reaction taking place in the droplets of the gas-liquid boundary layer. This means that the concentration distribution of droplets obtained in the bulk liquid phase does not change during their stay in the boundary layer.

If the reaction rate is fast enough, the absorbed component reacts completely in the bulk phase, that is, $a^o=0$ as well as $a_d(r)=0$. Solving differential Eq. 35 for that case (the second term of the righthand side of Eq. 35 will be equal to zero), the value of J_d can be expressed as:

$$J_d=B^o_d\left(1+\frac{k_{0d}}{sA^*_d}\right)A^*_d=B^o_d\left(1+\left\langle\frac{k_d}{s^o_d}\right\rangle^2\right)A^*_d \tag{37}$$

If $a_d(r)=a^o_d=$ constant, then for J_d it is:

$$J_d=B^o_d\left(1+\left\langle\frac{k_d}{s^o_d}\right\rangle^2-\frac{a^o_d}{A^*_d}\right)A^*_d \tag{38}$$

The value of J_d with the external mass-transfer coefficient can be provided by Eq. 39a or, using Eq. 37, by Eq. 39b:

$$J_d=B^oHA-B^o\left(a^*_d-\frac{j}{B^o_d}\right) \tag{39a}$$

$$J_d=\frac{HA}{\frac{H}{B^o_{cd}}+\frac{1}{B^o_d\left(1+\left\langle\frac{k_d}{s^o_d}\right\rangle^2\right)}}=B_0HA \tag{39b}$$

where

$$B^o=\frac{1}{\frac{H}{B^o_{cd}}+\frac{1}{B^o_d}} \tag{40}$$

Absorption rate at gas- (or solid-) liquid interface, J

To give the mass-transfer rate at the gas-liquid interface, the differential mass balance Eq. 1 has to be solved with initial and boundary conditions 2. The values of J_d, in general cases of first- and zero-order reactions, are defined by Eqs. 33, 39a and 39b, respectively. Thus, the Laplace transform of differential Eq. 1 for a first-order chemical reaction is:

$$D\frac{d^2\overline{A}}{dx^2}-\left(k_1+s+B_1H\frac{\omega}{1-\epsilon}\right)\overline{A}=-\left(a^o+\frac{B_1\omega}{(1-\epsilon)s}\left\langle a^*_d-\frac{j}{B_d}\right\rangle\right) \tag{41}$$

Taking Eqs. 12 and 14 into account for values j and a^*_d, the absorption rate for a first-order reaction is:

$$J=\frac{D}{\delta}\frac{\lambda_1}{th\,\lambda_1}A^*\left\{1-\frac{a^o}{A^*}\left(\frac{1}{ch\,\lambda_1}+\frac{(s^o)^2}{\lambda_1^2}\times\left[1+\frac{M'^2\beta_1H}{(th\,s^o)^2}\left\langle\frac{1}{\beta_d}-\frac{1}{B_d}\right\rangle\right]\left(1-\frac{1}{ch\,\lambda_1}\right)\right)\right\} \tag{42}$$

where

$$\lambda_1=s^o\sqrt{1+\frac{k_1}{s}+\frac{B_1H\omega}{s(1-\epsilon)}}=s^o\sqrt{1+\frac{M^2+M'^2H}{(th\,s^o)^2}} \tag{43}$$

$$M=\frac{\sqrt{k_1D}}{\beta^o}=\sqrt{\frac{k_1D}{sD}}\;th\,s^o \tag{44}$$

$$M'=\frac{\sqrt{\frac{B_1D\omega}{(1-\epsilon)}}}{\beta^o}=\frac{\sqrt{\frac{D}{\delta_d}3B_1S\frac{\epsilon}{(1-\epsilon)}}}{\beta^o} \tag{45}$$

and

$$s^o=\sqrt{\frac{s\delta^2}{D}}$$

For zero-order reactions the Laplace transform of the mass balance equation is defined by Eq. 46. Solving it with boundary conditions 2, the mass-transfer rate at the gas-liquid interface

$$D\frac{d^2\overline{A}}{dx^2}-B^o\frac{\omega H}{(1-\epsilon)}\overline{A}-s\overline{A}=\frac{k_0}{s}-B^o\frac{\omega}{s(1-\epsilon)}\left(a^*_d-\frac{j}{B^o_d}\right)-a^o \tag{46}$$

is provided by Eq. 47a, while in the case of $a^o/A^*=0$ (using Eq. 39b), by Eq. 47b:

$$J=\frac{D}{\delta}\frac{\lambda_0A^*}{th\,\lambda_0}\left\{1-\frac{a^o}{A^*}\left[\frac{1}{ch\,\lambda_0}+\frac{(s^o)^2}{\lambda_0^2}\left\langle1-\frac{M^2A^*}{(th\,s^o)^2a^o}+\frac{M'^2}{(th\,s^o)^2}\left(H+T_2H\frac{A^*}{a^o}-\frac{j}{B^o_da^o}\right)\right\rangle\left(1-\frac{1}{ch\,\lambda_0}\right)\right]\right\} \tag{47a}$$

where

$$\lambda_0 = s^o\sqrt{1+\frac{B^oH\omega}{s(1-\epsilon)}} = s^o\sqrt{1+\frac{M'^2H}{(th\ s^o)^2}} \tag{48a}$$

$$M' = \frac{\sqrt{\dfrac{B^oD\omega}{(1-\epsilon)}}}{\beta^o} = \frac{\sqrt{\dfrac{D}{\delta_d}\,3B^oS\,\dfrac{\epsilon}{(1-\epsilon)}}}{\beta^o} \tag{49}$$

and

$$M = \frac{\sqrt{\dfrac{k_0D}{A^*}}}{\beta^o} \tag{50}$$

$$J = \frac{D}{\delta}\frac{\lambda_0}{th\ \lambda_0}\left\{1+\frac{(s^o)^2}{\lambda_0^2}\frac{M^2}{(th\ s^o)^2}\left(1-\frac{1}{ch\ \lambda_0}\right)\right\}A^* \tag{47b}$$

where

$$\lambda_0 = s^o\sqrt{1+\frac{B_0H\omega}{s(1-\epsilon)}} \tag{48b}$$

The values of j and T_2 are expressed by Eqs. 17 and 19, respectively. If a reaction occurs neither in the continuous nor in the dispersed phase, the mass-transfer rate can be given from Eq. 42 or 47:

$$J^o = \frac{D}{\delta}\frac{\lambda_0}{th\ \lambda_0}\left[1-\frac{a^o}{A^*}\left\langle\frac{1}{ch\ \lambda_0}+\frac{(s^o)^2}{\lambda_0}\right.\right.$$
$$\left.\left.\times\left(1+\frac{M'^2H}{(th\ s^o)^2}\right)\left(1-\frac{1}{ch\ \lambda_0}\right)\right\rangle\right]A^* \tag{51}$$

Absorption rate at spherical gas- (or solid-) liquid interface, J_b

In common industrial processes the size of gas bubbles going through the liquid-liquid phase is larger than 100-200 μm. In this size range the value of δ/R_b is rather small and thus the spherical effect on the gas-liquid mass transfer can be neglected. However, if the third phase is a solid one (Mehra et al., 1988), the size of the particle can be smaller than the value of δ. If the particle size is of the same order of magnitude as the "film-thickness" is at the gas- (or solid-) liquid interface (Figure 1b), the spherical effect can no longer be neglected (Nagy et al., 1986). To define this effect, the mass balance (Eq. 52) has to be solved with initial and boundary conditions 53:

$$D\left(\frac{\partial^2A}{\partial r^2}+\frac{2}{r}\frac{\partial A}{\partial r}\right)-Q-J_d\frac{\omega}{1-\epsilon}=\frac{\partial A}{\partial t} \tag{52}$$

$$\begin{aligned} &\text{if}\quad t=0,\quad r>=R_b\quad \text{then}\quad A=a^o\\ &\text{if}\quad t>0,\quad r=R_b\quad \text{then}\quad A=A^*\\ &\text{if}\quad t>0,\quad r=R_b+\delta\quad \text{then}\quad A=a^o \end{aligned} \tag{53}$$

This equation can be solved with a similar method to that used for Eq. 1. The mass-transfer rate obtained for first- and zero-order reaction is expressed by Eqs. 54 and 56, respectively:

$$J = \frac{D}{\delta}\left(S_b+\frac{\lambda_1}{th\ \lambda_1}\right)\left\{1-\frac{a^o}{A^*}\left(\frac{1+S_b}{\dfrac{sh\ \lambda_1}{\lambda_1}S_b+ch\ \lambda_1}\right.\right.$$
$$\left.\left.+\frac{(s^o)^2}{\lambda_1^2}\left[1+\frac{M'^2\beta_1H}{(th\ s^o)^2}\left\langle\frac{1}{\beta_d}-\frac{1}{B_d}\right\rangle\right]T_3\right)\right\}A^* \tag{54}$$

where

$$T_3 = \frac{\dfrac{S_b}{\lambda_1}+ch\ \lambda_1-(1+S_b)}{\dfrac{sh\ \lambda_1}{\lambda_1}S_b+ch\ \lambda_1} \tag{55}$$

$$J = \frac{D}{\delta}\left(S_b+\frac{\lambda_0}{th\ \lambda_0}\right)\left\{1-\frac{a^o}{A^*}\left(\frac{1+S_b}{\dfrac{sh\ \lambda_0}{\lambda_0}S_b+ch\ \lambda_0}+\frac{(s^o)^2}{\lambda_0^2}\right.\right.$$
$$\times\left[1-\frac{M^2A^*}{(th\ s^o)^2a^o}+\frac{M'^2}{(th\ s^o)^2}\left\langle H+T_2H\frac{A^*}{a^o}\right.\right.$$
$$\left.\left.\left.\left.-\frac{j}{B_d^oa^o}\right\rangle\right]T_3\right)\right\}A^* \tag{56}$$

where

$$T_3 = \frac{\dfrac{S_b}{\lambda_0}+ch\ \lambda_0-(1+S_b)}{\dfrac{sh\ \lambda_0}{\lambda_0}S_b+ch\ \lambda_0} \tag{57}$$

Results and Discussion

Comparison between literature models and present ones

As has been mentioned, the main supposition of the homogeneous models in the literature is that the mass transport inside dispersed particles is instantaneous (Mehra, 1988). Obviously, this assumption is not fulfilled under all mass-transfer conditions, such as with increasing surface renewal frequency(s), or particle size (d_p). An important advantage of the model presented here is that the mass-transfer rate can also be predicted under extreme conditions, independently of whether there is a chemical reaction.

$$D\frac{\partial^2A}{\partial x^2}-k_1A-\frac{\epsilon}{1-\epsilon}\frac{\partial A_d}{\partial t}=\frac{\partial A}{\partial t} \tag{58}$$

Considering mass transport inside the particles as instantaneous, the mass-transfer rate (J_{inst}) at the gas-liquid interface can be obtained in this case solving the differential Eq. 58:

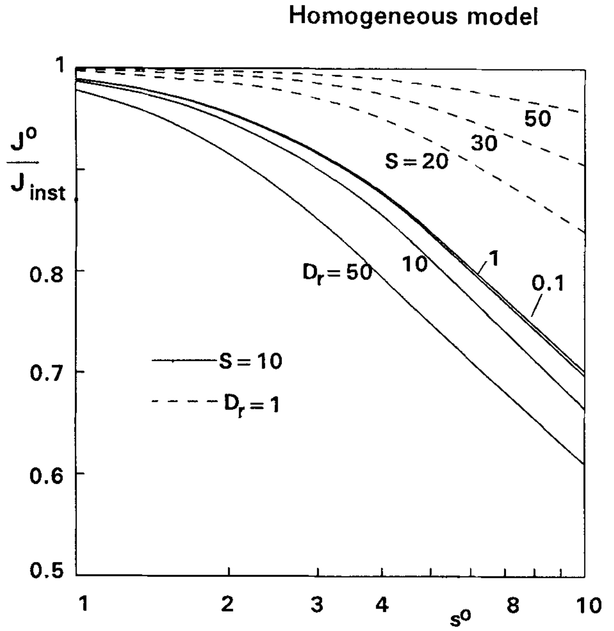

Figure 2. Effect of the dimensionless kinetic constant s^o on the ratio of physical mass-transfer rates: this model (J^o) vs. literature (J_{inst}) model.

$H=10$, $\epsilon=0.3$, $a^o=0$, $M=0$, $k_d=0$, $\delta_r=1$.

$$J_{inst}=\frac{D}{\delta}\frac{\lambda_{inst}}{th\,\lambda_{inst}}\left\{1-\frac{a^o}{A^*}\left(\frac{1}{ch\,\lambda_{inst}}+\frac{(s^o)^2}{\lambda_{inst}^2}\left[1-\frac{1}{ch\,\lambda_{inst}}\right]\right)\right\}A^* \tag{59}$$

where

$$\lambda_{inst}=s^o\sqrt{\frac{1-\epsilon+\epsilon H}{1-\epsilon}+\frac{M^2}{(th\,s^o)^2}} \tag{60}$$

In Figure 2 the ratio of mass-transfer rates calculated from Eqs. 51 and 59 is shown in the function of s^o for $a^o/A^*=0$ and $M=0$, as well as for different values of S and D_r. Increasing the value of s^o, the value of J^o/J_{inst} decreases indicating that the concentration distribution inside the drops is no longer uniform. Thus, Mehra's model is not valid any longer in that parameter regime. The mass-transfer rates obtained for values $s^o<0.3$ correspond to the film theory, while those for $s^o>3$ are identical with mass-transfer rates calculated from the surface renewal theory. The $0.3<s^o<3$ is the range of the so-called intermediate regime. It can be seen from Figure 2 that a rather wide range of s^o belongs to the value of $J^o/J_{inst}<1$. Both the particle size and the diffusivity ratio (D_r) have strong effects. For example, in the case of the surface renewal theory, $J^o/J_{inst}<1$ when $S<$cca. 30, which indicates a rather small particle size. Provided that the value of δ is between 20 to 60 μm, the intraparticle diffusion influences the absorption rate from the particle size of already $d_p>1.4-4$ μm. The value of H has a similar effect on J^o/J_{inst} than that of s^o (not figured here), such as for $H=10$ and $S=50$, its value decreases with s^o down to 0.95 (Figure 2), while for $H=1{,}000$ down to 0.68. Thus, the value of H can further reduce the particle size where $J^o/J_{inst}<1$.

The most important quantity in which the Mehra's model and the present one differ is the internal mass-transfer coefficient of the particles in the gas-liquid boundary layer, B_d, B_d^o. According to Mehra (1988), $B_d^o=2\pi^2D_d/(3d_p)$, that is, it is independent of the residence time in the layer, while in our proposed model defined by Eq. 31, B_d^o depends strongly on the surface renewal frequency and, consequently, on the time (note that the residence time $t=1/s$). In the limiting case, when s^o tends to become zero then $B_d^o\rightarrow 0$. The value of $(s_d^o/th\,s_d^o-1)$ in Eq. 31 changes by more than four orders of magnitude in the s^o range of 0.1 and 10 ($s_d^o=s^o\sqrt{D_r/S/\delta_r}$), thus the value of B_d^o changes accordingly. Another important point is that unlike Mehra's model, B_d (given by Eq. 27) also involves the effect of internal chemical reaction on the mass-transfer coefficient. The absorption rate in Mehra's model is given by Eq. 20a of his article with the use of the surface renewal theory (Mehra, 1988). In order to compare it with the J value of the present model, J, B_d, B_{cd}^o should be expressed using the surface renewal theory. In the case of $a^o=0$, the value of J is:

$$\text{if } \delta\rightarrow\infty \text{ then } \lim J=\sqrt{Ds(1+M^2+M'^2H)}A^* \tag{61}$$

with

$$B_d=\frac{2D_d}{d_p}\left(\frac{\lambda_d}{th\lambda_d}-1\right) \tag{62}$$

and

$$B_{cd}^o=\frac{2D}{d_p}+\sqrt{Ds} \tag{63}$$

The ratio of mass-transfer rates of the two models without reaction is shown in Figure 3 in the function of physical mass-transfer coefficient at the gas-liquid interface for different d_p and H values. With the increase of the surface renewal frequency (that is, the β^o value), the value of J^o/J_M^o increases as well. The effect of the particle size on the J^o/J_M^o ratio in the 2 to 8 μm range is rather low, (a few 10%). The solubility coefficient, H, has a strong effect indicating the important role of mass transport inside the particles.

The change of J/J_M ratio in the function of the internal reaction rate, more exactly in function of k_d, is shown in Figure 4 for different values of d_p, H and β^o. It can be seen that the difference between the two models can be very significant indicating the fact that the internal reaction can strongly alter the concentration distribution of fine particles in the boundary layer, even very low k_d values ($k_d\ll 1$). This is the consequence of the short residence time in the layer. As is well-known, the particle concentration is considered as constant, and it is in equilibrium with that of the continuous phase when $k_d>1$ [$k_d=\sqrt{k_{1d}R^2/D_d}$, that is, k_d is identical with the Thiele-modulus for steady-state mass transfer (Satterfield, 1969)]. Naturally, this limiting value decreases with decreasing contact time. The internal reaction rate constant changes between 3.2 and 320 1/s with $D_d=2\times10^{-9}$ m^2/s and $d_p=5$ μm in the k_d range of 0.1 to 1. Diffusivity in a catalyst particle can be even an order of magnitude lower. Thus k_{1d} will also become lower in the same manner in the above k_d range, such as the k_{1d} values for the hydrogenation of α-methyl-styrene on palladium-alu-

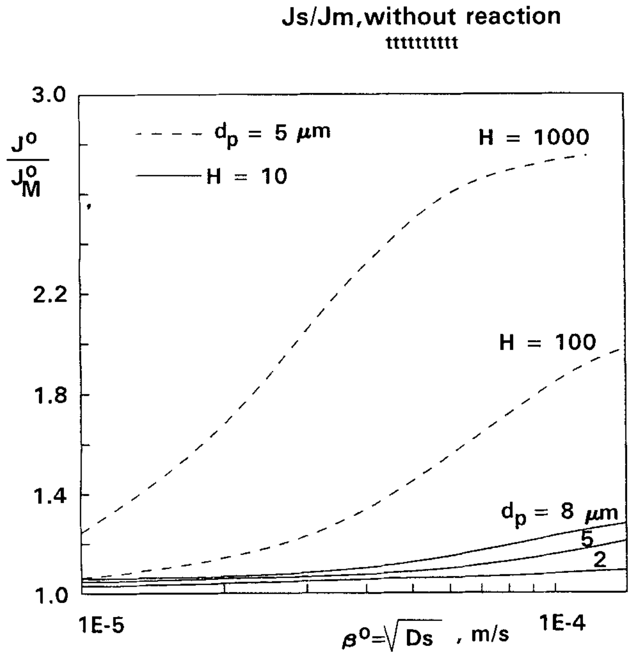

Figure 3. Change of J^o/J^o_M in function of the mass-transfer coefficient using the surface renewal theory.

$D = 2\times10^{-9}$ m²/s, $D_r = 1$, $\epsilon = 0.1$, $a^o = 0$, $M = 0$, $k_d = 0$, $\delta_r = 1$.

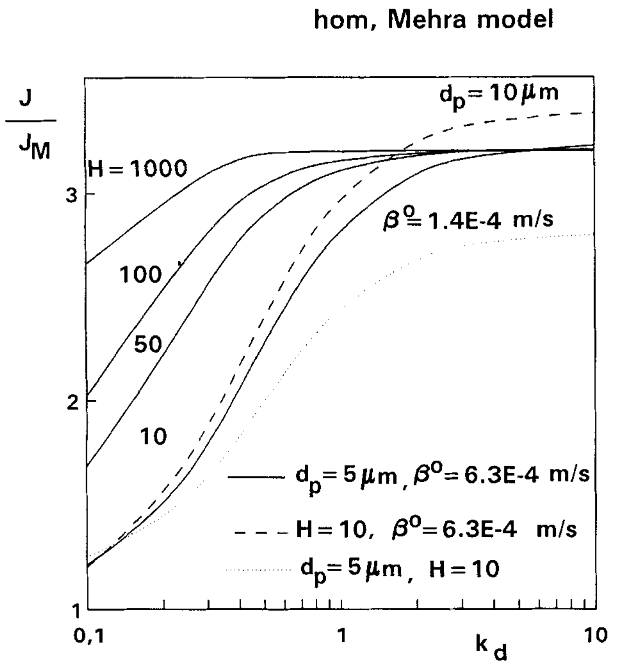

Figure 4. Change of J/J_M in the function of the internal reaction rate.

$\beta^o = \sqrt{Ds}$, $D = 2\times10^{-9}$ m²/s, $D_r = 1$, $\epsilon = 0.1$, $a^o = 0$, $M = 0$, $k_d = 0$, $\delta_r = 1$.

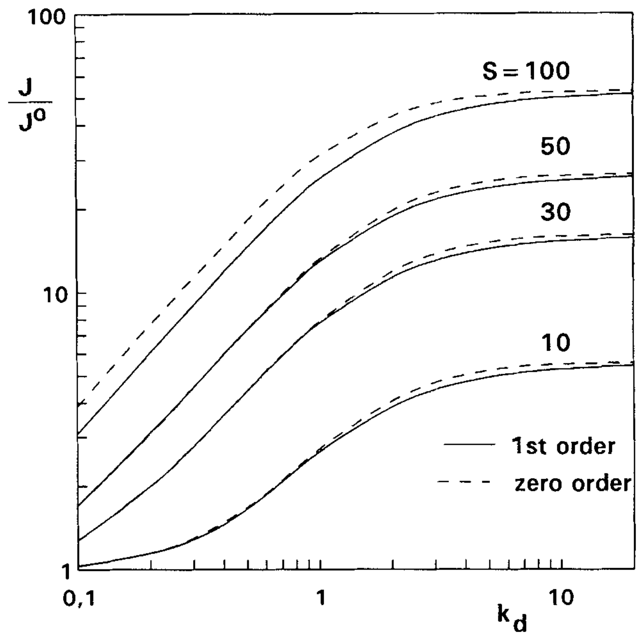

Figure 5. Effect of the dimensionless reaction rate constant k_d on the absorption rate related to that obtained without reaction in phases.

$D_r = 1$, $H = 1$, $\epsilon = 0.1$, $s^o = 0.5$, $\delta_r = 1$, $M = 0$, $a^o = 0$.

mina catalyst powder in a slurry reactor (Satterfield et al., 1969), or for that of aniline (Govindarao and Murthy, 1975) which falls in this range ($k_{1d} = 16.8$ and 2.8 1/s, respectively). Thus, the internal reaction can essentially affect the absorption of hydrogen when very fine catalyst particles are used in the process. Similar effects can appear in other petrochemical processes such as oxidation and amination.

Comparison between a first- and zero-order reaction

The effect of these reactions on the absorption rate is illustrated in Figure 5. Since $H = 1$ and $D_r = 1$ (parameters are listed in the legends), the dispersed phase without chemical reaction has no effect on the absorption rate; thus, $J/J^o \to 1$ if $k_d \to 0$. The value of a^o/A^* was chosen to be zero. Thus, the second term (in parenthesis) of Eq. 42 is zero. The zero-order reaction rate is independent of the absorbate concentration; therefore, these terms of Eqs. 47a and 47b will not be zero. The mass-transfer rate has been calculated by Eq. 47b for zero-order reaction (broken lines in the figure). The value of J/J^o approaches a limit with increasing reaction rate depending on the value of S. These limiting values are determined by the external mass-transfer coefficient between continuous and dispersed liquid phase. Thus, the maximum value of J_d, J_d^{max}, is:

$$J_d^{max} = B^o_{cd} A^* \tag{64}$$

Taking into account J_d^{max} in Eqs. 42 and 47a or 47b, the maximum value of the absorption rate, J^{max} can be given by Eq. 65 for a first-order reaction and by Eq. 66 for the zero-order case (a^o is considered to be zero):

$$\text{if } k_d \to \infty \text{ then } \lim J = J^{max} = \frac{D}{\delta}\frac{\lambda_1}{th\,\lambda_1} A^* \tag{65}$$

$$J^{max} = \frac{D}{\delta}\frac{\lambda_0}{th\,\lambda_0}\left\{1+\frac{(s^o)^2}{\lambda_0^2}\frac{M^2}{(th\,s^o)^2}\left(1-\frac{1}{ch\,\lambda_0}\right)\right\}A^* \tag{66}$$

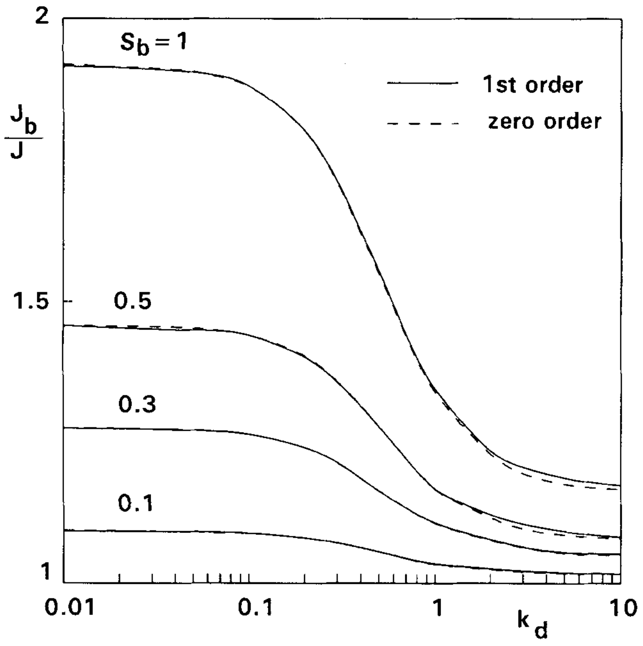

Figure 6. Change in the ratio of spherical (J_b) and planar (J) mass-transfer rates at gas-liquid (or solid-liquid) interface as a function of the reaction rate at different bubble (or solid particle) sizes.

$S = 10$, $D_r = 1$, $H = 1$, $\epsilon = 0.1$, $s^o = 0.5$, $\delta_r = 1$, $M = 0$, $a^o = 0$.

In accordance with Eq. 65, the limiting values of the lines in Figure 5 represent J values equal to J^{max}. Obviously, these are the limiting values for both cases, namely for mass transfer accompanied by first- or zero-order reactions. As can be seen in the figure, the spherical effect, S, which is practically the effect of the particle size, has also a strong effect on the enhancement. The reason for this is that with the increase of S, the value of k_d, as well as the value of the external mass-transfer coefficient B_d^o, also increases, and as a consequence the value of B_d and J increase more rapidly than those of B_d^o and J^o. J^{max}/J^o also changes very strongly with increasing S. In general, it can be stated that there is no significant difference between the effects of a first or zero-order reactions.

The diffusivity ratio, D_r, the solubility coefficient, H, and the dispersed phase holdup, ϵ, also play a significant role (not considered here) in the three-phase mass transfer. At a given internal reaction rate the J/J^o value increases with the decrease of D_r or the increase of ϵ. The J/J^o ratio has a maximum value with the increase of H. The chemical reaction dominates the J/J^o value in the rising part of the function, while in the falling part the external mass-transfer resistance predominates.

For the sake of completeness the spherical effect of the gas bubble (or solid particles) has also been expressed by Eqs. 54 and 56 for first- and zero-order reactions, respectively. This effect can be of practical importance only if the third phase is a solid. When gas is third-phase in current industrial processes, the bubble size is mostly larger than 200-300 μm, and in this size range the spherical effect is negligible. The particle size can have a substantial effect on the mass-transfer rate between gas and continuous liquid phases only if R_b is less than about 100 μm. Figure 6 illustrates the effect of reaction rate in the dispersed liquid phase if $S_b \leq 1$. The perpendicular axis contains the ratio of mass-transfer rates with spherical ($S_b > 0$) and planar ($S_b = 0$) interface. The value of J_b/J decreases monotonically with k_d in the cases of both chemical reactions. These values are equal to each other in both limiting value of k_d. These limits are given by Eqs. 68 and 69 (the value of λ_0 is defined by Eq. 48 while λ_1 by Eq. 43 with $B_d = B_{cd}^o/H$ and $M = 0$ for this case). The maximum value of J_b/J depends strongly on the spherical effect (S_b). From the figure,

$$\text{if } k_d \rightarrow 0 \text{ then } \lim \frac{J_b}{J} = \frac{S_b + \dfrac{\lambda_0}{th\,\lambda_0}}{\dfrac{\lambda_0}{th\,\lambda_0}} \tag{68}$$

$$\text{if } k_d \rightarrow \infty \text{ then } \lim \frac{J_b}{J} = \frac{S_b + \dfrac{\lambda_1}{th\,\lambda_1}}{\dfrac{\lambda_1}{th\,\lambda_1}} \tag{69}$$

it can be stated that the spherical effect of gas bubble (or solid particles) is not negligible if the value of S_b is larger than about 0.1.

Verification of the calculated results with experimental data

Mehra et al. (1988) have very extensively experimentally investigated the effect of dispersed second liquid phase on the absorption rate. As a dispersed phase, they used macroemulsions ($d_p = 1 - 12$ μm) and microemulsions with drop size less than 1 μm. The absorption of organic gases and hydrolysis of solid organic esters were used for experiments. The surface renewal theory was applied to evaluate the experimental data. Therefore, to compare the data of the present model to their experimental results, the model equations according to the surface renewal theory (Eqs. 61-63) were used for the calculation. In Tables 1a and 1b the results calculated by the present

Table 1a. Absorption Rates of Isobutylene into Macroemulsion of Chlorobenzene in Aqueous Solution of Sulfuric Acid*

		$J \times 10^7$, mol/m$^2\cdot$s			
k_1, 1/s	ϵ	Meas. Data	This Model	Mehra's Model	Data Used
0.32	0	0.37	0.4	0.4	$D = 1.14 \times 10^{-9}$ m^2/s
	0.01	0.79	1.1	1.04	$D_r = 1$
	0.05	2.16	2.37	2.35	$\beta^o = 1.74 \times 10^{-5}$ m/s
	0.2	4.11	5.12	4.66	$A^* = 1.55 \times 10^{-3}$ kmol/m^3
					$H = 1{,}617$
1.45	0	0.61	0.68	0.68	$s = 0.27$ 1/s
	0.2	5.14	5.17	4.6	$B_{cd}^o = 11.57 \times 10^{-4}$ m/s
					$B_d^o = 8.86 \times 10^{-8}$ m/s
4.86	0	1.08	1.18	1.18	$s_d^o = 1.53 \times 10^{-2}$
	0.05	2.03	2.63	2.55	$d_p = 2$ μm
	0.10	3.58	3.60	3.41	
	0.20	5.7	5.3	4.68	
13.0	0	1.7	1.9	1.9	
	0.10	4.25	3.98	3.69	
	0.20	6.58	5.57	4.87	

*The measured data are given in Table 4 of Mehra (1988) ($k_d = 0$, $a^o = 0$).

Table 1b. Absorption Rates of 1-Butene into Macroemulsion of Chlorobenzene in Aqueous Solution of Sulfuric Acid*

		$J \times 10^7$, mol/m$^2 \cdot$s			
k_1, 1/s	ϵ	Meas. Data	This Model	Mehra's Model	Data Used
0.25	0	0.15	0.24	0.24	$D = 6.19 \times 10^{-10}$ m^2/s
	0.02	1.11	1.06	1.06	$D_r = 1$
	0.05	1.88	1.67	1.60	$\beta^o = 1.27 \times 10^{-5}$ m/s
	0.1	2.74	2.40	2.34	$A^* = 1.33 \times 10^{-3}$ kmol/m^3
	0.2	3.91	3.60	3.31	$H = 2{,}014$
					$s = 0.26$ 1/s
3.52	0	0.55	0.64	0.64	$B^o_{cd} = 15.5 \times 10^{-4}$ m/s
	0.2	5.03	3.90	3.17	$B^o_d = 3.47 \times 10^{-8}$ m/s
					$s^o_d = 0.82 \times 10^{-2}$
					$d_p = 0.8$ μm

*Measured data are given in Table 6 in Mehra et al. (1988) ($k_d = 0$, $a^o = 0$).

and Mehra's model, as well as the experimental data of Mehra et al. (1988), are listed for absorption of organic gases into macroemulsion with $dp = 2$ μm (Table 1a) and with 0.8 μm (Table 1b) at different holdup values of the organic phase and reaction rate constants, k_1, for the continuous phase. It is assumed that there is no reaction in dispersed phase ($k_d = 0$) and that $a^o = 0$. The data of Mehra's model were also taken from their article (Mehra et al., 1988). As can be seen in Tables 1a and 1b, the figures obtained from the models are in good agreement with experimental data. Comparing the theoretical data, the following tendency can be observed: in the holdup range investigated the value of J by Mehra's model changes somewhat less than the experimental figure or the one obtained by the present model. For holdup values of 0.1 or 0.2, for example, the present model gives a much better agreement with experimental data than Mehra's one and seems superior.

An an example, the mass-transfer rates into microemulsion ($d_p = 100$ nm) are given for hydrolysis of solid esters in Table 2. Again, the calculated results are in a good agreement with the experimental ones. The tendency of the alteration between the calculated and measured mass-transfer rates is somewhat different, though not significant. The theoretical values are a bit higher for lower ϵ values and a bit lower for higher ones as compared to measured figures.

Janakiraman and Sharma (1985) experimentally studied the effect of fine carbon particles ($d_p = 4.3$ μm) on the mass-transfer rate between solid-liquid and liquid-liquid systems during alkaline hydrolysis of several organic compounds such as ethyl-p-nitrobenzoate (solid-liquid fast reaction) and the oximation of cyclododecanone (solid-liquid slow reaction). The enhancement of these processes is calculated also by Eqs. 61-63. The parameter values given by Janakiraman and Sharma together with the enhancement data are listed in Table 3. Again, the results of this model agree well with experimental data. It proves that the mass-transfer rate can also be correctly predicted by this model in the presence of solid catalytic particles.

Table 2. Mass-Transfer Rates (J) for Alkaline Hydrolysis of 2,4-dichlorophenyl Benzoate in Microemulsions of Chlorobenzene ($d_p = 100$ nm)*

		$J \times 10^9$, mol/m$^2 \cdot$s		
ϵ	H	Meas. Data	This Model	Data Used
0		0.69	0.75	$D = 5.9 \times 10^{-10}$ m^2/s
0.025	7,600	4.8	8.5	$D_r = 1$
0.05	6,340	9.0	11.3	$\beta^o = 9.8 \times 10^{-6}$ m/s
0.1	4,020	15.1	13.1	$A^* = 3.59 \times 10^{-5}$ kmol/m^3
0.14	1,620	17.9	16.9	$d_p = 100$ nm
				$k_1 = 0.58$ 1/s
				$s = 0.16$ 1/s
				$M = 1.89$

*Measured data are taken from Table 8 in Mehra et al. (1988), ($k_d = 0$, $a^o = 0$).

Table 3. Enhancement of the Mass-Transfer Rate in the Presence of Fine Carbon Particles ($d_p = 4.3$ μm)*

	Ethyl p-Nitrobenzoate		Cyclododecanone	
ϵ	Meas. Data	This Model	Meas. Data	This Model
0.01	1.8	1.7	1.66	2.0
0.02	2.3	2.2	2.35	2.7
0.03	2.9	2.6	2.91	3.3
0.04	—	—	3.46	3.8
Data Used:	$D = 3.3 \times 10^{-10}$ m^2/s		$D = 5.8 \times 10^{-10}$ m^2/s	
	$D_r = 1$		$D_r = 1$	
	$\beta^o = \sqrt{Ds} = 6.0 \times 10^{-6}$ m/s		$\beta^o = \sqrt{Ds} = 12.0 \times 10^{-6}$ m/s	
	$s = 0.11$ 1/s		$s = 0.25$ 1/s	
	$k_1 = 1.34$ 1/s		$k_1 = 0.033$ 1/s	
	$k_{1d} = 1.34$ 1/s		$k_{1d} = 0.033$ 1/s	
$H = 274 * 0.31/0.5/0.02 = 8{,}500$			$H = 440$	

*Measured data are taken from Janakiraman and Sharma (1985).

Conclusion

The improved pseudo-homogeneous model presented can be regarded as a general mass-transfer model for the three-phase system, which enables the user to calculate the effect of mass transport and chemical reaction inside the drops and that of particle size on the absorption rate. Both first- and zero-order reaction can considerably influence the absorption rate, and even the internal diffusivity of drops has an important role in the process. Although the particle-size range is very small in which the homogeneous model is really valid ($S \gg 1$), the mass transport in the drops, (especially if it is accompanied by reaction), must not also be regarded as an instantaneous process. The film-penetration theory used also includes the mass-transfer equations for both film and surface renewal theory as a limiting case. The model presented offers a possibility to predict the absorption rate under all important mass-transfer and reaction kinetic parameters.

Data calculated by the model presented are in very good agreement with published experimental data, further supporting the applicability of the model.

Acknowledgment

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Notation

a = concentration of A in the film layer of the particles located in the bulk liquid phase, mol/m^3
a^o = concentration of A in the bulk phase, mol/m^3
a^*, a^*_d = concentration of A at both sides of the interface of the particles located in the bulk liquid phase, $a^*_d = \mathrm{H}a^*$ (Eqs. 14 and 18), mol/m^3

$a_d(r)$, a_d = concentration of A in the dispersed particles in the bulk phase, mol/m^3
a_d^o = average concentration of A in the dispersed particles located in the bulk liquid phase, mol/m^3
A = concentration of component A (absorbed or dissolved) in the gas- (or solid) liquid boundary layer, mol/m^3
A^* = concentration of A at the gas-liquid interface, mol/m^3
A_d = concentration of A in the particles located in the gas-liquid boundary layer, mol/m^3
A_d^* = concentration of A at the interface of the particles located in the gas-liquid boundary layer, mol/m^3
$\overline{A}$, $\overline{A}_d$ = Laplace transform of concentration A and A_d, mol·s/m^3
B^o = physical mass-transfer coefficient with the total resistance (Eq. 40), m/s
B_{cd}^o = external mass-transfer coefficient between the continuous and dispersed phases in the gas-liquid boundary layer defined by Eq. 32, m/s
B_d = internal mass-transfer coefficient of the particles in the gas-liquid boundary layer (Eq. 27), m/s
B_d^o = internal physical mass-transfer coefficient of the particles in the boundary layer (Eq. 31), m/s
B_1, B_0 = mass-transfer coefficients with the total resistance between the continuous and dispersed phases in the gas-liquid boundary layer (Eq. 34) and Eq. 39b, respectively, m/s
ch = cosh
d_b = size of the gas bubbles or solid particles, m
d_p = size of the dispersed particles, m
D = diffusion coefficient of component A for the continuous phase, m^2/s
D_d = diffusivity of A in the dispersed phase, m^2/s
D_r = D/D_d
H = Henry's law constant between the continuous and dispersed phases
j = mass-transfer rate at the liquid-droplets interface in the bulk continuous phase (Eqs. 7 and 12 for first-order reaction and Eqs. 16 and 17 for zero-order one), mol/m^2·s
J = mass-transfer rate at the gas (or solid)-liquid interface, mol/m^2·s
J_b = mass-transfer rate at the spherical gas (or solid)-liquid interface, mol/m^2·s
J_d = mass-transfer rate between the continuous and dispersed phases in the gas (or solid)-liquid boundary layer, mol/m^2·s
J_M = mass-transfer rate at gas-liquid interface given by Mehra (1988), mol/m^2·s
J^o = mass-transfer rate obtained with the dispersed phase without reaction in the phases, mol/m^2·s
J_M^o = mass-transfer rate with the dispersed phase without reaction given by Mehra (1988), mol/m^2·s
$J^{\max}$ = maximum value of J, mol/m^2·s
k_1 = first-order reaction rate constant in the continuous phase, 1/s
k_{1d} = first-order reaction rate constant in the dispersed phase, 1/s
k_0 = zeroth-order reaction rate constant in the continuous phase, mol/m^3·s
k_{0d} = zeroth-order reaction rate constant in the dispersed phase, mol/m^3·s
k_{cd} = $\sqrt{(k_1\delta_d^2)/D_d}$ or $= \sqrt{(k_0\delta_d^2)/D_dA^*}$, dimensionless reaction rate constant for boundary layer around the particles
k_d = $\sqrt{(k_{1d}R^2)/D_d}$ or $= \sqrt{(k_{0d}R^2)/D_dA^*}$, dimensionless reaction rate constant for particles
M = $\sqrt{(k_1D)}/\beta^o$ or $= \sqrt{(k_0D)/A^*}/\beta^o$, reaction-diffusion parameter (Hatta number) for boundary layer at gas-liquid interface
M' = $\sqrt{(B_1D\omega)/(1-\epsilon)}/\beta^o$ or $= \sqrt{(B^oD\omega)/(1-\epsilon)}/\beta^o$, modified Hatta number
$\underline{Q}$ = reaction rate, mol/m^3·s
$\overline{Q}$ = Laplace transform of reaction rate, mol/m^3
r = radial space coordinate, m
R = radius of the dispersed droplets, m
R_b = radius of the gas bubbles or solid particles, m
s = surface renewal frequency in the gas- (or solid-) liquid boundary layer, 1/s
sh = sinh
s^o = $\sqrt{s\delta^2/D}$, dimensionless time for boundary layer at gas-liquid interface
s_{cd}^o = $\sqrt{s\delta_d^2/D_d} = s^o/\delta_r$, dimensionless time for boundary layer around the particles
s_d^o = $\sqrt{sR^2/D_d} = s_{cd}^o\sqrt{D_r}/S$, dimensionless time for particles
S = spherical coefficient, $S = \delta_d/R$
S_b = δ_d/R_b
t = time, s
th = tanh
T_1 = constant, Eq. 9
T_2 = constant, Eq. 19
T_3 = constant, Eqs. 55 and 57
x = space coordinate, m

Greek letters

β^o = physical mass-transfer coefficient of the gas- (or solid-) liquid boundary layer, $\beta^o = Ds^o/(\delta th\ s^o)$, m/s
β_{cd}^o = external mass-transfer coefficient between the dispersed and continuous phases in the bulk liquid phase, $\beta_{cd}^o = D(1+S)/\delta_d$, m/s
β_{cd} = external mass-transfer coefficient with reaction defined by Eq. 8, m/s
β_d = internal mass-transfer coefficient with reaction defined by Eq. 11, m/s
β_h = hydrodynamic part of the mass-transfer coefficient between the dispersed and continuous phases, $\beta_h = D/\delta_d$ or $Ds_{cd}^o/(\delta_d th\ [s_{cd}^o])$, m/s
β_1 = mass-transfer coefficient with the total resistance in bulk liquid phase (Eq. 13), m/s
δ = thickness of boundary layer at the gas- (or solid-) liquid interface, m
δ_d = thickness of film layer around of the dispersed particles, m
δ_r = δ/δ_d
ϵ = holdup of the dispersed liquid phase
θ = void fraction of catalyst
τ = tortuosity factor
λ_1 = defined by Eq. 43
λ_0 = defined by Eqs. 48a and 48b
ω = specific surface of the dispersed liquid phase, $\omega = 6\epsilon/d_p$, m^2/m^3

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