Int. J. Heat Mass Transfer. Vol. 21, pp. 445–453. Pergamon Press 1978. Printed in Great Britain

UNSTEADY STATE MASS TRANSFER THROUGH THE INTERFACE OF SPHERICAL PARTICLES—I

PHYSICAL AND MATHEMATICAL DESCRIPTION OF THE MASS-TRANSFER PROBLEM

H. BRAUER
Institut für Chemieingenieurtechnik, Technische Universität, Berlin, Germany

(*Received* 11 *October* 1976 *and in revised form* 16 *August* 1977)

Abstract—Unsteady state mass transfer through the interface of spherical particles has been thoroughly investigated using numerical methods. The particles may be bubbles, drops and solids. Mass transfer may occur in a motionless system and in a system with either the surrounding fluid only or both fluids being in motion. Creeping flow conditions are assumed for the surrounding fluid, so that the equations presented by Hadamard and Rybczinski for the velocity field can be used for calculations of the concentration field. The first part of the paper is devoted to a comprehensive discussion of the various mass-transfer conditions. This discussion is the basis for an understanding of the differential equations governing the concentration field inside and outside the sphere and the pertaining initial, boundary and interfacial conditions. These conditions are given for the general case of mass-transfer resistance in both phases as well as for the two limiting cases, for which mass-transfer resistance occurs in one of the two phases only.

NOMENCLATURE

A_p,	particle surface area;
d_p,	particle diameter;
D,	diffusion coefficient;
Fo_m,	Fourier-number, defined by (5);
M_A,	transferred mass;
$\dot{M}_{At}$,	surface mean instantaneous mass flux;
$\dot{m}_{At}$,	surface mean instantaneous mass flux density;
$\dot{m}_{A0t}$,	local instantaneous mass flux density;
r,	radical coordinate;
r^*,	normalized radical coordinate, defined by (21);
R,	radius of particle;
$ReSc$,	convection number, defined by (26);
Sh_{0t},	local instantaneous Sherwood number, defined by (43);
Sh_t,	surface mean instantaneous Sherwood number, defined by (45);
t,	time;
V_p,	particle volume;
w_∞,	fluid velocity in great distance of particle;
w_r,	radial component of fluid velocity;
w_θ,	tangential component of fluid velocity;
w_r^*,	normalized radial velocity, defined by (22);
w_θ^*,	normalized tangential velocity, defined by (24).

Greek symbols

β,	surface mean and time mean mass-transfer coefficient;
β_t,	surface mean instantaneous mass-transfer coefficient;
β_{0t},	local instantaneous mass-transfer coefficient;
θ,	angular coordinate;
ν,	kinematic viscosity;
ξ,	normalized local concentration, defined by (2) and (3);
$\bar{\bar{\xi}}$,	normalized mean concentration, defined by (16);
ξ_p,	normalized concentration in phase boundary, defined by (4);
ρ_A,	partial mass density of transferred mass;
ρ_{Ap},	partial mass density of transferred mass in phase boundary.

Subscripts

1, 2,	referring to phase 1 or 2.

1. INTRODUCTION

MASS transfer through the interface of particles is under all conditions a time dependent process. The mathematical description of this process turned out to be extremely difficult. To arrive at solutions important simplifying assumptions had to be introduced.

This paper is concerned with transfer processes through the interface of spherical particles. The spherical particles may be gas bubbles within a liquid continuous phase, liquid drops within a gaseous or a liquid environment and solid particles within a gaseous or a liquid continuous phase.

Several aspects of mass-transfer processes related to spherical particles have been investigated by means of theoretical methods by a relatively small number of scientists. Most of them assume that the relative motion between the spherical particle and the environment does not depend on time. Furthermore the relative motion is in most cases restricted to the low Reynolds number range. The fluid flow in the environment and inside of the spherical particles is therefore given by equations that have been presented by Stokes [1], Hadamard [2] and Rybczinski [3]. In other cases potential flow of the surrounding fluid has been assumed.

The mathematical treatment of the mass-transfer problem presented by Ruckenstein [4] is based on the assumption, that variations in the local concentration are restricted to the immediate vicinity of the interface. Boundary-layer simplifications may therefore be used to solve the differential equation for the concentration field. The results of this treatment are consequently restricted to large values of the parameter *ReSc* and to very short times. The same restrictions are valid for investigations carried out by Taunton and Lightfoot [5], Chao [6], Chao and Chen [7] and Konopliv and Sparrow [8] although different mathematical procedures have been adopted by these authors.

The case, where resistance to diffusional transport exists inside the spherical particle only, has been studied by Kronig and Brink [9] and Johns and Beckmann [10]. The results achieved are useful for very long times only. The results of publications mentioned so far are not useful for immediate application. Therefore Schlünder [11, 12] presented a method that may be used for the solution of problems with some practical importance. This method however makes use of steady state mass-transfer coefficients. The range of application, within which this method may be used with a fair degree of accuracy, cannot be given.

This brief survey on the available literature proves the inadequacy of existing knowledge pertaining to the mass-transfer problems under discussion. Therefore the author together with a variable number of coworkers is for several years now engaged in a comprehensive theoretical study of mass and heat transfer through the interface of spherical particles. Some of the results obtained by this study group have already been published by Plöcker and Schmidt-Traub [13] and Hilprecht [14]. Further results will be presented in this paper.

The paper is presented in two parts. In the first part the physical and mathematical description of the mass-transfer problem is presented. In the second part the results of the theoretical–numerical treatment of the problem are being discussed.

2. DISCUSSION OF MASS-TRANSFER CONDITIONS

2.1. *Definitions*

Mass-transfer conditions will be discussed on the basis of concentration profiles, given for the interior of the sphere and for its environment. To simplify the discussion it will be assumed that the environment is at rest. For this case the concentration profiles are independent of the angular coordinate θ.

There are three distinct cases of mass transfer through the interface of spherical particles:

(a) General case: resistance to mass transfer occurs in both phases; phase 1 is the sphere, phase 2 is the environment. The coefficients of molecular diffusion D_1 and D_2 are of the same order of magnitude: $D_1/D_2 \to 1$.

(b) First limiting case: resistance to mass transfer occurs in phase 1, the sphere only. The coefficient of molecular diffusion for the sphere, D_1, is extremely low compared to that for the surrounding fluid, D_2. This limiting case is therefore characterized by $D_1/D_2 \to 0$.

(c) Second limiting case: resistance to mass transfer occurs in phase 2, the environment only. The coefficient of molecular diffusion for the surrounding fluid, D_2, is extremely low compared to that for the sphere, D_1. The second limiting case is therefore characterized by $D_1/D_2 \to \infty$.

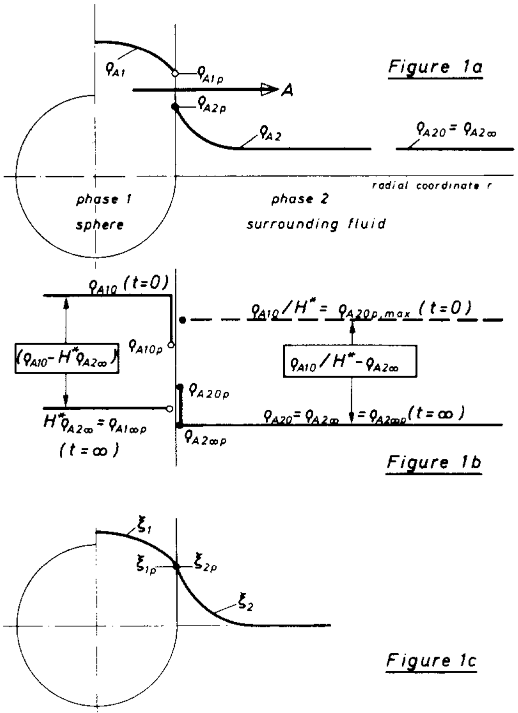

FIG. 1. For explanation of concentration profiles and definitions.

The concentration of the diffusing matter A is given by the partial mass density ρ_A. An example for concentration profiles in both phases is shown in Fig. 1(a). Within the sphere the partial mass density ρ_{A1} decreases from its highest value in the centre of the sphere to its lowest value ρ_{A1p} in the phase boundary (index p), on the interface. The concentration in the environment, given by ρ_{A2}, decreases from its highest value ρ_{A2p} in the interface to its lowest value $\rho_{A20} = \rho_{A2\infty}$ at a great distance from the interface.

In the interface the concentration jumps from ρ_{A1p} to ρ_{A2p}. The ratio of these two concentrations is given by the Henry number:

$$H^* \equiv \frac{\rho_{A1p}}{\rho_{A2p}}. \tag{1}$$

The Henry number depends on the components involved; it is a function of temperature and pressure.

For a dimensionless presentation of the concentration profiles use is made of the following definitions:

$$\xi_1 \equiv \frac{\rho_{A1}-H^*\rho_{A2\infty}}{\rho_{A10}-H^*\rho_{A2\infty}}, \tag{2}$$

$$\xi_2 \equiv \frac{\rho_{A2}-\rho_{A2\infty}}{\rho_{A10}/H^*-\rho_{A2\infty}}. \tag{3}$$

The index 0 designates values at the time $t = 0$, that is at starting time, while the index ∞ designates values at $t = \infty$. The dimensionless concentrations ξ_1 and ξ_2 are to be interpreted as follows:

$$\xi_{1,2} \triangleq \frac{\text{difference of concentrations at } t \text{ and } t=\infty \text{ in considered phase}}{\text{maximum possible difference of concentrations at } t=0 \text{ and } t=\infty \text{ in considered phase}}$$

The maximum possible concentration differences in the considered phase is shown in Fig. 1(b). The definitions of ξ_1 and ξ_2 fulfill the following conditions:

1. $0 \leqslant \xi_{1,2} \leqslant 1$
2. $\xi_{1p} = \xi_{2p}$.

The second condition avoids a concentration jump in the interface:

$$\xi_{1p} \equiv \frac{\rho_{A1p}-H^*\rho_{A2\infty}}{\rho_{A10}-H^*\rho_{A2\infty}} = \frac{H^*(\rho_{A1p}/H^*-\rho_{A2\infty})}{H^*(\rho_{A10}/H^*-\rho_{A2\infty})}$$

$$= \frac{\rho_{A2p}-\rho_{A2\infty}}{\rho_{A10}/H-\rho_{A2\infty}} = \xi_{2p}. \tag{4}$$

A concentration profile with $\xi_{1p} = \xi_{2p}$ is given in Fig. 1(c). To present the time coordinate in a dimensionless way, use is made of the Fourier number Fo_{m1} and Fo_{m2} for mass transfer, that is defined as follows:

$$Fo_{m1} \equiv \frac{t}{R^2/D_1} = Fo_{m2}\frac{D_1}{D_2} \tag{5}$$

$$Fo_{m2} \equiv \frac{t}{R^2/D_2} = Fo_{m1}\frac{D_2}{D_1}. \tag{6}$$

The Fourier number may be interpreted in the following way:

$$Fo_{m1} \triangleq \frac{\text{absolute time } t}{\text{diffusion time } R^2/D_2 \text{ specific for particle}}.$$

2.2 *Resistance to diffusion in phase* 1 (*sphere*) *only*

Resistance to diffusion in phase 1 only requires the following condition:

$$D_1/D_2 \to 0.$$

According to available experience the Henry number for this case is always smaller than one:

$$H^* \equiv \frac{\rho_{A1p}}{\rho_{A2p}} < 1.$$

The concentration profiles are depicted in Figs. 2 and 3 for the two possible directions of the mass flux. Figures

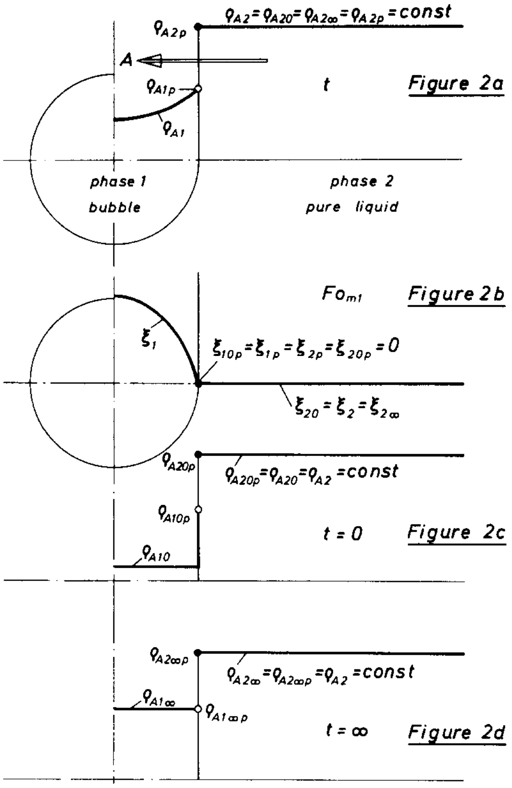

FIG. 2. Concentration profiles at various values of time t when resistance to diffusion occurs in the sphere only and when the mass flux is directed into the sphere.

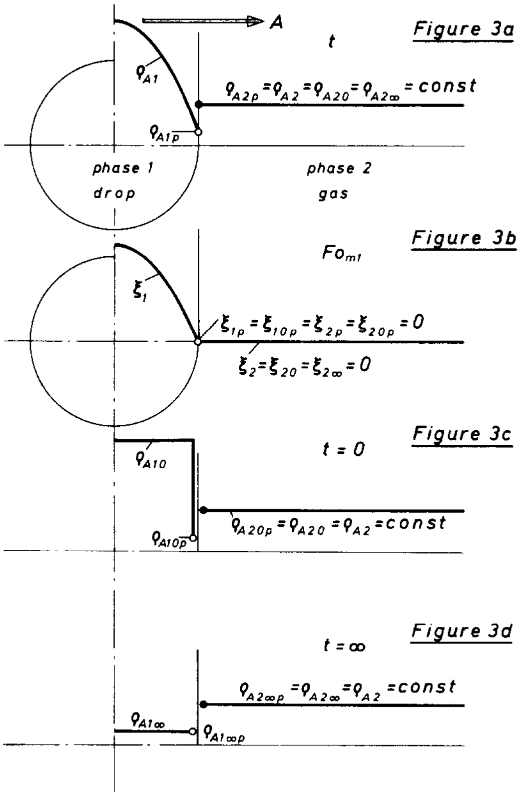

FIG. 3. Concentration profiles at various values of time t when resistance to diffusion occurs in the sphere only and when the mass flux is directed into the environment.

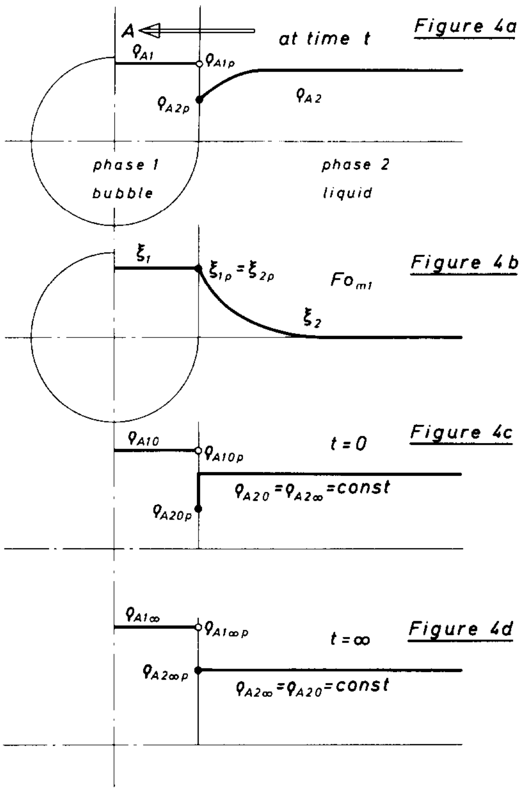

FIG. 4. Concentration profiles at various values of time t when resistance to diffusion occurs in the surrounding fluid only and when the mass flux is directed into the sphere.

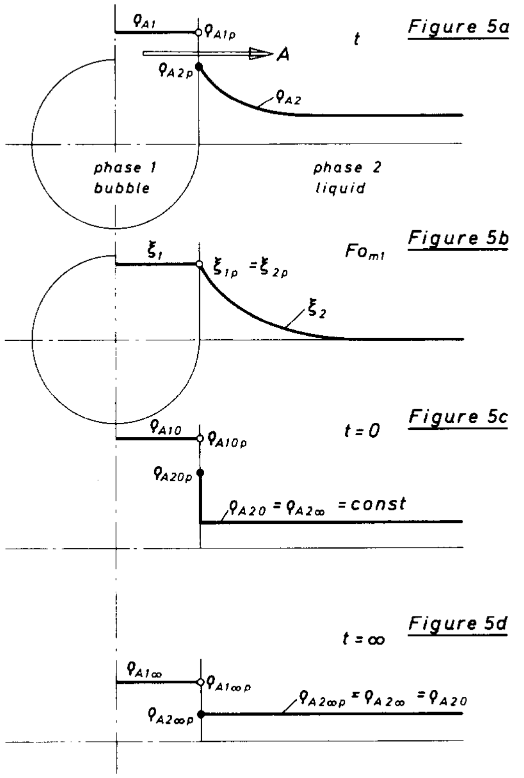

FIG. 5. Concentration profiles at various values of time t when resistance to diffusion occurs in the surrounding fluid only and when the mass flux is directed into the environment.

2(a) and (b) as well as 3(a) and (b) give the concentration profiles at time t or Fo_{m1}. The Figs. 2(c) and (d) as well as 3(c) and (d) give the concentration profiles at time $t = 0$ and $t = \infty$. At $t = \infty$ equilibrium has been obtained. It may be noted the identical concentration profiles are obtained, when dimensionless coordinates are introduced.

2.3. *Resistance to diffusion in phase* 2 (*surrounding fluid*) *only*

Resistance to diffusion in phase 2 only requires the following condition:

$$D_1/D_2 \rightarrow \infty.$$

According to available experience the Henry number for this case is always greater than one:

$$H^* \equiv \frac{\rho_{A1p}}{\rho_{A2p}} > 1.$$

The concentration profiles are shown in Figs. 4 and 5 for the two possible directions of the mass flux. It is to be noted that the concentration in the surrounding fluid in a great distance from the interface does not change at all. This is due to the fact, that the volume of the surrounding fluid is infinite compared with the volume of the sphere. Therefore the concentration at $t = 0$ is equal to that at $t = \infty$, that is $\rho_{A20} = \rho_{A2\infty}$.

2.4. *Resistance to diffusion in both phases*

If diffusional resistances occur in both phases, the coefficients of diffusion must be of the same order of magnitude:

$$D_1/D_2 \approx 1.$$

In this case the Henry number H^* may be greater or smaller than one:

$$H^* \equiv \frac{\rho_{A1p}}{\rho_{A2p}} \gtrless 1.$$

Concentration profiles for $H^* > 1$ are given in Figs. 6 and 7.

There are two cases with different direction of the mass flux for the component A. Making use of dimensionless coordinates, the concentration profile is for both cases identical.

3. CALCULATION OF TRANSFERRED MASS

In the time interval from $t = 0$ to t the mass M_A has moved across the interfacial area A_p of the sphere and either penetrated the sphere or left the sphere. Introducing a time mean value of the mass transfer coefficient, designated by β, the mass M_A may be calculated by the equation:

$$M_A = \beta A_p t(\rho_{A10} - \rho_{A1\infty}). \tag{7}$$

Introducing the relations

$$\rho_{A1\infty} = H^* \rho_{A2\infty}, \tag{8}$$

$$A_p = 6\frac{V_{p1}}{d_p} \tag{9}$$

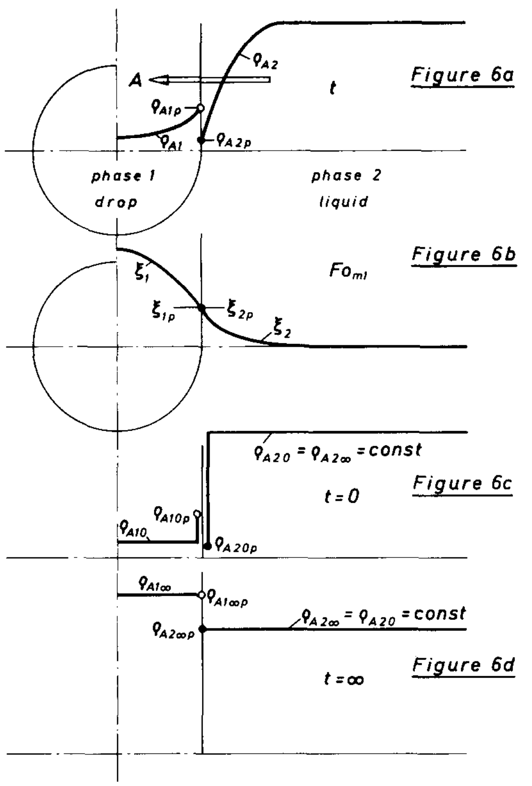

FIG. 6. Concentration profiles at various values of time t when resistance to diffusion occurs in both phases and when the mass flux is directed into the sphere.

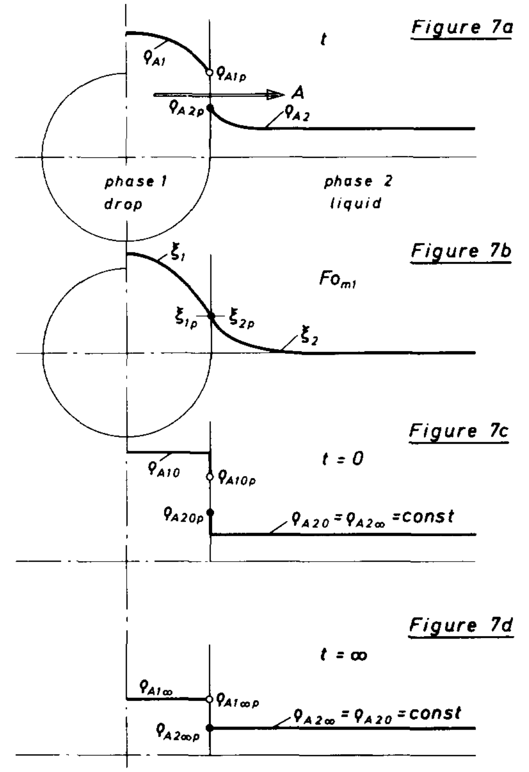

FIG. 7. Concentration profiles at various values of time t when resistance to diffusion occurs in both phases and when the mass flux is directed into the environment.

and rearranging will lead to the following equation:

$$\frac{M_A}{V_{p1}\rho_{A10}} = \frac{6\beta t}{d_p}\left(1 - H^*\frac{\rho_{A2\infty}}{\rho_{A10}}\right). \tag{10}$$

With the dimensionless numbers,

$$Sh_1 \equiv \frac{\beta d_p}{D_1} \text{ mean time value of Sherwood number} \tag{11}$$

$$Fo_{m1} \equiv \frac{tD_1}{d_p^2/4} \text{ Fourier number pertaining to phase 1} \tag{12}$$

one obtains

$$\frac{6\beta t}{d_p} = \tfrac{3}{2}Sh_1 Fo_{m1}, \tag{13}$$

and equation (10) may be rewritten as follows:

$$\frac{M_A}{V_{p1}\rho_{A10}} = Sh_1 Fo_{m1}\frac{3}{2}\left(1 - H^*\frac{\rho_{A2\infty}}{\rho_{A10}}\right). \tag{14}$$

On the LHS of this equation stands the ratio of the mass M_A transferred across the interface in the time interval between $t = 0$ and t and the mass $V_{p1}\rho_{A10}$ of component A that is within the sphere at the starting time $t = 0$. The concentration ratio $H^*\rho_{A2\infty}/\rho_{A10}$ may be interpreted as ratio of concentration in the sphere at $t = \infty$ given by $\rho_{A1\infty} = H^*\rho_{A2\infty}$ and the concentration in the sphere at $t = 0$ given by ρ_{A10}. The concentrations, the volume V_{p1} of the sphere and the Fourier number Fo_{m1} may be assumed as given.

The mass M_A transferred across the interfacial area must be equal to the change of mass of component A within the sphere:

$$M_A = V_{p1}(\rho_{A10} - \bar{\rho}_{A1}) = V_{p1}\rho_{A10}\left(1 - \frac{\bar{\rho}_{A1}}{\rho_{A10}}\right). \tag{15}$$

ρ_{A1} is the volumetric mean value of the concentration in phase 1 at time t. Introducing the dimensionless mean concentration

$$\bar{\xi}_1 \equiv \frac{\bar{\rho}_{A1} - H^*\rho_{A2\infty}}{\rho_{A10} - H^*\rho_{A2\infty}} \tag{16}$$

and rearranging delivers the result:

$$\frac{M_A}{V_{p1}\rho_{A10}} = (1 - \bar{\xi})\left(1 - H^*\frac{\rho_{A2\infty}}{\rho_{A10}}\right). \tag{17}$$

Comparing equations (14) and (17) one obtains:

$$1 - \bar{\xi} = \tfrac{3}{2}Sh_1 Fo_{m1}. \tag{18}$$

Calculation of the transferred mass M_A requires knowledge of either the Sherwood number Sh_1 or the mean concentration $\bar{\xi}_1$ at the Fourier number Fo_{m1}. Theoretical methods to determine Sh_1 and $\bar{\xi}_1$ will be explained and some of the results obtained will be discussed.

4. DIFFERENTIAL EQUATIONS FOR CONCENTRATION FIELDS

The differential equations for the concentration field within the sphere and the concentration field within the surrounding fluid will be given together with pertaining initial and boundary values for the different cases of mass transfer.

4.1. *Differential equations*

The concentration field within the sphere is given by the following differential equation:

$$\frac{\partial \xi_1}{\partial Fo_{m2}} \frac{D_1}{D_2} + \left(w_{r1}^* \frac{\partial \xi_1}{\partial r^*} + \frac{w_{\theta 1}^*}{r^*} \frac{\partial \xi_1}{\partial \theta} \right) \frac{(ReSc)_2}{2} \frac{D_2}{D_1} = \frac{1}{r^{*2}} \left[\frac{\partial}{\partial r^*} \left(r^{*2} \frac{\partial \xi_1}{\partial r^*} \right) + \frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left(\sin\theta \frac{\partial \xi_1}{\partial \theta} \right) \right]. \quad (19)$$

Similarly the differential equation governing the concentration field in the fluid surrounding the sphere is as follows:

$$\frac{\partial \xi_2}{\partial Fo_{m2}} + \left(w_{r2}^* \frac{\partial \xi_2}{\partial r^*} + \frac{w_{\theta 2}^*}{r^*} \frac{\partial \xi_2}{\partial \theta} \right) \frac{(ReSc)_2}{2} = \frac{1}{r^{*2}} \left[\frac{\partial}{\partial r^*} \left(r^{*2} \frac{\partial \xi_2}{\partial r^*} \right) + \frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left(\sin\theta \frac{\partial \xi_2}{\partial \theta} \right) \right]. \quad (20)$$

The dimensionless concentrations ξ_1 and ξ_2 as well as the Fourier numbers Fo_{m1} and Fo_{m2} have been defined by equations (2), (3), (5) and (6). The other dimensionless quantities are defined as follows:

$r^* \equiv r/R = r/(d_p/2)$
radial coordinate (21)

θ
tangential coordinate

$w_{r1}^* \equiv w_{r1}/w_\infty$
radial velocity in phase 1 (22)

$w_{r2}^* \equiv w_{r2}/w_\infty$
radial velocity in phase 2 (23)

$w_{\theta 1}^* \equiv w_{\theta 1}/w_\infty$
tangential velocity in phase 1 (24)

$w_{\theta 2}^* \equiv w_{\theta 2}/w_\infty$
tangential velocity in phase 2 (25)

$(ReSc)_2 \equiv \frac{w_\infty d_p}{v_2} \frac{v_2}{D_2} = \frac{w_\infty d_p}{D_2}$
convection number (26)

R radius of sphere, d_p diameter of sphere, w_∞ velocity of fluid surrounding the sphere at $r \rightarrow \infty$, v_2 kinematic viscosity of surrounding fluid and D_2 diffusion coefficient for species A in fluid surrounding the sphere.

Solution of the differential equations requires knowledge of the velocity field. For creeping flow conditions, $Re \rightarrow 0$, equations for radial and tangential velocity components in both phases have been worked out by Stokes, Hadamard and Rybczinsky; they are available in [15]. For the higher range of Reynolds number, $Re_2 \leqslant 150$, numerical solutions of the differential equations for the velocity field have been presented; results are available in [15].

Integration of equations (19) and (20) requires defined initial and boundary values. These will be discussed in the next chapter.

4.2. *Initial, boundary and interface conditions*

The initial and boundary conditions are of a general nature while the interface conditions depend on special mass-transfer conditions.

4.2.1. *Initial condition.* The mass-transfer process starts at time $t = 0$ or $Fo_{m1} = Fo_{m2} = 0$. At this time the concentration in phase 1 is ρ_{A10} and therefore $\xi_{10} = 1$:

$$\left.\begin{array}{l} \mathrm{Fo}_{m1} = Fo_{m2} = 0 \\ r^* < 1 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \quad \xi_{10} = 1. \quad (27)$$

This condition is independent of mass-transfer direction and distribution of mass transfer resistances in the two phases.

4.2.2. *Boundary conditions.* The fluid flow is symmetrical to the axis of the sphere, so that the concentration field is symmetrical to this axis too. The following symmetry conditions are to be observed for all values of the Fourier number, $0 \leqslant Fo_{m1} = Fo_{m2} \leqslant \infty$:

1. $$\left.\begin{array}{l} 0 \leqslant r^* \leqslant 1 \\ \theta = \left\{\begin{array}{l} 0 \\ \text{and} \\ \pi \end{array}\right. \end{array}\right\} \quad \partial \xi_1 / \partial \theta = 0. \quad (28)$$

2. $$\left.\begin{array}{l} r^* = 0 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \quad \partial \xi_1 / \partial r^* = 0. \quad (29)$$

3. $$\left.\begin{array}{l} 1 \leqslant r^* \leqslant \infty \\ \theta = \left\{\begin{array}{l} 0 \\ \text{and} \\ \pi \end{array}\right. \end{array}\right\} \quad \partial \xi_2 / \partial \theta = 0. \quad (30)$$

The last boundary condition fixes the concentration at infinite distance from the interface; it is the infinity condition:

4. $$\left.\begin{array}{l} r^* = \infty \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \quad \xi_2 = 0. \quad (31)$$

These conditions are independent of mass-transfer direction and distribution of mass-transfer resistance in the two phases.

4.2.3. *Interface conditions.* The interface conditions depend on the distribution of mass-transfer resistance in the two phases. They are independent of mass-transfer direction. They are valid for all values of the Fourier number, $0 \leqslant Fo_{m1} = Fo_{m2} \leqslant \infty$.

A general condition for the interface is the validity of Henry's law:

$$H^* \equiv \frac{\rho_{A1p}}{\rho_{A2p}}. \quad (1)$$

Making use of introduced dimensionless quantities this leads to the equality of the concentrations of both phases in the interface:

$$\left.\begin{array}{l} r^* = 1 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \quad \xi_{1p} = \xi_{2p}. \quad (32)$$

4.2.3.1. Mass-transfer resistance in phase 1 (sphere) only—According to Figs. 2(b) and 3(b) the interface condition reads as follows:

$$\left.\begin{array}{l} r^* = 1 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \quad \xi_{1p} = \xi_{2p} = 0. \quad (33)$$

4.2.3.2. Mass-transfer resistance in phase 2 (surrounding fluid) only—Figures 4(b) and 5(b) show that the concentration in phase 1 is independent of the radial coordinate. But with time ξ_1 moves from 1 to 0. The change of mass A inside the sphere with time t, is given by:

$$\left(\frac{\partial M_A}{\partial t}\right)_1 = \tfrac{4}{3}\pi R^3 \frac{\partial \bar{\rho}_{A1}}{\partial t}. \tag{34}$$

This mass flux moves across the interface and must therefore be equal to the change of mass A in phase 2:

$$\left(\frac{\partial M_A}{\partial t}\right)_2 = -\iint_{A_p} D_2\left(\frac{\partial \rho_{A2}}{\partial r}\right)_p R^2 \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi. \tag{35}$$

With A_p the interfacial area of the particle and with ϕ the angular coordinate is given. With the symmetry conditions mentioned the mass flux is independent of ϕ. Therefore equation (34) by introducing dimensionless quantities and equalized with equation (33) leads to the following interface condition:

$$\left.\begin{array}{l} r^* = 1 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \frac{\partial \bar{\bar{\xi}}_1}{\partial Fo_{m2}} = -\frac{3}{2}\frac{1}{H^*}\int_0^\pi \left(\frac{\partial \xi_2}{\partial r^*}\right)_p \sin\theta\,\mathrm{d}\theta. \tag{36}$$

It is to be noted, that the mean concentration $\bar{\bar{\xi}}_1$ is equal to the local concentration ξ_1 and that the concentration in the interface ξ_{2p} is equal to ξ_{1p} and $\bar{\bar{\xi}}_1$.

4.2.3.3. Mass-transfer resistance in both phases—For all conditions the mass flux density leaving one phase must move into the other phase. Using Ficks law for the mass flux density at both sides of the interface the following interface condition is obtained:

$$\left.\begin{array}{l} r^* = 1 \\ 0 \leqslant \theta \leqslant \pi \end{array}\right\} \frac{D_1}{D_2} H^* = \frac{(\partial \xi_2/\partial r^*)_p}{(\partial \xi_1/\partial r^*)_p}. \tag{37}$$

4.3. *Some remarks on the solution of the differential equations*

The differential equations for the concentration fields have been solved by numerical methods, based on the finite difference procedure developed by Crank–Nicolson [16]. A comprehensive description of this procedure is included in unpublished reports prepared by Schmidt-Traub [17] and Carrubba [18].

For the special cases of $Fo_{m1} \to 0$ and $Fo_{m1} \to \infty$ the differential equations allow simplifications so that analytical solutions are possible [13, 15].

Immediate results of the solved differential equations are the concentrations ξ_1 and ξ_2 for the two involved phases. The general nature of the solutions is shown by the following equations:

$$\xi_1 = f_{10}[r^*; \theta; (ReSc)_2; Sc_2; Fo_{m2}; D_1/D_2; H^*], \tag{38}$$

$$\xi_2 = f_{20}[r^*; \theta; (ReSc)_2; Sc_2; Fo_{m2}; D_1/D_2; H^*]. \tag{39}$$

These equations are the basis for further calculations such as mean concentration $\bar{\bar{\xi}}_1$ and various Sherwood numbers.

5. CALCULATION OF SHERWOOD NUMBERS

The Sherwood number is a dimensionless quantity that includes the mass-transfer coefficient, which is defined as follows:

$$\beta_{1\theta t} = \frac{\dot{m}_{A\theta t}}{\rho_{A10} - H^* \rho_{A2\infty}} \tag{40}$$

$\dot{m}_{A\theta t}$ is the instantaneous local mass flux density at the angular coordinate θ and at the time t, for which Ficks law is assumed to apply:

$$\dot{m}_{A\theta t} = -D_1\left(\frac{\partial \rho_{A1}}{\partial r}\right)_p. \tag{41}$$

Introducing equation (41) into equation (40) one obtains after rearranging an equation for the local instantaneous Sherwood number for phase 1:

$$Sh_{1\theta t} = -2\left(\frac{\partial \xi_1}{\partial r^*}\right)_p. \tag{42}$$

The Sherwood number is defined as follows:

$$Sh_{1\theta t} \equiv \frac{\beta_{1\theta t} d_p}{D_1}. \tag{43}$$

Integration over the interfacial area of the sphere leads to the surface mean instantaneous Sherwood number:

$$Sh_{1t} = \tfrac{1}{2}\int_0^\pi Sh_{1\theta t} \sin\theta\,\mathrm{d}\theta. \tag{44}$$

Definition of the Sherwood number:

$$Sh_{1t} \equiv \frac{\beta_{1t} d_p}{D_1}. \tag{45}$$

The surface mean instantaneous mass flux density $\dot{m}_{At}$ and mass flux $\dot{M}_{At}$ are calculated by means of the mass-transfer coefficient β_{1t}:

$$\dot{m}_{At} = \beta_{1t}(\rho_{A10} - H^* \rho_{A2\infty}), \tag{46}$$

$$\dot{M}_{At} = \beta_{1t} A_p (\rho_{A10} - H^* \rho_{A2\infty}). \tag{47}$$

Integrating Sh_{1t} over the Fourier number the time mean value of the Sherwood number is obtained:

$$Sh_1 = \frac{1}{Fo_{m2}} \int_0^{Fo_{m2}} Sh_{1t}\, dFo_{m2}. \tag{48}$$

The definition of Sh_1 is as follows:

$$Sh_1 \equiv \frac{\beta_1 d_p}{D_1}. \tag{49}$$

The time mean value of the mass-transfer coefficient β_1 serves to calculate the mass M_A that has been transferred across the interface in the time interval between $t = 0$ and t:

$$M_A = \beta_1 A_p t(\rho_{A10} - H^* \rho_{A2\infty}). \tag{50}$$

This equation is identical with equation (7) combined with equation (8). The equations (40)–(50) refer to phase 1. Equations pertaining to phase 2 may be developed in the same way. Therefore the equations for phase 2 are given without further comments.

$$\beta_{2\theta t} = \frac{\dot{m}_{A\theta t}}{\rho_{A10} - H^* \rho_{A2\infty}}, \tag{40a}$$

$$\dot{m}_{A\theta t} = -D_2\left(\frac{\partial \rho_{A2}}{\partial r}\right)_p, \tag{41a}$$

$$Sh_{2\theta t} = -\frac{2}{H^*}\left(\frac{\partial \xi_2}{\partial r^*}\right)_p, \tag{42a}$$

$$Sh_{2\theta t} \equiv \frac{\beta_{2\theta t} d_p}{D_2}, \tag{43a}$$

$$Sh_{2t} = \tfrac{1}{2}\int_0^{\pi} Sh_{2\theta t} \sin\theta \, \mathrm{d}\theta, \tag{44a}$$

$$Sh_{2t} \equiv \frac{\beta_{2t} d_p}{D_2}, \tag{45a}$$

$$Sh_2 = \frac{1}{Fo_{m2}}\int_0^{Fo_{m2}} Sh_{2t} dFo_{m2}. \tag{48a}$$

Comparison of these equations shows that

$$\left.\begin{aligned} \beta_{1\theta t} &= \beta_{2\theta t}, \\ \beta_{1t} &= \beta_{2t}, \\ \beta_1 &= \beta_2. \end{aligned}\right\} \tag{51}$$

For mass-transfer calculations it is therefore of no importance whether the mass-transfer coefficient is determined for phase 1 or for phase 2.

A comprehensive discussion of the results obtained by solving the differential equations will be presented in Part II of the investigation.

REFERENCES

1. G. G. Stokes, On the effect of internal friction of fluids on the motion of pendulums, *Trans. Camb. Phil. Soc.* **9**(II), 8–106 (1851).
2. J. Hadamard, Mouvement permanente lent d'une sphére liquide et visqueuse dans un liquid visqueux, *C. R. Hebd. Séanc. Acad. Sci., Paris* **152**, 1735–1817 (1911).
3. W. Rybczinski, *Bull. Int. Acad. Sci. Lett. Cracovie A* **40** (1911).
4. E. Ruckenstein, Mass transfer between a single drop and a continuous phase, *Int. J. Heat Mass Transfer* **10**, 1785–1792 (1967).
5. J. W. Taunton and W. N. Lightfoot, Transient effects in mass transfer to circulating drops, *Int. J. Heat Mass Transfer* **12**, 1817–1822 (1969).
6. B. T. Chao, Transient heat and mass transfer to a translating droplet, *J. Heat Transfer* **91C**(2), 273–281 (1969).
7. B. T. Chao and L. S. Chen, Series solution of unsteady heat or mass transfer to a translating fluid sphere, *Int. J. Heat Mass Transfer* **13**, 359–367 (1970).
8. N. Konopliv and E. M. Sparrow, Unsteady heat transfer and temperature for Stokesian flow about a sphere, *J. Heat Transfer* **94C**(3), 266–272 (1972).
9. R. Kronig and J. C. Brink, On the theory of extraction from falling droplets, *Appl. Scient. Res.* **A2**, 142–154 (1950).
10. L. E. Johns and R. B. Beckmann, Mechanism of dispersed-phase mass transfer in viscous, single-drop extraction systems, *A.I.Ch.E. Jl* **12**(1), 10–16 (1969).
11. E. U. Schlünder, Über die Brauchbarkeit des Newtonschen Abkühlungsgesetzes im Licht der Fourierschen Wärmeleitungstheorie, *Wärme- und Stoffübertragung* **5**(1), 9–14 (1972).
12. E. U. Schlünder, Thermische Verfahrenstechnik, *Beil. Z. Verfahrenstechnik* (1972/1973).
13. U. Plöcker and H. Schmidt-Traub, Instationärer Stofftransport zwischen einer Einzelkugel und einer ruhenden Umgebung, *Chemie-Ingr-Tech.* **44**, 313–319 (1972).
14. L. Hilprecht, Instationärer Impuls- und Stoffaustausch bei beschleunigter Bewegung von Einzelpartikeln, Diss. D83, Techn. Univ. Berlin (1975).
15. H. Brauer and D. Mewes, *Stoffaustausch einschließlich chemischer Reaktionen.* Sauerländer, Aarau (1971).
16. J. Crank, *Mathematics of Diffusion.* Oxford University Press, Oxford (1956).
17. H. Schmidt-Traub, Final report on the research project: Instationärer Stofftransport an festen Partikeln, Tropfen und Blasen. Unpublished report.
18. G. Carrubba, Untersuchung des instationären Stofftransportes in Kugeln mit innerer Zirkulation. Internal, unpublished report.

TRANSFERT MASSIQUE INSTATIONNAIRE A TRAVERS L'INTERFACE DE PARTICULES SPHERIQUES—1^e PARTIE: DESCRIPTION PHYSIQUE ET MATHEMATIQUE DU PROBLEME DE TRANSFERT MASSIQUE

Résumé—Le transfert massique instationnaire à travers l'interface de particules sphériques a été soigneusement étudié à partir de méthodes numériques. Les particules peuvent être des bulles, des gouttes et des solides. Le transfert massique peut avoir lieu dans un système immobile et dans un système avec mouvement pour le fluide environnant ou pour les deux fluides. Les conditions d'écoulement rampant sont supposées pour le fluide environnant, de telle sorte que les équations données par Hadamard et Rybozinski pour le champ des vitesses peuvent être utilisées dans le calcul du champ de concentration. La première partie de l'article est consacrée à une discussion des conditions variées de transfert massique. On se base sur les équations aux dérivées partielles du champ des concentrations à l'intérieur et à l'extérieur de la sphère et des conditions initiales, aux limites et à l'interface. Ces conditions sont données dans le cas général d'une résistance au transfert massique dans les deux phases et aussi dans les deux cas limites d'une seule résistance dans l'une ou l'autre phase.

INSTATIONÄRER STOFFTRANSPORT DURCH DIE GRENZFLÄCHE KUGELFÖRMIGER PARTIKELN TEIL 1 PHYSIKALISCHE UND MATHEMATISCHE BESCHREIBUNG DES STOFFTRANSPORTPROBLEMS

Zusammenfassung—Der instationäre Stofftransport durch die Grenzfläche kugelförmiger Partikeln wurde mittels numerischer Methoden theoretisch untersucht. Die Partikeln können Blasen, Tropfen und Körner sein. Der Stofftransport kann in einem ruhenden System und in einem System mit Bewegung in dem umgebenden oder auch in beiden Fluiden stattfinden. Für das umgebende Fluid wird schleichende Bewegung angenommen, so daß die von Hadamard und Rybczinski angegebenen Gleichungen für die Geschwindigkeitsfelder zur Berechnung der Konzentrationsfelder herangezogen werden können. Im ersten Teil der Untersuchung werden die physikalischen Bedingungen für den Stofftransport ausführlich erörtert. Das bildet die Grundlage zum Verständnis der Differentialgleichungen mit Randbedingungen, die die Konzentrationsfelder innerhalb und außerhalb der Partikeln beschreiben. Die Randbedingungen werden für die folgenden Fälle gegeben: 1. Stofftransportwiderstand in beiden Phasen; 2. Stofftransportwiderstände in nur jeweils einer der beiden Phasen.

НЕСТАЦИОНАРНЫЙ МАССОПЕРЕНОС ЧЕРЕЗ ПОВЕРХНОСТЬ СФЕРИЧЕСКИХ ЧАСТИЦ. ЧАСТЬ 1. ФИЗИЧЕСКОЕ И МАТЕМАТИЧЕСКОЕ ОПИСАНИЕ ПРОБЛЕМЫ

Аннотация — С помощью численных методов проведено детальное исследование нестационарного переноса массы через поверхность сферических частиц, примерами которых могут служить пузырьки, капли и твердые тела. Перенос массы может иметь место как в неподвижной системе, так и в системе, в которой движется только окружающая среда, или обе фазы (жидкость и частицы). Предполагается, что течение окружающей среды ползущее, поэтому для расчета поля концентрации можно использовать уравнения Адамара и Рыбчинского для поля скоростей. Дан подробный анализ различных условий массообмена, который необходим для понимания дифференциальных уравнений, описывающих поля концентрации внутри сферы и вне её, и соответствующих начальных и граничных условий, а также условий на поверхности раздела фаз. Данные условия приведены для общего случая наличия сопротивления переносу массы в обеих фазах, а также для двух предельных случаев, когда сопротивление переносу массы имеет место только в одной из фаз.