UDC 532.72 : 532.529

UNSTEADY MASS EXCHANGE BETWEEN A BUBBLE AND MEDIUM IN A REACTOR WITH FLUIDIZED LAYER

PMM Vol. 39, №1, 1975, pp. 118-129

Iu. P. GUPALO, Iu. S. RIAZANTSEV and Iu. A. SERGEEV
(Moscow)

(Received October 1, 1974)

Approximate solution of the problem of unsteady mass exchange between a bubble and medium in a reactor with a fluidized layer in the presence of first order volume chemical reaction is derived. It is assumed that the bubble velocity exceeds the rate of fluidization, which results in the formation of a closed circulation region containing the bubble [1]. The problem reduces to solving the equation of unsteady convective diffusion outside the closed circulation region, and also of the equation of balance of the reagent in that region. Variation of the reagent concentration along the reactor resulting from the volume reaction and lengthwise agitation is taken into consideration. The field of concentration outside the closed circulation region, the change of reagent concentration inside that region with time, and the coefficient of mass exchange are determined by the method of joining asymptotic expansions in small Péclet numbers.

The problem of mass exchange between a bubble with a fluidized layer with volume reaction without allowance for variation of concentration along the reactor was solved in [2] by numerical methods for considerable Péclet numbers. The Authors had investigated the less essential process of mass exchange between the bubble and the surrounding cloud inside the region of circulation. A similar problem of mass exchange between a bubble and a stationary fluid was also considered in [3] for considerable Péclet numbers and specially selected boundary conditions for concentration at considerable distance from the bubble.

An analytic solution of the problem of unsteady mass exchange between a bubble and the continuous phase of a fluidized layer is derived below for the case of small Péclet numbers with allowance for a chemical reaction in the layer. The results obtained in [4] represent a particular case of this solution.

1. Statement of problem. Method of solution. Let us consider a spherical bubble rising at constant velocity $u_b > v_0$ in a reactor with a layer of fluidized catalyst (v_0 is the mean velocity of the homogeneous stream of fluid in the interstices of particles away from the bubble). The bubble radius a_b is assumed fairly small in comparison with the transverse and longitudinal dimensions of reactor and the distance of the bubble from the walls and in- and outlet sections of the reactor is sufficiently great so that the effect of the reactor boundaries on the flow around the bubble can be neglected.

The flow field of the fluid phase outside the bubble is defined by the stream function [1]

$$\psi = (u_b - v_0)\left(1 - \frac{a_c^3}{r^3}\right)\frac{r^2}{2}\sin^2\theta \tag{1.1}$$

$$\frac{a_c^3}{a_b^3} = \frac{1+2\delta}{1-\delta}, \qquad \delta = \frac{v_0}{u_b}$$

where a_c is the radius of the cloud surrounding the bubble whose external surface is impermeable to the fluid phase. Inside the region bounded by that surface there is intense circulation of fluid, hence it is assumed that the fluid is thoroughly agitated there.

In a fluidized layer a chemical reaction takes place on the surface of catalyst particles, which corresponds to a first-order effective volume reaction whose rate constant is denoted by k. The lengthwise agitation of reagent in the reactor is determined by the convective diffusion process whose effective coefficient is denoted by D. The characteristic numbers of the problem are: the Péclet number P and the Thiele modulus Ψ defined, respectively, by

$$P = \frac{(u_b - v_0)\, a_c}{D}, \qquad \Psi = a_c \sqrt{\frac{k}{D}}$$

As the result of the reaction, diffusion and convective transport of the reagent, a certain lengthwise concentration distribution $c_\infty(x)$ sets in the reactor. This concentration must be taken into account in establishing boundary conditions at considerable distance from the bubble. It is determined by the solution of the one-dimensional equation of steady convective diffusion with boundary conditions

$$D\frac{d^2 c_\infty}{dx^2} - v_0 \frac{dc_\infty}{dx} - kc_\infty = 0$$

$$x = -x_1, \quad c_\infty = c_1; \quad x \to \infty, \quad c_\infty \to 0$$

(where coordinate $x = -x_1$ $(x_1 > 0)$ corresponds to the reactor inlet) for a semi-infinite reactor. The solution for the c_∞, with allowance for subsequent analysis can be written as

$$c_\infty = c_0 \exp\left(-P\eta \frac{x}{a_c}\right), \qquad c_0 = c_1 \exp\left(-P\eta\frac{x_1}{a_c}\right) \tag{1.2}$$

$$\eta = \sqrt{\frac{1}{4}\frac{\delta^2}{(1-\delta)^2} + \frac{\Psi^2}{P^2}} - \frac{1}{2}\frac{\delta}{1-\delta}$$

where c_0 is the reagent concentration at point $x = 0$ to which we shall relate henceforth the bubble center at the initial instant of time.

The problem reduces to the determination of the effect of the time-dependent concentration of the diffusion flux outside the cloud on its external boundary and of the reagent concentration c_+ inside the closed circulation region. Since solid phase particles are present inside the cloud, it is reasonable to assume that a chemical reaction of the first order whose effective rate coefficient is k_+ takes place inside that region. We neglect the effect of reactor walls and inlet on the diffusion process, and assume that away from the bubble concentration is determined by formula (1.2).

The concentration field outside the closed circulation region is determined by the equation of convective diffusion which in dimensionless variables

$$r' = \frac{r}{a}, \qquad \mu = \cos\theta, \qquad \tau = \frac{D}{a_c^2}\, t \tag{1.3}$$

$$c' = \frac{c}{c_0}, \quad c_+' = \frac{c_+}{c_0}, \quad \psi' = \frac{\psi}{a_c^2\,(u_b - v_0)}$$

is of the form (the prime is subsequently omitted)

$$\frac{\partial c}{\partial \tau} = \Delta c - \frac{P}{r^2}\frac{\partial(\psi, c)}{\partial(r, \mu)} - \Psi^2 c \tag{1.4}$$

In specifying boundary conditions away from the bubble and of the initial condition at $\tau = 0$, it should be noted that coordinates x and r are related by formula

$$x = \frac{u_b a_c^2}{D}\tau - a_c r\mu \equiv a_c P \frac{1}{1-\delta}\tau - a_c r\mu$$

With the use of (1.2) from this we obtain the boundary and initial conditions of the form

$$r \to \infty, \quad c \to \exp\left(-P^2\eta \frac{1}{1-\delta}\tau\right)\exp(P\eta r\mu) \tag{1.5}$$

$$\tau = 0, \quad r > 1, \quad c = \exp(P\eta r\mu) \tag{1.6}$$

The equation for the concentration c_+ inside the closed circulation region is derived from the expression for the concentration change rate c_+ produced by the reaction inside the cloud and by the inflow of substance from outside. It is of the form

$$\frac{dc_+}{d\tau} + \Psi_+^2 c_+ = \frac{3}{2}\int_{-1}^{1} \frac{\partial c}{\partial r}\bigg|_{r=1} d\mu, \quad \Psi_+ = a_c\sqrt{\frac{k_+}{D}} \tag{1.7}$$

where Ψ_+ is the Thiele modulus for the closed circulation region.

The initial condition for Eq. (1.7) is

$$\tau = 0, \qquad c_+ = c_{+0} \tag{1.8}$$

where generally $c_{+0} \neq c_0$

Boundary condition for $r = 1$ is obtained from the condition of concentration continuity at the external boundary of the cloud

$$r = 1, \qquad c = c_+ \tag{1.9}$$

Thus the problem of determination of c and c_+ reduces to solving the system of Eqs. (1.4) and (1.7) with boundary and initial conditions (1.5), (1.6), (1.8) and (1.9).

We introduce functions ξ and ξ_+ by formulas

$$c = (1 - \xi(r, \mu, \tau))\exp\left(-P^2\eta\frac{1}{1-\delta}\tau\right)\exp(P\eta r\mu) \tag{1.10}$$

$$c_+ = \exp\left(-P^2\eta\frac{1}{1-\delta}\tau\right)\xi_+(\tau)$$

The problem of determination of functions ξ and ξ_+ is defined by

$$\frac{\partial\xi}{\partial\tau} = \Delta\xi - \frac{P}{r^2}\frac{\partial(\psi, \xi)}{\partial(r, \mu)} + 2P\eta\left(\mu\frac{\partial\xi}{\partial r} + \frac{1-\mu^2}{r}\frac{\partial\xi}{\partial\mu}\right) + \tag{1.11}$$

$$P^2\eta\left(-\frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\mu}{r^2}\frac{\partial\psi}{\partial\mu} + 1\right)(\xi - 1)$$

$$\frac{d\xi_+}{d\tau} - P^2\eta\frac{1}{1-\delta}\xi_+ + \Psi_+^2\xi_+ =$$

$$\frac{3}{2}P\eta\int_{-1}^{1}\mu e^{P\eta\mu}(1-\xi)|_{r=1}\,d\mu - \frac{3}{2}\int_{-1}^{1} e^{P\eta\mu}\frac{\partial\xi}{\partial r}\bigg|_{r=1} d\mu$$

$$\tau = 0, \quad r > 1, \quad \xi = 0; \quad \tau = 0, \quad \xi_+ = c_{+0}$$

$$r = 1, \quad e^{P\eta\mu}(1-\xi) = \xi_+; \quad r \to \infty, \quad \xi \to 0$$

In deriving Eqs. (1.11) we used the identity which follows from the relationship between η, Ψ and P in accordance with the formula in (1.2)

$$P^2\eta^2 - \Psi^2 + P^2\eta\frac{1}{1-\delta} = P^2\eta$$

The solution of problem (1.11) is constructed by the method of joining asymptotic expansions in small Péclet number. It is sought in the form of external and internal expansion of ξ in Péclet number in regions $r > O(1/P)$, and $1 \leqslant r < O(1/P)$, respectively, and of an asymptotic expansion of ξ_+ in P. Such solution is presented below in the form of asymptotics for time $\tau > O(P^{-2})$.

The dimensionless stream function ψ in regions $1 \leqslant r < O(1/P)$ and $r > O(1/P)$ is, in accordance with (1.1) and (1.3), of the form

$$1 \leqslant r < O(1/P), \quad \psi = \psi_* \equiv \left(r^2 - \frac{1}{r}\right)\frac{1-\mu^2}{2} \tag{1.12}$$

$$r > O(1/P), \quad \psi = \frac{\psi^*}{P^2}, \quad \psi^* = \rho^2\frac{1-\mu^2}{2} + O(P^3), \quad (\rho = rP)$$

To solve problem (1.11) we use the Laplace transformation

$$\zeta = \int_0^\infty \exp(-P^2 s\tau)\,\xi(r,\mu,\tau)\,d\tau, \quad \zeta_+ = \int_0^\infty \exp(-P^2 s\tau)\,\xi_+(\tau)\,d\tau$$

With this definition of Laplace transformation the pre-image asymptotics with respect to time $\tau > O(P^{-2})$ correspond to image values $|s| < O(1)$ as implied by the theorem on the relation between pre-image and image values. Owing to the boundedness of ξ and ξ_+ when $\tau \to +\infty$ finctions $\zeta(s)$ and $\zeta_+(s)$ are analytic functions of s when $\operatorname{Re} s > 0$.

From (1.11) for $\zeta(s)$ and $\zeta_+(s)$ we obtain the problem

$$P^2 s\zeta = \Delta\zeta - \frac{P}{r^2}\frac{\partial(\psi,\zeta)}{\partial(r,\mu)} + 2P\eta\left(\mu\frac{\partial\zeta}{\partial r} + \frac{1-\mu^2}{r}\frac{\partial\zeta}{\partial\mu}\right) + \tag{1.13}$$
$$P^2\eta\left(-\frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\mu}{r^2}\frac{\partial\psi}{\partial\mu} + 1\right)\zeta - \frac{\eta}{s}\left(-\frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\mu}{r^2}\frac{\partial\psi}{\partial\mu} + 1\right)$$

$$P^2 s\zeta_+ - c_{+0} - P^2\eta\frac{1}{1-\delta}\zeta_+ + \Psi_+^2\zeta_+ = \tag{1.14}$$
$$\frac{3}{2}P\eta\int_{-1}^{1}\mu e^{P\eta\mu}\left(\frac{1}{P^2 s} - \zeta\right)\Big|_{r=1}d\mu - \frac{3}{2}\int_{-1}^{1}e^{P\eta\mu}\frac{\partial\zeta}{\partial r}\Big|_{r=1}d\mu$$

$$r = 1, \quad \frac{1}{P^2 s} - \zeta = e^{-P\eta\mu}\zeta_+ \tag{1.15}$$

$$r \to \infty, \quad \zeta \to 0 \tag{1.16}$$

We seek the solution of problem (1.13) – (1.16) in the form of external (denoted by ζ^*) and internal (denoted by ζ_*) expansions for function ζ and an asymptotic expansion of function ζ_+ in Péclet number in the form

$$\zeta_* = \sum_{n=0}^{\infty}\alpha_n(P)\,\zeta_n(r,\mu,s), \quad \zeta^* = \sum_{n=0}^{\infty}\alpha^{(n)}(P)\,\zeta^{(n)}(\rho,\mu,s)$$

$$\zeta_+ = \sum_{n=0}^{\infty}\alpha_{+n}(P)\,\zeta_{+n}(s)$$

As to functions $\alpha_n(P)$, $\alpha^{(n)}(P)$ and $\alpha_{+n}(P)$ the only assumption is that for $P \to 0$,

$$\frac{\alpha_{n+1}(P)}{\alpha_n(P)} \to 0, \quad \frac{\alpha^{(n+1)}(P)}{\alpha^{(n)}(P)} \to 0, \quad \frac{\alpha_{+n+1}(P)}{\alpha_{+n}(P)} \to 0$$

In region $1 \leqslant r < O(1/P)$ the problem for functions ζ_* and ζ_+ is of the form (1.13)–(1.15). In the region $r > O(1/P)$ the problem for function ζ^* is of the form

$$s\zeta^* = \Delta^*\zeta^* - \frac{1}{\rho^2}\frac{\partial(\psi^*, \zeta^*)}{\partial(\rho, \mu)} + 2\eta\left(\mu\frac{\partial\zeta^*}{\partial\rho} + \frac{1-\mu^2}{\rho}\frac{\partial\zeta^*}{\partial\mu}\right) + \tag{1.17}$$
$$\eta\left(-\frac{1}{\rho}\frac{\partial\psi^*}{\partial\rho} + \frac{\mu}{\rho^2}\frac{\partial\psi^*}{\partial\mu} + 1\right)\zeta^* -$$
$$\frac{\eta}{P^2 s}\left(-\frac{1}{\rho}\frac{\partial\psi^*}{\partial\rho} + \frac{\mu}{\rho^2}\frac{\partial\psi^*}{\partial\mu} + 1\right), \quad \rho \to \infty, \ \zeta^* \to 0$$

where $\rho = rP$ is the contracted radial coordinate and Δ^* is the Laplace operator in coordinates ρ, θ.

The constants which appear in the solution of problem (1.13) – (1.15) and (1.17) are determined by joining the internal and external expansions for ζ.

2. The zero and first approximations for ζ and ζ_+. Noting that

$$-\frac{1}{\rho}\frac{\partial\psi^*}{\partial\rho} + \frac{\mu}{\rho^2}\frac{\partial\psi^*}{\partial\mu} + 1 = O(P^3) \tag{2.1}$$

for the zero term of external expansion $\zeta^{(0)}$ we have the problem which obtains from problem (1.17) as the result of discarding the last two terms of the equation in accordance with (2.1). This problem has the trivial solution

$$\zeta^{(0)} = 0$$

The form of the boundary condition (1.15) implies that in the zero approximation $\alpha_0(P) = \alpha_{+0}(P) = P^{-2}$. Hence for the zero term of the internal expansion ζ_0 from (1.13) and (1.15) we obtain the problem

$$\Delta\zeta_0 = 0; \quad r = 1, \quad \zeta_0 = -\zeta_{+0} + \frac{1}{s}$$

The solution of this problem after joining ζ_0 and $\zeta^{(0)}$ is

$$\zeta_0 = \left(-\zeta_{+0} + \frac{1}{s}\right)\frac{1}{r} \tag{2.2}$$

The obtained solution makes it possible to determine ζ_{+0} with the use of Eq. (1.14). Retaining in it terms of order $1/P^2$ and taking into account the asymptotic expansions of exponents appearing in it in P, we obtain the equation for ζ_{+0}

$$\Psi_+^2\zeta_{+0} = -\frac{3}{2}\int\limits_{-1}^{1}\frac{\partial\zeta_0}{\partial r}\bigg|_{r=1} d\mu \tag{2.3}$$

Having determined ζ_{+0} from (2.3), from (2.2) we obtain the explicit form of ξ_0. Finally, we obtain

$$\zeta_{+0} = \frac{1-q}{s}, \quad \zeta_0 = \frac{q}{sr}, \quad q = \frac{\Psi_+^2}{\Psi_+^2 + 3} \tag{2.4}$$

It follows from (2.4) that $\alpha^{(1)}(P) = 1/P$. Retaining in (1.17) terms of order $1/P$ and taking into account (1.12) and (2.1), we obtain for the determination of the first term of external expansion $\zeta^{(1)}$ the problem

$$\Delta^*\zeta^{(1)} - (1 - 2\eta)\left(\mu\frac{\partial\zeta^{(1)}}{\partial\rho} + \frac{1-\mu^2}{\rho}\frac{\partial\zeta^{(1)}}{\partial\mu}\right) - s\zeta^{(1)} = 0; \tag{2.5}$$
$$\rho \to \infty, \quad \zeta^{(1)} \to 0$$

The substitution

$$\zeta^{(1)}(\rho, \mu) = \exp\left(\frac{1-2\eta}{2}\rho\mu\right)\zeta'^{(1)}(\rho, \mu)$$

reduces Eq. (2.5) to the Helmholtz equation

$$\Delta^*\zeta'^{(1)} - \left[\frac{(1-2\eta)^2}{4} + s\right]\zeta'^{(1)} = 0$$

From this we obtain the general solution of problem (2.5)

$$\zeta^{(1)} = \exp\left(\frac{1-2\eta}{2}\rho\mu\right)\rho^{-1/2}\sum_{n=0}^{\infty} A_n K_{n+1/2}\left(\frac{\rho}{2}\sqrt{(1-4\eta)^2+4s}\right)P_n(\mu) \qquad (2.6)$$

$$K_{n+1/2}(x) = \sqrt{\frac{\pi}{2x}}\, e^{-x}\sum_{m=0}^{n}(2x)^{-m}\frac{(m+n+1)!}{m!\,(n-m+1)!}$$

where $(P_n(\mu)$ is the Legendre polynomial Henceforth we select that branch of function $\sqrt{(1-2\eta)^2+4s}$, which yields positive roots at the real semi-axis $0 < s < +\infty$.

Using the condition of joining $P^{-1}\zeta^{(1)}$ with $P^{-2}\zeta_0$ for determining A_n, we obtain the solution

$$\zeta^{(1)} = \frac{q}{s\rho}\exp\left\{\frac{\rho}{2}\left[(1-2\eta)\mu - \sqrt{(1-2\eta)^2+4s}\right]\right\} \qquad (2.7)$$

Equation (2.7) implies that $\alpha_1(P) = 1/P$, hence the boundary condition (1.15) yields $\alpha_1(P) = 1/P$ (*). Using (1.13), we obtain for ζ_1 the equation

$$\Delta\zeta_1 = -\frac{q}{s}\left(\frac{1-2\eta}{r^2} - \frac{1}{r^5}\right)P_1(\mu)$$

whose general solution is

$$\zeta_1 = \frac{q}{s}\left(\frac{1-2\eta}{2} + \frac{1}{4r^3}\right)P_1(\mu) + \sum_{n=0}^{\infty}(a_n r^n + b_n r^{-n-1})P_n(\mu) \qquad (2.8)$$

Retaining in (1.15) terms of order $1/P$, we obtain the boundary condition for ζ_1

$$r = 1, \quad \zeta_1 = -\zeta_{+1} + \zeta_{+0}\eta\mu$$

which yields the linear relationship between coefficients a_n and b_n

$$b_0 = -a_0 - \zeta_{+1}, \quad b_1 = -a_1 - \frac{q}{s}\left(\frac{3}{4} - \eta\right) + \zeta_{+0}\eta \qquad (2.9)$$

$$b_n + a_n = 0, \qquad n = 2,\ 3,$$

To determine the unknown coefficients in (2.8) we carry out the joining of the first approximations $\zeta_{*1} = P^{-2}\zeta_0 + P^{-1}\zeta_1$ for $r \to \infty$ and $\zeta^{*1} = P^{-1}\zeta^{(1)}$ for $\rho \to 0$. Using the results of the joining operation and (2.9), we obtain for function ζ_1

$$\zeta_1 = -\frac{q}{2s}\sqrt{(1-2\eta)^2+4s} + \left(\frac{q}{2s}\sqrt{(1-2\eta)^2+4s} - \zeta_{+1}\right)\times \qquad (2.10)$$

$$\frac{1}{r} + \frac{1}{s}\xi_{1,1}P_1(\mu)$$

$$\xi_{1,1} = q\left(\frac{1-2\eta}{2} - \frac{3}{4r^2} + \frac{1}{4r^3}\right) + \frac{\eta}{r^2}$$

*) Editor's Note. Literal translation of the original Russian sentence.

It remains to determine ζ_{+1}. For this from (1.14) we obtain the equation

$$\Psi_+{}^2\zeta_{+1} = \frac{3}{2}\eta\int\limits_{-1}^{1}\mu\left(\frac{1}{s}-\zeta_0\right)\Big|_{r=1}d\mu - \frac{3}{2}\int\limits_{-1}^{1}\frac{\partial\zeta_1}{\partial r}\Big|_{r=1}d\mu -$$

$$\frac{3}{2}\eta\int\limits_{-1}^{1}\mu\frac{\partial\zeta_0}{\partial r}\Big|_{r=1}d\mu$$

whose solution, after the substitution of expressions from (2.4) and (2.10) for ζ_0 and ζ_1, respectively, is

$$\zeta_{+1} = \frac{q(1-q)}{2s}\sqrt{(1-2\eta)^2+4s} \tag{2.11}$$

The explicit form of ζ_1 can now be determined from (2.10)

$$\zeta_1 = \frac{q}{2s}\sqrt{(1-2\eta)^2+4s}\left(-1+\frac{q}{r}\right)+\frac{1}{s}\xi_{1,1}P_1(\mu) \tag{2.12}$$

3. The second approximation for ζ and ζ_+. It follows from (2.4) and (2.12) that $\alpha^{(2)}(P) = 1$. Using (2.1), from (1.17) we obtain for $\zeta^{(2)}$ the same problem as for $\zeta^{(1)}$, whose general solution is of the form (2.6). After the determination of coefficients A_n by joining $\zeta^{*2} = P^{-1}\zeta^{(1)} + \zeta^{(2)}$ for $\rho \to 0$ with $\zeta^{*1} = P^{-2}\zeta_0 + P^{-1}\zeta_1$ for $r \to \infty$, we obtain

$$\zeta^{(2)} = \frac{q}{2}\sqrt{(1-2\eta)^2+4s}\,\zeta^{(1)} \tag{3.1}$$

It follows from (3.1) and (1.14) that $\alpha_2(P) = \alpha_{+2}(P) = 1$. This makes it possible to obtain equations for second approximations of the internal expansion and the expansion of ζ_+. Solutions of these equations are derived by using the procedure similar to that described in Sect. 2. Expressions for ζ_{+2} and ζ_2 are not adduced here because of their unwieldiness. They will be taken into account in final formulas for ξ, ξ_+ and Sh.

4. Distribution of concentration. Inflow of matter to the closed circulation region. From the theorem on the relation between limit pre-image and image values, which for the considered Laplace transformation is of the form

$$\lim_{\tau\to\infty}\xi(r,\mu,\tau) = \lim_{s\to 0}P^2 s\zeta(r,\mu,s)$$

we find that in the case of homogeneous concentration distribution away from the bubble ($\eta = 0$) and total absorption of matter in the cloud ($q = 1$) with $\tau \to \infty$ a stationary concentration distribution is established, and that the latter is the same as calculated for this case in [4].

To determine the concentration field it is generally necessary to resort to the inverse Laplace transformation of derived functions $\zeta^{(n)}$, ζ_n and ζ_{+n}. For the external expansion ξ^* this yields (4.1)

$$\xi^* = q\exp\left(\frac{1-2\eta}{2}\rho\mu\right)\left\{\int\limits_0^{\tau}\exp\left(-\frac{\rho^2}{4P^2t}\right)\exp\left[-\frac{(1-2\eta)^2}{4}P^2t\right]\frac{dt}{2\sqrt{\pi t^3}}+\right.$$

$$P\frac{q}{2}\int\limits_0^{\tau}\Phi(P^2t,\eta)\exp\left[-\frac{(1-2\eta)^2}{4}P^2(\tau-t)\right]\times$$

$$\left.\exp\left[-\frac{\rho^2}{4P^2(\tau-t)}\right]\frac{dt}{\sqrt{\pi(\tau-t)^3}}\right\}+O(P^3)$$

where

$$\Phi(P^2\tau, \eta) = \frac{1}{\sqrt{\pi P^2 \tau}} \exp\left[-\frac{(1-2\eta)^2}{4} P^2\tau\right] + \frac{1-2\eta}{2} \operatorname{erf}\left(\frac{1-2\eta}{2} P\sqrt{\tau}\right) \tag{4.2}$$

Function ξ in the proximity of the closed circulation region (internal asymptotic expansion) is of the form

$$\xi_* = \frac{q}{r} + P\left[\Phi(P^2\tau, \eta)\left(-q + \frac{q^2}{r}\right) + \xi_{1,1}P_1(\mu)\right] + P^2\xi_2 + O(P^3) \tag{4.3}$$

$$\xi_2 \approx -\frac{\xi_{+2}(\tau)}{r} + \sum_{n=0}^{2} \xi_{2,n}(r) P_n(\mu) \tag{4.4}$$

$$\xi_{2,0} = q\left[\frac{(1-2\eta)^2}{6} r - \frac{(1-2\eta)^2}{4} q + \frac{S_0}{r} + \frac{1-2\eta}{24r^2} - \frac{1}{16r^4} + \frac{1}{60r^5}\right] - \frac{T_0}{r} + \frac{\eta}{12r^4}$$

$$\xi_{2,1} = 2q\Phi(P^2\tau, \eta)\left(-\frac{1-2\eta}{4} r + \frac{1-2\eta}{4} q + \frac{S_1}{r^2} + \frac{q}{8r^3}\right)$$

$$\xi_{2,2} = q\left[\frac{(1-2\eta)^2}{12} r - \frac{1-2\eta}{4r} + \frac{5(1-2\eta)}{24r^2} - \frac{S_2}{r^3} - \frac{1}{8r^4} + \frac{5}{168r^5}\right] - \frac{\eta(1-2\eta)}{3r} - \frac{T_2}{r^3} + \frac{\eta}{6r^4}$$

$$S_0 = \frac{(1-2\eta)^2}{4} q - \frac{(1-2\eta)^2}{6} - \frac{1-2\eta}{24} + \frac{11}{240}$$

$$T_0 = \frac{\eta}{12} + \frac{\eta^2(1-q)}{6}, \quad S_1 = \frac{1-2\eta}{4} - \frac{q(3-4\eta)}{8} + \frac{\eta(1-r)}{2}, \quad S_2 = \frac{(1-2\eta)^2}{12} - \frac{1-2\eta}{24} - \frac{2}{21}$$

$$T_2 = \frac{\eta(5-2\eta)}{6} + \frac{\eta^2(1-q)}{3}$$

where $\xi_{1,1}$ is defined by formula (2.10) and ξ_{+2} by expression (4.6) (see below). The approximate expression for ξ_2 was obtained by inverse Laplace transformation of ζ_2.

The asymptotic expression for function $\xi_+(\tau)$ is of the form

$$\xi_+(\tau) = 1 - q + Pq(1-q)\Phi(P^2\tau, \eta) + P^2\xi_{+2} + O(P^3) \tag{4.5}$$

where function ξ_{+2} is the pre-image of $P^{-2}\zeta_{+2}(s)$, whose approximate expression is

$$\xi_{+2} \approx \frac{1}{24}(1-2\eta)^2 q(1-q)(6q-7) + \frac{1}{2}\eta(1-q) \times \left[\eta + \frac{1}{2} + \frac{q}{3}(\eta - 1) + \frac{2}{3}\frac{1}{1-\delta}\right] \tag{4.6}$$

Concentration distribution c outside the closed circulation region and concentration c_+ inside it is determined by formulas (1.10) with the use of obtained expansions (4.1)–(4.6).

We define the cloud mass exchange by the mean Sherwood number in which the "initial" concentration c_0 is chosen as the characteristic value. In dimensionless variables the mean Sherwood number is of the form

$$\mathrm{Sh} = \frac{1}{2}\int_{-1}^{1} \left.\frac{\partial c}{\partial r}\right|_{r=1} d\mu \tag{4.7}$$

or, using (1.10)

$$\mathrm{Sh} = \exp\left(-P^2\eta\frac{1}{1-\delta}\tau\right)\mathrm{Sh}^\circ \tag{4.8}$$

$$\mathrm{Sh}^\circ = \frac{1}{2}\left[P\eta\int_{-1}^{1}\mu e^{P\eta\mu}(1-\xi_*)\big|_{r=1}d\mu - \int_{-1}^{1}e^{P\eta\mu}\frac{\partial\xi_*}{\partial r}\Big|_{r=1}d\mu\right]$$

It is convenient to express the Sherwood number in the form of an asymptotic expansion in the Péclet number. Taking into account the expression for ξ_2 and the asymptotic expansion of exponents in (4.7), from (4.1) – (4.5) we obtain

$$\mathrm{Sh}^\circ = q + Pq^2\Phi(P^2\tau,\ \eta) + P^2\mathrm{Sh}_2^\circ + O(P^3) \tag{4.9}$$

$$\mathrm{Sh}_2^\circ \approx \frac{\eta^2(7-2q)}{6} - \frac{\eta(1-q)}{4} + \xi_{+2}(\tau) -$$

$$q\left[(1-2\eta)^2\frac{4-3q}{12} - \frac{1-2\eta}{24} + \frac{29}{240}\right]$$

5. Region of applicability of results. The derived expansions of ξ, ξ_+ and Sh° in Péclet number are valid for any η, q and $\delta \neq 1$. The method of asymptotic expansions capable of being joined does not permit the determination of the upper limit of Péclet numbers for which expansions (4.1), (4.3), (4.5) and (4.9) are valid. However the procedure of solution derivations makes it reasonable to expect a widening of the limits of solution by Péclet numbers with decreasing parameters η and δ.

In the derivation of solution the effect of walls and of the inlet section of the reactor on the diffusion process was neglected here. Owing to the distortion of the cloud shape and the necessity to allow for diffusion transport in the process of bubble formation, it is extremely difficult to take fully into account the effect of the inlet section. It can be shown, however, that if the initial concentration distribution for a formed bubble at a distance $r > O(1/P)$ differs only slightly from the concentration (1.2) in the absence of a bubble, the effect of the inlet section for times $\tau > O(1/P^2)$ introduces in the terms of expansions $O(P^2)$ only an insignificant correction. Otherwise the derived expansions are valid up to and including terms $O(P)$.

In practice in a number of cases the formation of a stable bubble takes place at distances of the order of one or two radii from the reactor inlet, as the result of coalescence of very small bubbles. Inside these and in the surrounding medium the concentration levels out almost instantaneously and the effect of the inlet section on the formed bubble can be neglected. Hence it is possible to use the derived solution with an accuracy to within terms $O(P^2)$.

The derived solution may also be used for analyzing the mass exchange between an isolated bubble and a medium in laboratory reactors in which the introduction of the bubble into a homogeneous fluidized layer is done by means of a special device and at considerable distance from the reactor inlet. In that case the effect of the inlet section is negligible, the time of bubble formation is short, the concentration distribution at the initial instant of time is noticeably different from (1.2) only at small distances from the bubble, and the solution is valid up to terms $O(P^2)$.

The assumption of constancy of bubble dimensions during its ascent in the reactor is important, since growth and fractionation of bubbles is often observed. Assumption about the isotropy of effective diffusion coefficients in the continuous phase is also important.

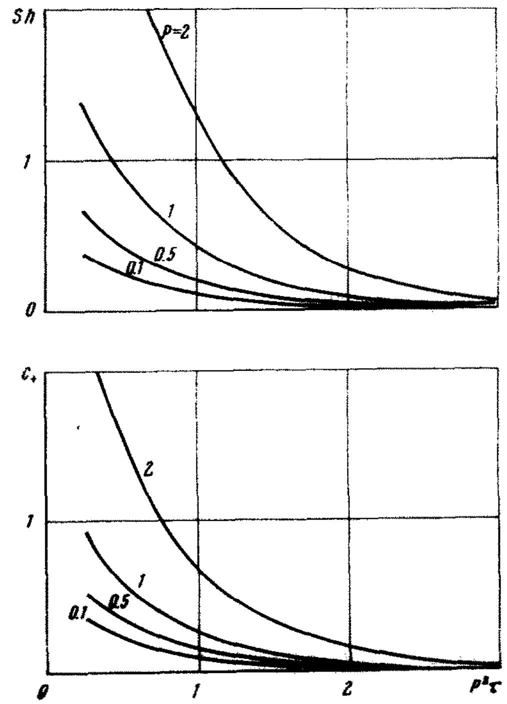

Fig. 1

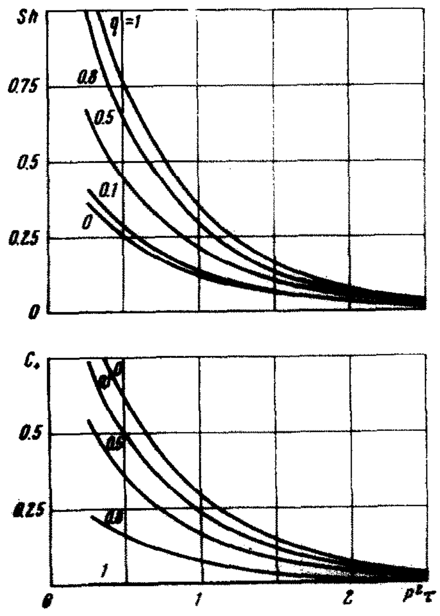

Fig. 2

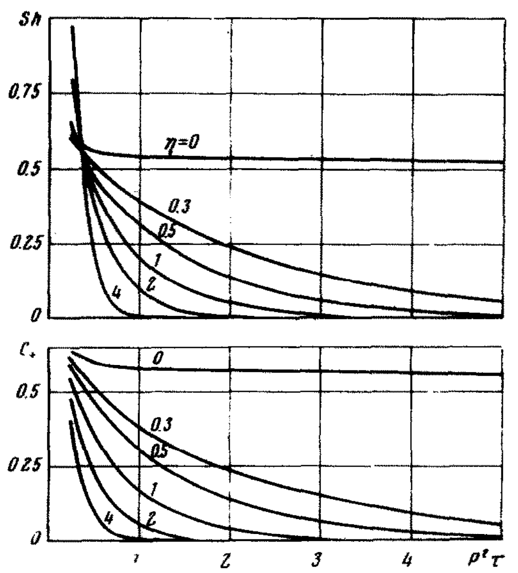

Fig. 3

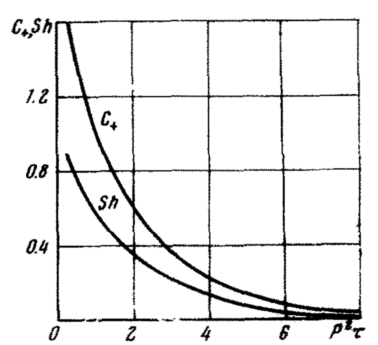

Fig. 4

The dependence of concentration inside the cloud and of the mean Sherwood number on time for $\delta = 0.333$ and several values of parameters P, q and η is shown in Figs. 1–3. Figure 1 illustrates the effect of the Péclet number on the time-dependence of concentration inside the cloud and of the mean Sherwood number for $q = 0.5$ and $\eta = 1$. The dependence of solution on the reaction rate constant inside the cloud for $P = 0.5$ and $\eta = 1$ is shown in Fig. 2. The form of functions $c_+ (P^2\tau)$ and $\mathrm{Sh}(P^2\tau)$ for several η (i.e. the degree of the concentration profile irregularity in the reactor) is shown in Fig. 3.

The derived solution for $\eta = 0$ and $q = 1$ corresponds to the heat and mass exchange between a sphere and a uniform stream of perfect fluid in conditions of total absorption of matter at the surface of the sphere (infinitely fast reaction on the surface abruptly beginning at instant $\tau = 0$). A stationary mode is established with $\tau \to \infty$, and the formulas for concentration distribution and mass exchange convert to those derived in [5] for that case.

The solution is considerably simplified in the case of systems of small particles for which the ratio v_0 / u_b is, as a rule, very small. The circulation region is virtually the same as the bubble and does not contain particles, hence $q = 0$. In that case

$$c_+ = \exp\left(-\eta \frac{1}{1-\delta} P^2\tau\right)\left\{1 + P^2\left[\frac{\eta}{3(1-\delta)} + \frac{\eta(3 \mp 8\eta)}{12}\right] + O(P^2)\right\}$$

$$Sh = P^2 \exp\left(-\eta \frac{1}{1-\delta} P^2\tau\right)\left[\frac{\eta}{3(1-\delta)} + \frac{\eta^2}{2} + O(P)\right]$$

Let us consider in conclusion a model example. Numerical value of parameters appearing in this example correspond as to their order of magnitude to specific systems cited in publications.

Let the initial rate of fluidization be $v_0 = 15$ cm/sec (e.g. glass balls of approximately 0.3 mm diameter fluidized by air). For a bubble of radius $a_b = 1$ cm, we have $u_b \approx 30$ cm/sec, $\delta = 0.5$, and $a_c = 1.59$ cm. We assume the effective diffusion coefficient to be $D = 11.9$ cm^2/sec, so that $P = a_c (u_b - v_0) / D = 2$. Let $k = 5.9$ sec^{-1}. From this, using formula (1.2), we obtain $\eta = 0.25$. We determine the effective constant of the reaction rate inside the bubble using the rough approximation based on the assumption that $k_+ = kV_c / (V_c + V_b)$, where V_c and V_b are the volumes of the cloud and the bubble, respectively. Then $k_+ = 3k\delta / (1 + 2\delta) = 4.42$ sec^{-1}, $\Psi_+^2 = 0.94$ and $q = 0.24$. The dependence of c_+ and Sh on time for this system is shown in Fig. 4. It will be seen that in this case the mass exchange between the bubble and the continuous phase at distances from the inlet not exceeding by one or two orders of magnitude the bubble dimensions, is essentially unsteady.

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Translated by J.J.D.

UDC 532.529

EQUATIONS OF KINETICS FOR AGGREGATION PROCESSES IN SUSPENSIONS

PMM Vol.39, №1, 1975, pp.130-143
A.S.POPEL, S.A.REGIRER and N.Kh.SHADRINA
(Moscow, Leningrad)
(Received January 2, 1974)

Generalized kinetic equations for determining the aggregation of particles in suspensions are derived with allowance for dispersion and multiple and exchange interactions. A system of equations is derived in a general form for moments of the distribution function, and a method for determining equilibrium distribution is indicated. Some exact solutions, including self-similar, of the proposed kinetic equations are obtained.

Physical properties of many suspensions substantially depend on the processes of aggregation and dispersion of suspended particles. Such processes are defined by special kinetic equations, an example of which is the equation of drop coagulation (see [1]). The latter takes into account only one aggregation process, viz., the amalgamation of drops produced by double collisions. Theories which take into account also the dispersion of particles (see, e.g., [2]) are known. However for some systems with high concentration of suspended particles such as, for instance, blood in which erythrocytes occupy about half of the volume, it is necessary to take into consideration a more complex interaction between particles.

Thus in a concentrated suspension the determining effect may be that of collisions other than double, which in the case of blood become significant for an erythrocyte concentration $H \geqslant 5\%$ [3]. Besides aggregation and dispersion of particles, exchange interactions are possible when two or more particles not identical to the original ones are formed as the result of collisions (*). If under certain conditions there exists a limit dimension for the aggregate but with possible collisions of arbitrary particles, exchange interactions must necessarily occur.

The above phenomena are taken into consideration in the kinetic equation which is derived and analyzed below in Sects. 1 – 4 and 7. Certain exact solutions of that equation are presented in Sects. 5 and 6.

1. The kinetic equation. Let us consider a suspension in the form of a mixture of a "carrier" fluid and suspended particles which may coalesce into aggregates of any arbitrary form as the result of effective collisions, i.e. leading to the sticking to-

*) This was brought to the attention of the authors by A.G.Kulikovskii.