EXTRINSIC PROBLEM OF MASS EXCHANGE FOR A SOLID SPHERE AND A DROP AT HIGH PECLET NUMBERS AND Re  $\leq$  100

#### G. M. Ryskin and G. A. Fishbein

Based on the solution of the problem of flow around a liquid sphere for  $0.5 \le 100$ , values are found for the mass-transfer coefficients for a solid sphere, a drop, and a gas bubble in diffusion boundary-layer approximation.

The calculation of the process of mass exchange between a spherical particle and a continuous medium at high values of the Peclet number usually is carried out in approximation of diffusion boundary-layer theory. These solutions are well known for particles moving under Stokes conditions (Re < 1). Formulas for the Nusselt criterion of a solid sphere, drop, and bubble, when Re < 1 and for high Peclet values obtained in diffusion boundary-layer approximation in papers by various authors, are shown in [1]. The present paper considers mass exchange in the region of high Reynolds numbers, 0.5 < Re < 100.

We apply the Prandtl-Mises transformation to the diffusion boundary-layer equation at the surface of a sphere

$$v_r \frac{\partial C}{\partial r} + \frac{v_{\theta}}{r} \cdot \frac{\partial C}{\partial \theta} = \frac{2}{\text{Pe}} \cdot \frac{\partial^2 C}{\partial r^2}; \qquad (1)$$

i.e., we convert from the variables r,  $\theta$  to  $\psi$ ,  $\theta$  and we expand the flow function near the boundary in Taylor series, preserving the first nonvanishing term of the series. Then we obtain for  $\psi$  the following expressions:\*

for the drop

$$\psi_1 = -v_0 y \sin \theta, \tag{2}$$

for the solid sphere

$$\psi_2 = -\frac{1}{2}\zeta_0 y^2 \sin\theta, \qquad (3)$$

where y = r - 1(y <<1);  $v_0$  is the velocity and  $\zeta_0$  is the vorticity at the surface of the sphere. Conversion to dimensionless quantities in Eqs. (1)-(3) is effected by introducing scaling-ratios: for the velocity – the velocity remote from the particle; for distance – the radius of the sphere; and for concentration – the difference between the concentrations at the surface of the sphere and remote from it.

After this transformation with conditions of constancy of the concentrations at the surface of the sphere and in the core of the stream, Eq. (1) assumes the form of the thermalconductivity equation; the problem becomes self-similar and its solution leads to the following expressions for the Nusselt criterion:

\*Formulas (2) and (3) can be obtained easily, by using the expressions for the velocity and vorticity at the boundary in terms of the flow function

$$v_0 = -\frac{1}{\sin\theta} \left(\frac{\partial\psi}{\partial r}\right)_{r=1}; \quad \zeta_0 = -\frac{1}{\sin\theta} \left(\frac{\partial^2\psi}{\partial r^2}\right)_{r=1}.$$

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Fig. 1. Vorticity distribution at the surface of a solid sphere: 1) Re = 0.5; 2) Re = 20; 3) Re = 50; 4) Re = 100.



Fig. 2. Velocity distribution of liquid over the surface of a drop: 1) Re = 0.5,  $\mu$  = 1; 2) 20, 0; 3) 20, 1; 4) 20, 3; 5) 100, 0; 6) 100, 1; 7) 100, 3.

(4)

(5)

for the drop

$$Nu_1 = \varphi(v_0) \sqrt{Pe}$$

for the solid sphere

where

$$\operatorname{Nu}_{2} = f(\zeta_{0})^{3}/\overline{\operatorname{Pe}}$$

$$\varphi(v_0) = \left(\frac{2}{\pi}\int_0^{\theta_s} v_0 \sin^2\theta d\theta\right)^{\frac{1}{2}}; \quad f(\zeta_0) = 0.641 \left(\int_0^{\theta_s} \sqrt{\zeta_0 \sin^3\theta} d\theta\right)^{\frac{2}{3}}$$

Here  $\theta_s$  is the angle of flow separation (for nonseparating streamline flow  $\theta_s = \pi$ ).

Formula (4) is derived in [2] and an expression similar to Eq. (5) is obtained in [3].

In the limiting case of small Re values, by substituting in Eq. (4) and (5) the values of  $v_0$  and  $\zeta_0$  for Stokes streamline flow conditions, we obtain for Nu the very well-known formulas

$$Nu_{I} = \frac{0.65}{\sqrt{1+\mu}} \sqrt{Pe}, \qquad (6)$$

$$Nu_2 = 0.99 \sqrt[3]{Pe}.$$
 (7)

It should be noted that the flow function represented in the form of Eq. (2) is not valid when  $\mu \rightarrow \infty$ . Expansion of Eq. (2) postulates constancy of the tangential component of the velocity across the boundary layer, which is approximately true only for small values of  $\mu$  (the greater is Pe, the greater is the validity of this assumption for large values of  $\mu$ ). Because of this, formulas (4) and (6) can be used in the case of not very large values of  $\mu$ . This can be seen also from the fact that when  $\mu \rightarrow \infty$  they do not give a limiting transition in formulas (5) and (7). Thus, for example, when Pe = 10<sup>4</sup>, formula (6) even for  $\mu > 8$  gives a value of Nu which is lower than for a solid sphere according to formula (7).

It follows from expressions (4) and (5) that for solving the extrinsic problem of mass exchange it is necessary to know the velocity distribution or the vorticity at the surface of the drop or the solid sphere, respectively. These quantities for Re > 1 can be obtained from the solution of the Navier-Stokes equations for the problem of flow around a spherical

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Fig. 3. Dependence of the coefficients f and  $\varphi$  on Re and  $\mu$ : 1) for a solid sphere; 2) for a gas bubble ( $\mu = 0$ ); 3, 4, 5) for a drop when  $\mu = 0.333$ , 1, and 3, respectively. I)  $\mu = 0.38$ ; II)  $\mu = 0.42$ ; III)  $\mu = 2.6$  (I-III are Griffith's experimental data [6]).

drop (solid sphere and gas bubble are limiting cases of this problem). The formulation and method for the numerical solution of this problem are given in [4]. Further improvement of the method of solution and the use of the BÉSM-6 computer increase considerably the accuracy of the calculation. Numerical calculations have been carried out for  $0.5 \le \text{Re} \le 100$  and  $\mu = 0$ , 0.333, 1, 3, and  $\infty$ .

Figures 1 and 2 show the vorticity distribution at the surface of a sphere and the velocity at the surface of a drop for several computed alternatives. The coefficients  $f(\zeta_0)$ (curve 1) and  $\varphi(v_0)$  (curves 2-5), obtained by integration of the corresponding expressions [see formulas (4) and (5)] and which are necessary for calculating the Nusselt criterion, are plotted in Fig. 3.

With the values of  $\pi$  and Re considered, the flow around the drop is nonseparable and integration in formula (4) is carried out over the limits from 0 to  $\pi$ . For a solid sphere, flow separation is observed even when Re  $\approx 20$ . Curve 1 is plotted by taking account of the mass exchange in the breakaway zone. It has been assumed that in the breakaway zone a diffusion boundary layer is formed and that the mass exchange between a combined vortex and the external flow is quite intensive, so that the concentration in the leading flow is equal to the concentration remote from the drop (the point of advance of the flow in the breakaway zone is the point  $\theta = \pi$ ). The total diffusion flow is defined as the sum of the flows in the boundary layers up to the point of breakaway and in the zone of flow separation. The dashed part of curve 1 corresponds to the solution without taking account of mass exchange in the breakaway zone.

It can be seen from Fig. 3 that when Re < 1 formulas (6) and (7) can be used for the Nusselt criterion. For fixed values of Pe, with a change of Re from 0.5 to 100, the coefficient of mass exchange for a solid sphere increases approximately by a factor of 1.6. For a drop, the effect of Re on the rate of transfer becomes more obvious with increase of  $\mu$ .

At large values of Re and  $\mu \rightarrow 0$ , it can be supposed that the streamline flow differs only slightly from ideal. Then, substituting the expression for v<sub>0</sub> for the ideal flow around the sphere in formula (4), we obtain the well-known Boussinesq solution [5]

$$Nu = 1.13 \sqrt{Pe}$$
 (8)

It is clear that the number 1.13 is the upper limit for the function  $\Psi(v_0)$ . It can be seen from Fig. 3 that when Re = 100, the value for the gas bubble differs from this maximum value in all by 15%.

The experimental data of Griffith [6] are also plotted in Fig. 3 for a drop with a viscosity ratio of  $\mu = 0.38$ , 0.42, and 2.6. For a solid sphere, the experimental data in a number of papers have been processed in the form of a correlation function of the type

$$Nu_{2} = 2 + \alpha \operatorname{Re}^{1/2} \operatorname{Pr}^{1/3} = 2 + \alpha \operatorname{Re}^{1/6} \operatorname{Pe}^{1/3}.$$
(9)

For the coefficient  $\alpha$ , values of 0.55, 0.95, and 0.72 were obtained in [7-9], respectively. When Pe >> 1, it follows from Eq. (9) that

$$\frac{\mathrm{Nu}_2}{\sqrt[3]{\mathrm{Pe}}} = \alpha \,\mathrm{Re}^{1/6}.$$
(10)

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Comparison of relation (10) with the quantity  $f(\zeta_0)$  [see formula (5)], shows that when Re > 10, the best agreement is when  $\alpha = 0.72$ .

#### NOTATION

r, $\theta$ , spherical coordinates;  $\psi$ , flow function;  $v_r$ ,  $v_{\theta}$ , velocity components;  $\zeta$ , vorticity; C, concentration;  $\mu = \mu_d/\mu_c$ , ratio of viscosities of the dispersed and continuous phases; U, velocity of steady motion of liquid;  $\alpha$ , radius of sphere;  $\nu$ , kinematic viscosity of medium; D, coefficient of diffusion; k, mass-transfer coefficient; Nu =  $2k\alpha/D$ , Nusselt number; Pe = 2Ua/D, Peclet number; Re = 2Ua/v, Reynold's number; Pr = v/D, Prandtl number. Indices: 1, drop; 2, solid sphere; d, dispersed phase; c, continuous phase; s, point of flow separation; 0, values at the surface of the sphere.

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# UNSTEADY MASS TRANSFER WITH A HETEROGENEOUS CHEMICAL

REACTION DURING LAMINAR FLOW PAST A SPHERE

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Unsteady mass transfer toward a solid sphere is investigated in the region of Peclet numbers  $1 \le Pe < 1000$ . Diffusion flow in the presence of a first-order chemical reaction is calculated and the relaxation time of the steady regime as a function of the Peclet number is determined.

The process of mass transfer between a moving spherical particle and a continuous flow was investigated earlier in a quasistationary approximation for limiting cases of small and large Peclet (Pe) values. In [1, 2] solutions were obtained for small Pe values by the method of joining asymptotic expansions [3]. Although theoretically this method is suitable only for Pe < 1, the results of such calculations were used sometimes also for Pe > 1. Solutions obtained in an approximation of the theory of a diffusion boundary layer are known for large Pe (see, for example, [4, 5]). In the transition region of Peclet numbers (1 < Pe ~ 100), when the diffusion boundary layer has still not formed and the contribution to the magnitude of the diffusion flow from the molecular and convective terms in the transfer equation is commensurable, the field of concentrations cannot be determined by a single one of the approximate methods. In [6] the problem of steady mass transfer was solved for the particular case of evaporation of water drops by the finite-difference method for 0 < Pe  $\leq$  200. In this article we will consider the most general case when the transfer process is unsteady and a chemical reaction occurs on the surface of the particle.

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