NONSTATIONARY DIFFUSION FLOW TO A MOVING DROP AT LOW REYNOLDS NUMBERS

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The process of nonstationary convective diffusion to a moving drop at low Re is examined. Equations for the dimensionless diffusion flow Nu to the surface of the drop and for the length of the nonstationary region and the time required for the establishment of steady-state diffusion conditions are obtained.

Levich [1] examined steady-state convective diffusion to a moving drop at Re < 1 and obtained an equation for the dimensionless diffusion flow to the surface of the drop

$$\mathrm{Nu} = \frac{2}{\sqrt{6\pi}} \mathrm{Re}^{1/2}\mathrm{Pr}^{1/2} \left(\frac{\mu}{\mu+\mu'}\right)^{1/2}. \tag{1}$$

The velocity u of the drop is calculated from the Hadamard-Rybchinskii formula, application of which is restricted to the region Re < 1:

$$u = \frac{2}{3} g a^2 \frac{\rho' - \rho}{\mu} \frac{\mu+\mu'}{2\mu+3\mu'}. \tag{2}$$

Regarding the diffusion process as unsteady and assuming the phase contact time to be short, Higbie [2] obtained the mass transfer coefficient in the form

$$k = 2(D/\pi t)^{1/2}. \tag{3}$$

It is clear from Eq. (3) that at the initial instant ($t \to 0$) the mass transfer coefficient can be arbitrarily large. As the solution at the surface of the drop is depleted (with increase in t), the density of the diffusion flow decreases. Then, convective mass transfer, which was not considered in deriving (3), begins to have a significant effect. Higbie's theory does not permit an estimate of the time during which formula (3) still gives reliable practical results.

In this paper nonstationary mass transfer to a moving drop at low Re is examined and the time required for the establishment of steady-state conditions is estimated.

The concentration distribution in the nonstationary laminar boundary layer of a spherical drop is given by the equation

$$\frac{\partial c}{\partial t} + v_r \frac{\partial c}{\partial r} + \frac{v_\theta}{r}\frac{\partial c}{\partial \theta} = D\frac{\partial^2 c}{\partial r^2}. \tag{4}$$

We use the Prandtl-Mises transformation [3] and as a new independent variable instead of r we introduce the stream function

$$\psi = v_0 a y \sin^2\theta. \tag{5}$$

The quantity v_0 in (5) is the velocity of the liquid on the drop surface,

$$v_0 = \frac{u}{2}\frac{\mu}{\mu+\mu'}. \tag{6}$$

In the boundary layer of the drop we have $y \approx 0$ and $r = a + y \approx a$, $v_\theta = v_0 \sin\theta$.

In view of the above, in the variables (t, ψ) Eq. (4) takes the form

$$\frac{\partial c}{\partial t} + \frac{v_0}{a}\sin\theta \frac{\partial c}{\partial \theta} = D a^2 v_0^2 \sin^4\theta \frac{\partial^2 c}{\partial \psi^2}. \tag{7}$$

We will seek the solution of this equation by the method of successive approximations and write

$$C=\sum_{1}^{\infty} C_i,$$

where C_1 is the solution of the diffusion equation in a stationary medium, and C_2, C_3, . . . are certain small corrections.

When $t \to 0$ the convective term in Eq. (7) can be omitted. We then obtain the following equation:

$$\frac{\partial C_1}{\partial t}=Da^2v_0^2\sin^4\theta\,\frac{\partial^2 C_1}{\partial\psi^2}. \tag{8}$$

The solution of Eq. (8) satisfying the boundary conditions

$$C_1=C_0,\quad \psi\to\infty,$$
$$C_1=C^*,\quad \psi\to 0,$$
$$C_1=C_0,\quad t=0,$$

is

$$C_1=\frac{2(C_0-C^*)}{\sqrt{\pi}}\int\limits_0^{\psi/\sqrt{Bt}}\exp(-z^2)\,dz+C^*, \tag{9}$$

where $B = Da^2v_0^2 \sin^4\theta$.

Differentiating (9) with respect to θ with ψ = const, we find the derivative

$$\frac{\partial C_1}{\partial\theta}=-\frac{2(C_0-C^*)}{\sqrt{\pi}}\,\frac{\psi}{av_0}\,\frac{\cos\theta}{\sin^3\theta}\exp(-z^2),$$

where $z=\psi/2\sqrt{Bt}$.

In the boundary layer of the drop $\psi \approx 0$, and hence $z \approx 0$ and $\exp(-z^2) \approx 1$. Thus we finally obtain

$$\frac{\partial C_1}{\partial\theta}\simeq-\frac{2(C_0-C^*)}{\sqrt{\pi}}\,\frac{\psi}{av_0}\,\frac{\cos\theta}{\sin^3\theta}. \tag{10}$$

The correction C_2 is given by the equation

$$\frac{\partial C_2}{\partial t}-\frac{2(C_0-C^*)}{a^2\sqrt{\pi Dt}}\,\psi\,\frac{\cos\theta}{\sin^2\theta}=Da^2v_0^2\sin^4\theta\,\frac{\partial^2 C_2}{\partial\psi^2}. \tag{11}$$

In deriving this equation we neglected the term

$$\frac{v_0}{a}\sin\theta\,\frac{\partial C_2}{\partial\theta}\ll Da^2v_0^2\sin^4\theta\,\frac{\partial^2 C_2}{\partial\psi^2},$$

which is valid when $\psi \to 0$.

Transforming Eq. (11) by the Laplace method, we obtain an equation with total derivatives for the transform of the required function. The solution of this last equation has the form

$$\bar{C}_2=\alpha_1\exp\left(\frac{\psi}{av_0\sin^2\theta}\sqrt{\frac{s}{D}}\right)+\alpha_2\exp\left(-\frac{\psi}{av_0\sin^2\theta}\sqrt{\frac{s}{D}}\right)+$$
$$+2(C_0-C^*)\frac{\cos\theta\psi}{a^2\sin^2\theta\sqrt{D}}\,s^{-1.5}.$$

For the transform $\bar{C}_2$ we have the following boundary conditions: $\bar{C}_2(0) = 0$, $\bar{C}_2(\infty) = 0$. Using these, we find the constants of integration $\alpha_1 = \alpha_2 = 0$. We finally obtain

$$\bar{C}_2=2(C_0-C^*)\frac{\psi\cos\theta}{a^2\sin^2\theta\sqrt{D}}\,s^{-1.5}. \tag{12}$$

Using tables of transforms and their originals [4], we find the value of the function

$$C_2 = 4\frac{C_0 - C^*}{a^2}\sqrt{\frac{t}{\pi D}}\frac{\cos\theta}{\sin^2\theta}\psi. \tag{13}$$

Substituting ψ from Eq. (5) in (9) and (13), we find

$$C_1 = \frac{2(C_0 - C^*)}{\sqrt{\pi}}\int_0^{y/2\sqrt{Dt}} \exp(-z^2)\,dz + C^*,$$
$$C_2 = 4(C_0 - C^*)\frac{v_0}{a}\sqrt{\frac{t}{\pi D}}\,y\cos\theta. \tag{14}$$

By repeating the calculation used to determine C_2, we can find all the remaining corrections C_i, and for even values i = 2n we obtain

$$C_{2n} = (C_0 - C^*)\frac{v_0}{a}\sqrt{\frac{t}{\pi D}}\left(\frac{v_0 t}{a}\right)^{2n-2}\left[\frac{2^{4n-2}}{1\cdot 3\cdot 5 \ldots (4n-3)}\cos\theta\right]y. \tag{15}$$

For odd values i = 2n + 1 we have

$$C_{2n+1} = (C_0 - C^*)\frac{v_0}{a}\sqrt{\frac{t}{\pi D}}\left(\frac{v_0 t}{a}\right)^{2n-1}\times$$
$$\times\left[\frac{2^{4n-1}}{1\cdot 3\cdot 5 \ldots (4n-1)}(1+\cos^2\theta)\right]y. \tag{16}$$

The total mass flow to the surface of the drop in time t, measured from the start of the process (t = 0), is

$$I = \frac{2\pi a^2}{t}D\left[\int_0^t\int_0^\pi\left(\frac{\partial C_1}{\partial y}\right)_{y=0}\sin\theta\,d\theta\,dt + \int_0^t\int_0^\pi\sum_2^\infty\left(\frac{\partial C_i}{\partial y}\right)_{y=0}\sin\theta\,d\theta\,dt\right] \tag{17}$$

Substituting in (17) the values of the derivatives $\left(\frac{\partial C_i}{\partial y}\right)_{y=0}$, calculated from Eqs. (14), (15), and (16), we find the dimensionless diffusion flow to the surface of the drop

$$\mathrm{Nu} = \frac{2}{\sqrt{\pi}}\mathrm{Re}^{1/2}\mathrm{Pr}^{1/2}\left(\frac{\mu}{\mu+\mu'}\right)^{1/2}F(h), \tag{18}$$

where

$$F(h) = h^{-1/2}\left[1 + \frac{2}{3}\sum_1^\infty\frac{(2h)^{2n}}{1\cdot 3\cdot 5 \ldots (4n+1)}\right],$$
$$h = \frac{\mu}{\mu+\mu'}H,\ H = \frac{ut}{a},\ \mathrm{Nu} = \frac{ka}{D},\ k = \frac{I}{4\pi a^2(C_0 - C^*)}. \tag{19}$$

Function F(h) in (18) is a correction for the unsteadiness of the process. When $t \to 0$, Eq. (18) takes the form

$$\mathrm{Nu} = \frac{2}{\sqrt{\pi}}\mathrm{Re}^{1/2}\mathrm{Pr}^{1/2}H^{-1/2}. \tag{20}$$

By expanding the criteria in Eq. (20), we can easily verify that this expression reduces to Higbie's formula (3).

However, when $t \to \infty$, Eq. (18) does not lead to Levich's steady-state solution (1). This understandable, since when $t \to \infty$ the assumption of a convective flow small in comparison with the molecular flow, an assumption on which the derivation of Eq. (18) was based, becomes erroneous. It is obvious that the order of magnitude of the characteristic time T at which the above assumption can no longer be regarded as valid must be found from Eq. (19). In fact, the series $\frac{2}{3}\sum_{1}^{\infty}\frac{(2h)^{2n}}{1\cdot 3\cdot 5\ldots(4n+1)}$ in the square brackets in Eq. (19) is a correction to Higbie's equation for convective mass transfer. The equality

$$1=\frac{2}{3}\sum_{1}^{\infty}\frac{(2h)^{2n}}{1\cdot 3\cdot 5\ldots(4n+1)}$$

is the condition under which the convective flow becomes equal to the molecular mass transfer, i.e., may serve for an estimate of the order of magnitude of T.

In view of the rapid convergence of the series on the right-hand side of Eq. (21), we can restrict ourselves to the first two terms of this series. The solution of the equation obtained is h ≈ 2.1, whence $T\simeq 2.1\frac{a}{u}\frac{\mu+\mu'}{\mu}$. A more accurate estimate of T can be obtained from the condition $\frac{\partial F(h)}{\partial h}=0$, which gives the minimum of the function F(h). As calculations show, the minimum value of function F(h) is reached when h ≈ 1. When h = 1 is substited in Eq. (18),

$$\mathrm{Nu}=\frac{106}{45\sqrt{\pi}}\mathrm{Re}^{1/2}\mathrm{Pr}^{1/2}\left(\frac{\mu}{\mu+\mu'}\right)^{1/2},$$

i.e., a value approximately 2.9 times greater than that given by Levich's equation (1) for steady-state mass transfer.

The presence of a minimum of function F(h) when h = 1 leads to a cessation of the regular decrease in Nu with increase in t and even to some increase in the intensity of the diffusion process (when h > 1). This is a result of the unjustified extension of the approximate Eq. (18) into the region of large t, where it is invalid. Analysis shows that the contradiction noted is due to the assumption that the convective flow is small in comparison with the molecular flow, an assumption used in the derivation of Eq. (18).

At large t the terms $v_r\frac{\partial c}{\partial r}+\frac{v_\theta}{r}\frac{\partial c}{\partial\theta}$, in the equation of convective diffusion [4] are no longer small, but the term $\partial c/\partial t$ is and may be dropped. In view of the foregoing, the order of magnitude of the characteristic time T can be determined from the condition h = 1:

$$T\simeq\frac{a}{u}\frac{\mu+\mu'}{\mu}. \tag{22}$$

We can use Eq. (22) to estimate the order of the length of the region characterized by nonstationary diffusion:

$$l\simeq a(1+\mu'/\mu). \tag{23}$$

Substituting in (22) the values of u from formula (2), we find

$$T\simeq\frac{3}{2}\frac{2\mu+3\mu'}{\rho'-\rho}\frac{1}{ga}. \tag{24}$$

The results obtained can be used in the case of motion of drops at Re < 1 (this restriction is a result of using the Hadamard-Rybchinskii formula in the calculations). Substituting in the inequality Re < 1 the velocity of the drop from Eq. (2) and solving the expression obtained for the radius, we find

$$a<\left(\frac{3}{2}\frac{\mu^2(2\mu+3\mu')}{\mu+\mu'}\frac{1}{g\rho(\rho'-\rho)}\right)^{1/3}. \tag{25}$$

Inequality (25) sets a limit to the size of drops to which the results derived in this paper apply.

In the case of bubbles moving in water we have $a\simeq 10^{-2}$ cm, $\nu\simeq 10^{-2}$ cm^2/sec, $\mu'\ll\mu$, and $\rho'\ll\rho$.

Substituting these values in (23) and (24), we find $l\simeq a\simeq 10^{-2}$ cm and T ≈ 0.003 sec, from which it is clear that in the case of bubbles moving in water steady-state diffusion sets in almost instantaneously. However, when $\rho'\simeq\rho$ and $\mu'\gg\mu$ [5], the order of magnitude of the characteristic time T and the length of the nonstationary region l, as calculations from Eqs. (23) and (24) show, may be very great.

When t < T the nonstationary convective diffusion process should follow Eq. (18).

NOTATION

k – mass transfer coefficient; a – radius of drop; D – coefficient of molecular diffusion; ν – kinematic viscosity of medium; μ, μ' – viscosity of medium and substance of drop; $Nu = ka/D$ – Nusselt number; $Re = ua/\nu$ – Reynolds number; $Pr = \nu/D$ – Prandtl number; g – acceleration of gravity; ρ, ρ' – density of medium and substance of drop; c – concentration of distributed substance; v_r, v_θ – radial and angular velocity components; r – radius; θ – angle in spherical coordinates; y – coordinate measured along exterior normal to surface of drop.

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