

# HEAT AND MASS TRANSFER IN INTERACTION OF SPHERICAL DROPS AND GAS BUBBLES WITH A LIQUID FLOW

B. I. BROUNSHTEIN, A. S. ZHELEZNYAK and G. A. FISHBEIN

All-Union Research Institute of Petrochemical Processes, Leningrad, U.S.S.R.

(Received 18 May 1968)

**Аннотация**—Приведен критический обзор работ по массо-и теплопередаче в единичную сферическую каплю, опубликованных после 1965 года. Проведено обобщение данных, полученных авторами статьи. Теоретически рассмотрены предельные случаи массопередачи при лимитирующих сопротивлениях сплошной и диспергированной фаз для  $0 < Re \leq 80$ , а также общий случай соизмеримых фазовых сопротивлений. Результаты теоретических расчетов сопоставлены с имеющимися экспериментальными данными.

## NOMENCLATURE

- $R$ , radius of drop;  
 $c$ , concentration;  
 $\bar{c}_1$ , volume average concentration in drop;  
 $c_{10}$ , initial concentration in drop;  
 $c_{20}$ , concentration in flow core;  
 $\psi$ , distribution coefficient  $= \frac{c_1}{(c_2)_{eq}}$ ;  
 $D$ , diffusion coefficient;  
 $x$ , viscosity ratio of disperse to continuous phase  $= \frac{\mu_1}{\mu_2}$ ;  
 $\nu$ , kinematic viscosity of medium;  
 $\Psi$ , stream function;  
 $u$ , steady-state velocity of drop;  
 $v_0$ , velocity of liquid at drop equator;  
 $k_b$ , individual mass transfer coefficient;  
 $K_b$ , overall mass transfer coefficient;  
 $Re$ , Reynolds number;  
 $Pe$ , Péclet number;  
 $\tau$ , Fourier number;  
 $Sh_b$ , individual Sherwood number;  
 $Sh_{ob}$ , overall Sherwood number;  
 $A$ , degree of saturation;  
 $V$ , volumetric velocity of continuous phase;

$$Pe' = \frac{Pe}{4(1 + X)}$$

## Subscripts

- 1, disperse phase;  
 2, continuous phase;  
 eq, equilibrium concentration.

## INTRODUCTION

THE WORKS on mass transfer to a moving drop published before 1965 were reviewed in [1]. Heat transfer for interaction between a drop and a flow was studied in [2-4]. Recently a number of works on experimental and theoretical study of mass transfer have been published [5-12].

Without affecting the generality of the subject all the subsequent discussion is presented in terms of mass transfer since the most comprehensive experimental information is accumulated in the field of liquid extraction. The shape of drops is assumed close to spherical.

Viscous flow around drops has been studied by Hadamard [13] and Rybczynski [14] for  $Re < 1$  and by Hamielec and Johnson [15] for  $1 < Re \leq 80$ .

In the present paper the limiting cases of mass transfer for controlling resistances in the continuous and disperse phases and the general case of comparable phase resistances for  $0 < Re \leq 80$  have been considered (though experiments were carried out even for larger  $Re$  numbers).

**HYDRODYNAMICS OF FLOW AROUND DROPS**

If the origin of the coordinate system is placed in the centre of the drop and the polar axis is directed to meet the flow then, according to Hadamard and Rybczynski, for small values of  $Re$  ( $Re < 1$ ) the expressions for the stream functions inside and outside the drop in spherical coordinates are of the form

$$\Psi_1 = -\frac{1}{2}v_0\rho^2(1 - \rho^2)\sin^2 \theta \tag{1}$$

$$\Psi_2 = \frac{1}{2}v_0 \left[ \frac{X(1 - \rho^2)}{\rho} - (2 + 3X)\rho(1 - \rho) \right] \sin^2 \theta. \tag{2}$$

According to Hamielec and Johnson [15] at  $1 < Re \leq 80$  the approximate expressions for the stream functions are of the form

$$\Psi_1 = (E_1\rho^2 + E_3\rho^4)\sin^2 \theta + (F_1\rho^2 + F_3\rho^4)\sin^2 \theta \cos \theta \tag{3}$$

$$\Psi_2 = \left( 0.5\rho^2 + \sum_{k=1}^4 \frac{A_k}{\rho_k} \right) \sin^2 \theta + \left( \sum_{k=1}^4 \frac{B_k}{\rho_k} \right) \sin^2 \theta \cos \theta. \tag{4}$$

The values of the coefficients  $E_1, E_3, F_1, F_3, A_1, A_2, A_3, A_4, B_1, B_2, B_3, B_4$  were determined by the authors as functions of  $Re$  and  $X$ .

The liquid velocity components are related to the stream function by the known expressions

$$v_{\rho i} = \frac{1}{\rho^2 \sin \theta} \frac{\partial \Psi_i}{\partial \theta} \tag{5}$$

$$v_{\theta i} = -\frac{1}{\rho \sin \theta} \frac{\partial \Psi_i}{\partial \rho}. \tag{6}$$

**THE LIMITING CASE OF THE CONTROLLING RESISTANCE IN THE CONTINUOUS PHASE**

The distribution of concentrations inside and outside the drop is governed by the convective diffusion equations

$$\frac{\partial c_i}{\partial t} + v_{ri} \frac{\partial c_i}{\partial r} + \frac{v_{\theta i}}{r} \frac{\partial c_i}{\partial \theta} = D_i \left[ \frac{\partial^2 c_i}{\partial r^2} + \frac{2}{r} \frac{\partial c_i}{\partial r} + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left( \sin \theta \frac{\partial c_i}{\partial \theta} \right) \right] \quad (i = 1, 2). \tag{7}$$

Since the diffusion coefficient for liquids is rather small ( $D = 10^{-5}$  cm<sup>2</sup>/s) then, for the time of liquid flow around the drop, the diffusion front will move by a distance much less than the drop radius. Therefore it may be considered that a thin diffusional boundary layer exists on the outside drop surface and the mass transfer process is a steady-state one.

In the approximation of the diffusional boundary layer the last two terms in expression (7) for the Laplace operator in spherical coordinates [16] may be neglected and the equation of convective diffusion for a steady-state case assumes the form

$$v_{r2} \frac{\partial c_2}{\partial r} + \frac{v_{\theta 2}}{r} \frac{\partial c_2}{\partial \theta} = D_2 \frac{\partial^2 c_2}{\partial r^2}. \tag{8}$$

For the case of all resistance in the continuous phase equation (8) is solved with the boundary conditions

$$c_2(r, \theta) \Big|_{r \rightarrow \infty} = c_{20} \quad c_2(r, \theta) \Big|_{r=R} = \frac{c_1}{\Psi} \tag{9}$$

where  $c_1$  is the constant volumetric concentration inside the drop.

Under the assumption that the thickness of the diffusional layer is much smaller than the drop radius, equation (8) in Prandtl and Mizes variables is transformed to

$$\frac{\partial c_2}{\partial \theta} = D_2 R^{-1} (v_{\theta})_{\rho=1} \sin^2 \theta \frac{\partial^2 c_2}{\partial \Psi^2}. \tag{10}$$

The expressions for the stream functions, linearized with respect to  $\rho^1 = \rho - 1$ , are of one and

the same form

$$\Psi_i = -v_{\text{eq}} \rho^1 \sin^2 \theta \quad (11)$$

where  $v_{\text{eq}}$  is absolute value of the liquid velocity at the drop equator.

For  $Re < 1$ ,  $v_{\text{eq}} = v_0$ . For  $1 < Re \leq 80$  the surface velocity of liquid at the equator is approximately determined by Baird and Hamielec [17] as

$$v_{\text{eq}} = \frac{10 + 2A_1}{8 + 3X}. \quad (12)$$

The boundary conditions (9) in terms of the variables  $\theta$ ,  $\Psi$  are transformed into

$$c_2(\theta, \Psi) \Big|_{\Psi \rightarrow \infty} = c_{20} \quad c_2(\theta, \Psi) = \Big|_{\Psi \rightarrow 0} \frac{c_1}{\psi} \quad (13)$$

For small Reynolds numbers equations (10) and (11) with boundary conditions (13) were solved by Levich [16]. Levich's solution is

$$Sh_2 = \frac{2}{\sqrt{[3\pi(1 + X)]}} \sqrt{(Pe_2)}. \quad (14)$$

Equation (14) may be represented by

$$Sh_2 = 4 \sqrt{\left( \frac{Rv_{\text{eq}}}{3\pi D_2} \right)}. \quad (15)$$

Since the expression for the stream function (11) is of the same form both for small and large  $Re$ , then equation (15) is also valid for the range of Reynolds numbers  $1 < Re \leq 80$ . In the latter case the velocity of flow at the drop equator is determined by equation (12). By substituting the value of  $v_{\text{eq}}$  from (12) into equation (15)

$$Sh_2 = 1.13 \sqrt{\left( \frac{20 + 4A_1}{24 + 9X} \right)} \sqrt{(Pe_2)}. \quad (16)$$

Equation (16) was obtained earlier by Baird and Hamielec [17] in the solution of the external problem within the framework of the theory of diffusional boundary layers by a somewhat cumbersome method.

#### THE LIMITING CASE OF THE CONTROLLING RESISTANCE IN THE DISPERSE PHASE

In the case of the total resistance being located in the disperse phase the distribution of the concentrations of the extracted component is determined by the solution of the equations of convective diffusion (7) with the boundary conditions

$$c_1(r, \theta, t) \Big|_{r=R} = \psi c_{20} \quad c_1(r, \theta, t) \Big|_{r=0} \neq \infty \quad (17)$$

and initial condition

$$c_1(r, \theta, t) \Big|_{t=0} = c_{10}. \quad (18)$$

For the first time the equation of convective diffusion for  $Re < 1$  was solved by Kronig and Brink (18) on the assumption that the concentration of the extracted component along a streamline is constant. The authors substantiated the adopted assumptions by an estimation according to which the time of circulation along the streamline is much shorter than the time of relaxation of drop saturation by molecular diffusion. With the above assumptions the authors obtained the convection diffusion equation in the form

$$\frac{\partial}{\partial x} \left[ P(x) \frac{\partial c_1(x, t)}{\partial x} \right] = \frac{R^2}{16D_1} q(x) \frac{\partial c_1(x, t)}{\partial t} \quad (19)$$

where the curvilinear coordinate  $x = 4\rho^2(1 - \rho^2) \sin^2 \theta$  coincides with the expressions for the stream function except for the constant factor. The coefficients  $P(x)$  and  $q(x)$  are functions of total elliptic integrals.

Boundary conditions (17) in terms of variables  $x$ ,  $t$  assume the form

$$c_1(x, t) \Big|_{x=0} = \psi c_{20} \quad c_1(x, t) \Big|_{x=1} \neq \infty \quad (20)$$

The solution of Kronig and Brink for the degree of saturation is of the form

$$A(\tau) = 1 - \frac{3}{8} \sum_{n=1}^{\infty} B_n \exp(-16 \lambda_n \tau). \quad (21)$$

Kronig and Brink calculated seven values of the coefficients  $B_n$  and  $\lambda_n$  by the Ritz method.

Later on equation (19) with boundary (20) and initial conditions (18) was computed on the electronic digital computer M-20 for a wide range of Fourier numbers from  $\tau = 10^{-4}$ . The results of computations for  $A$  are presented in Table 1 of the Appendix to [1]. Table 4-2 in [1] represents the values of  $A$  calculated by equation (21) for  $n = 7$ . The comparison of calculations for  $\tau > 10^{-3}$  shows satisfactory agreement. However at  $\tau = 10^{-4}$  the values of  $A$  calculated more accurately on the electronic digital computer were 2.6 times smaller than those given by equation (21).

In [7, 8] the solution of [18] is criticized in connection with the fact that at large  $Pe$  numbers the basic assumptions of Kronig and Brink do not hold in a thin surface layer of a drop. At the same time, according to the authors' of [7, 8] the main resistance to mass transfer in the drop is concentrated in a thin diffusional boundary layer of the drop.

In this connection, to give more comprehensive explanation of the Kronig and Brink model, application in [19] of the solution of the convective diffusion equation (7) is carried out under conditions (17) and (18) for the case when the  $Pe$  number is large enough. The solution is performed by the Bubnov-Galerkin method [20]. In the drop the concentration distribution is found in the form of the series

$$c(\tau, r, \theta) = \sum_{i=1}^{\infty} a_i(\tau) \varphi_i(r, \theta). \quad (22)$$

The succession of approximating functions was chosen to satisfy the boundary conditions (17) and the steady-state convective diffusion equation in the first approximation. According to the Bubnov-Galerkin method the boundary-value problem is reduced to the system of ordinary differential equations in terms of the coefficients  $a_i(\tau)$

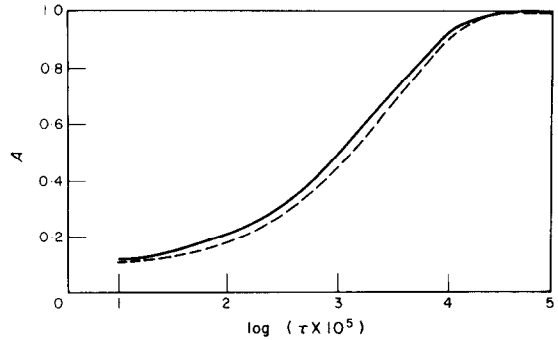


FIG. 1. Degree of saturation  $A$  against Fourier number. Solid curve, Kronig and Brink's solution [18]; dotted line, Brounshtein and Fishbein's solution [19].

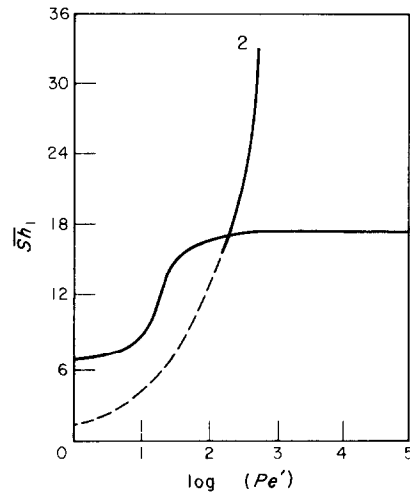


FIG. 2.  $Sh_1$  number as function of  $Pe'$  number at  $\tau > 0.5$ . 1, Johns and Beckman's solution [11]; 2, Levich, *et al.* [7] and Ruckenstein's [12] solutions for large  $Pe_1$ .

$$\frac{da_i}{d\tau} + \sum_{k=1}^n \lambda_{ik} a_k(\tau) = 0 \quad (i = 1, 2, \dots, n) \quad (23)$$

with initial conditions

$$a_i(0) = \beta_i. \quad (24)$$

For the degree of saturation the following expression is found

$$A(\tau) = 1 - \frac{3}{4\pi} \sum_{i=1}^n a_i(\tau) \beta_i. \quad (25)$$

The series (25) converges somewhat quicker than the series (21). The numerical calculations have been carried out for  $n = 5$ . Figure 1 represents the calculations by equations (21) and (25).

The solution of equation (7) with conditions (17) and (18) for various Péclet numbers was obtained by Johnson and Beckman [11] on an electronic digital computer. The computational results for large  $\tau$  ( $\tau > 0.15$ ) are presented in Fig. 2 (Curve 1). As follows from the graph the Sherwood number is practically independent of  $Pe'$  at  $Pe' > 100$ . The asymptotic value of  $Sh$  obtained by Johns and Beckman (i.e. the value of  $Sh$  at large  $Pe$  and  $\tau$ ) is 17.9. The same value of  $Sh$  is asymptotic for the Kronig and Brink solution. Thus Kronig and Brink's solution may be used for the calculation of heat and mass transfer into a spherical drop for the total resistance present in the disperse phase if  $\tau > 0.1$   $Pe' > 100$ . At  $\tau < 0.1$  the solution by Kronig

responding to the values of Péclet numbers from  $Pe' = 0$  (pure molecular diffusion) to  $Pe' = 80$  converge into a single curve described, according to Johns and Beckmann, by the equation

$$\bar{Sh}_1 = -\frac{2}{3\tau} \ln \frac{\bar{c}_1}{c_{10}}$$

where

$$\frac{\bar{c}_1}{c_{10}} = 1 - 3.38 \sqrt{\tau}. \tag{26}$$

The dotted line which corresponds to the computer solution by Kronig and Brink satisfies equation (26) for  $\tau \leq 10^{-4}$ .

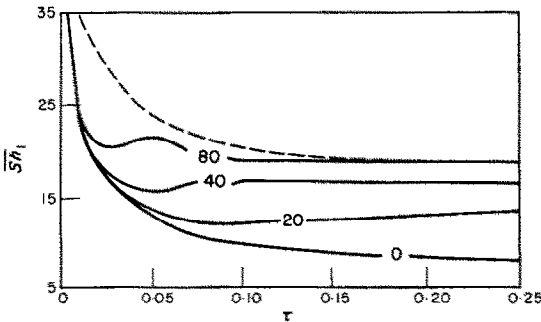


FIG. 3.  $Sh_1$  number as a function of Fourier number for various  $Pe_1$ . Solid curves, Johns and Beckman's solution; dotted line, solution of Kronig and Brink's equations on electronic digital computer.

may already be applied at  $Pe' \gg 100$ . In Fig. 3 the results of calculations by Johns and Beckman show the dependence of the Sherwood number on the Fourier number at various Péclet numbers. The dashed curve corresponds to the accurate solution of equation (19) on an electronic digital computer at the Computation Centre of Leningrad University.

For  $\tau < 0.015$  the different curves corre-

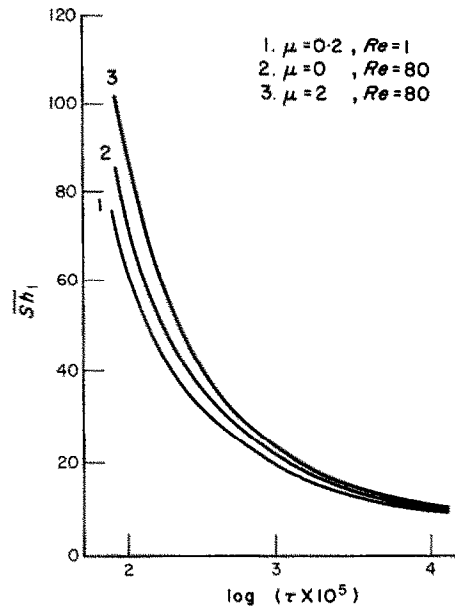


FIG. 4.  $Sh_1$  number as a function of  $\tau$  at  $X = 0, 2$ . 1:  $X = 0, 2$ ;  $Re < 1$ ; 2:  $X = 0$ ;  $Re = 80$ ; 3:  $X = 2$ ;  $Re = 80$ .

For Reynolds numbers much greater than unity ( $1 < Re \leq 80$ ) equation (7) with conditions (17) and (18) was solved with the use of the expression for the stream function (3) in [21]. The solution for large  $Pe$  was carried out by the Bubnov-Galerkin method. The calculation results are plotted in Fig. 4 for  $X = 0$  and 2 and in Fig. 5, for  $X = 10$ . As follows from the graphs for  $X \leq 2$  and  $\tau > 10^{-3}$  the Sherwood

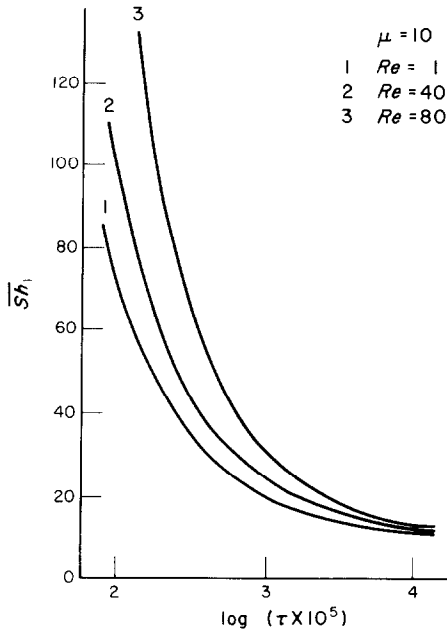


FIG. 5.  $Sh_1$  number as a function of  $\tau$  at  $X = 10$ . 1:  $Re < 1$ ; 2:  $Re = 40$ ; 3:  $Re = 80$ .

number slightly depends on  $Re$ . However at  $X = 10$  the value of  $Sh_1$  for  $Re = 80$  and  $\tau = 10^{-3}$  is rather greater than for  $Re < 1$ .

**COMPARABLE PHASE RESISTANCES**

The concentration change in the continuous phase in counter-flow for the case of comparable phase resistances is considered in [6], where equation (19) is solved approximately for boundary conditions corresponding to the averaged material balance in counter-flow of the extracted component. The numerical values of  $A$  as a function of the Fourier number and parameters  $\alpha = V_1\psi/V_2$ ;  $\beta = D_1\psi/2K_2R$  are included in [1]\*. The time average overall Sherwood number for the disperse phase at  $\alpha = 0$  is related with the degree of extraction  $A$  by the expression

$$\overline{Sh}_{01} = -\frac{2}{3\tau} \ln(1 - A). \tag{27}$$

\* In [1], Appendix 1, pp. 299–306; text, p. 115  $\beta = D_1\psi/k_2R$  should be substituted by  $\beta = D_1\psi/2k_2R$ .

The results of calculations for  $\alpha = 0$  are plotted in Figs. 6–8. As follows from the graphs in Figs. 6 and 7 the resistance of the disperse phase may be considered controlling at  $\beta < 10^{-3}$  (the limiting case of the controlling resistance in the disperse phase is  $\psi \rightarrow 0$  and  $\beta \rightarrow 0$ , respectively). The value  $\beta \rightarrow \infty$  ( $\psi \rightarrow \infty$ ) corresponds to the limiting case of the controlling resistance in the continuous phase. As follows from the data of Fig. 8 the continuous phase resistance is controlling up to  $\tau \approx 1$  at  $\beta > 1$ . Thus the range of  $\beta$  values  $10^{-3} < \beta < 1$  corresponds to the intermediate case of comparable phase resistances.

As is known for the steady-state conditions of mass transfer the additivity equations [1] may be applied

$$\frac{1}{K_1} = \frac{1}{k_1} + \frac{\psi}{k_2}; \quad \frac{1}{K_2} = \frac{1}{k_2} + \frac{1}{\psi k_1}. \tag{28}$$

Since for comparable phase resistances the mass transfer regime is non-steady only at  $\tau < 0.1$  (Fig. 7), then at  $\tau > 0.1$  the additivity equation (28) may be used for the calculation of the overall mass transfer coefficient. At  $\tau < 0.1$  the correction for the additivity formula may be calculated by the equation

$$\frac{K_1}{(\overline{K}_1)_{ad}} = Sh_{01} \left( \beta + \frac{1}{Sh_1} \right) \tag{29}$$

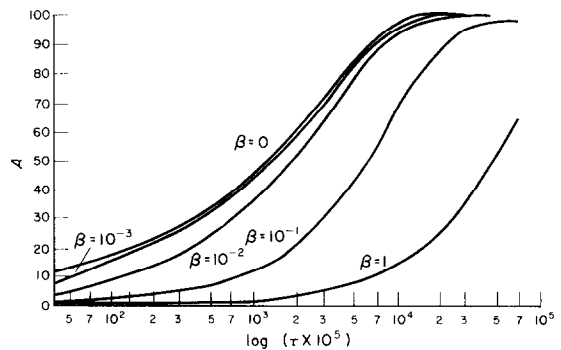


FIG. 6. Degree of saturation  $A$  against  $\tau$  at  $\alpha = 0$  and  $\beta = 0, 10^{-3}, 10^{-2}, 10^{-1}, 1$ .

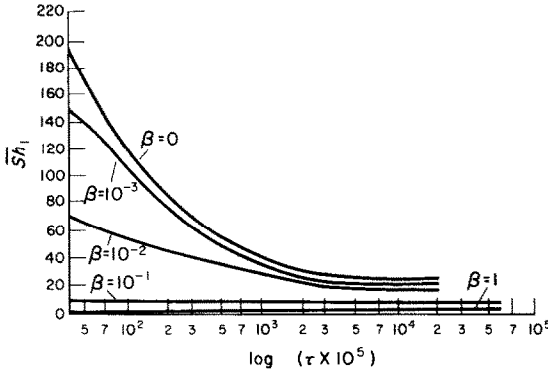


FIG. 7.  $Sh_1$  as a function of  $A$  at  $\tau = 0$  and  $\beta = 0, 10^{-3}, 10^{-2}, 10^{-1}, 1$ .

where  $(\bar{K}_1)_{ad}$  is the time-average mass transfer coefficient predicted by the additivity equation (28).

#### THE SOLUTION OF THE INTERNAL PROBLEM BY USING THE APPROXIMATION OF A THIN DIFFUSION BOUNDARY LAYER

In recently published works [7, 8, 12] the case of comparable phase resistances for  $Re < 1$  is considered on the assumption of the existence of a thin diffusional boundary layer both on the external and internal sides of the drop surface at large  $Pe$  numbers.

Levich [16] arrives at the existence of a thin diffusional boundary layer at large  $Pe$  after considering the equation of convective diffusion, the dimensionless form of which is

$$(\vec{\nabla}\vec{\nabla})C = \frac{1}{Pe} \Delta C. \quad (30)$$

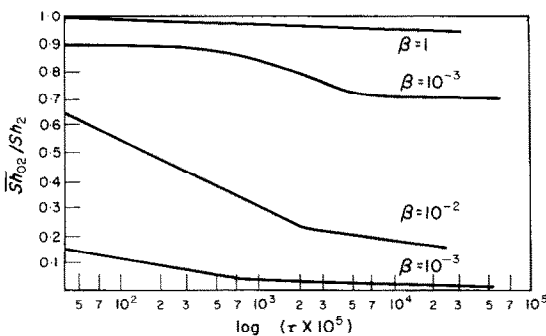


FIG. 8.  $Sh_2$  as a function of  $\tau$  at  $\alpha = 0$  and  $\beta = 10^{-3}, 10^{-2}, 10^{-1}, 1$ .

At large  $Pe$  the left hand side of the equation in the flow core is zero. Hence Levich concludes that always  $\nabla C = 0$ , i.e.  $C = \text{const}$ . However  $(\vec{\nabla}\vec{\nabla})C = 0$  not only at  $\nabla C = 0$  but also in the case when the concentration gradient is normal to the streamlines. This is the case found by Kronig and Brink for mass transfer to the drop with the resistance present only in the disperse phase.

Since Johns and Beckmann [11] obtained an exact solution of the convective diffusion equation (7) for the case of the controlling resistance in the disperse phase, this solution may be compared to those by Levich *et al.* [7] and Ruckenstein [12]. As the expressions for the stream function inside and outside the drop in the approximation of a thin diffusional boundary layer are of the same form (11) as the convective diffusion equation (10) and boundary conditions (13), then equation (14) in the form

$$Sh_1 = \frac{2}{\sqrt{[3\pi(1+X)]}} \sqrt{(Pe_1)} \quad (31)$$

is the solution. Equation (31) may be obtained from general equations for comparable phase resistances cited in [7] and [12] as the limiting case. The comparison of calculations by equation (31) with the exact solution by Johns and Beckman for  $\tau > 0.15$  (when the mass transfer process is steady) is presented in Fig. 2 (curve 2). For  $\tau < 0.15$  the process of mass transfer is nonsteady according to the exact calculation, while  $Sh_1$  determined by equation (31) does not depend on  $\tau$ . According to [8] the process is unsteady only at the very initial moment of formation of the diffusional boundary layer. The relaxation time for the boundary layer is small and equal to the ratio of drop diameter to its rising velocity. The Fourier number  $\tau = 4/Pe$  corresponds to the above condition. The authors of [7, 8] consider that the solution obtained is valid for the Fourier numbers  $\tau, \ll \tau \ll \tau_d$ , where  $\tau_d$  is the Fourier number corresponding to the time in which the total amount of the substance dissolved in a drop decreases  $e$  times the case of molecular diffusion. In reality

equation (31) holds on the assumption of the existence of a diffusional boundary layer for a quasi-stationary mass transfer mechanism, i.e. in the case when for the time of relaxation of the boundary layer the relative driving force  $(C_{ec} - C_1)/(C_{eq} - C_{01})$  changes slightly. Assume this condition. The concentration change inside the drop is found from the solution of the differential equation

$$\frac{dc_1}{dt} = \frac{3}{R} k_1 (c_{eq} - c_1)$$

at the initial condition  $c_1 = c_{01}$  at  $t = 0$ . The solution is of the form

$$\frac{c_{eq} - c_1}{c_{eq} - c_{01}} = \exp - \frac{3k_1 t}{R}$$

Consequently, the time for the relative driving force to decrease  $e$  times  $t_e = R/3k_1$  or  $\tau_e = 2/3Sh_1$ .

By substituting into expression for  $\tau_e$  the value of  $Sh_1$ , from (31) gives

$$\tau_e = \frac{\sqrt{[3\pi(1 + X)]}}{3\sqrt{(Pe_1)}}$$

The process is nonsteady at  $\tau_e \gg \tau_r$ . Since  $\tau_r \approx 4/Pe$  then the quasi-stationary condition may be written in the form

$$\sqrt{(Pe_1)} \gg \frac{12}{\sqrt{[3\pi(1 + X)]}}$$

which always holds at large  $Pe$ .

Thus, on the assumption of a diffusional boundary layer inside the drop (if such a layer existed) equation (31) would hold at large  $Pe$  for any  $\tau > 4/Pe$ , rather than for a finite range of numbers  $\tau_r < \tau < \tau_d$ , as considered by the authors of [7, 8].

#### COMPARISON WITH EXPERIMENTAL DATA

As was pointed out in the Introduction, a review of the works on mass transfer to moving drops published before 1965 (including experimental ones) is given in monograph [1]. Recently a number of new works have been

published which include the comparison of the predicted data with experimental ones.

Zheleznyak and Brounshtein [5, 22, 23] after extraction from single drops in five systems with the ratio of the viscosities of disperse and solid phases  $0 < X \leq 2$  and Reynolds numbers  $15 \leq Re \leq 650$ , established that for drops  $d = 1-3$  mm at  $Re \leq 200$  the deviation between the experimental data and those predicted by Kronig and Brink do not exceed 10-12 per cent. At  $Re > 200$  the deviation is much greater. Thus for example, at  $200 < Re < 500$  the relative deviation (predicted to experimental mass transfer coefficient ratio) ranges from 0.85 to 0.45. Similar results were obtained when treating the experimental data on extraction of propionic acid from aqueous solution with benzene by Smirnov and Kuznetsov [24, 25]. The results of the comparison of experimental data of [22, 23] with the predicted ones by Kronig and Brink are presented in Fig. 9. Comprehensive experimental material was obtained for the extraction of acetic acid from aqueous solutions with different concentrations of ethyl acetate [26]. These experiments are in good agreement with Kronig and Brink's calculations.

Skelland and Wellek [9] carried out experiments with four binary systems with resistance in the disperse phase only and Reynolds numbers  $37 \leq Re \leq 546$ . The ratio between viscosities of disperse and continuous phases for three systems was  $X \approx 2$  and for one  $X \approx 12$ . All the experimental data have been correlated for all the systems within the above range of  $Re$  numbers. The experimental data have been compared with the data predicted by Kronig and Brink. In this work the scatter of the experimental data is very large (the root mean square deviation is 46 per cent). As was shown earlier (Fig. 4) the equations by Kronig and Brink may be used for calculating the rate of mass transfer in the range  $1 < Re < 80$  if  $X \leq 2$ . Zheleznyak and Brounshtein have found that at  $X \leq 2$  the experimental data are in satisfactory agreement with those predicted by Kronig and Brink



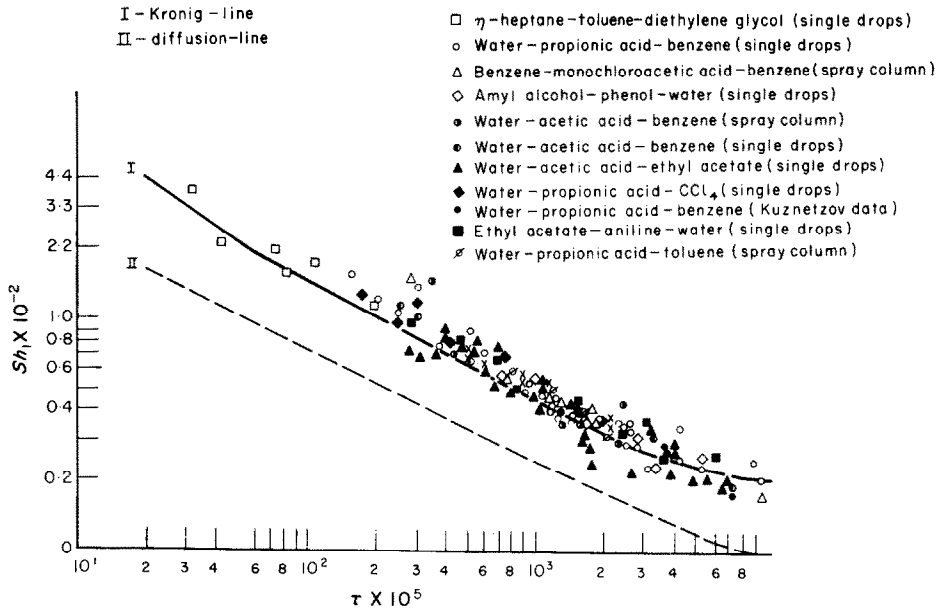


FIG. 9. Comparison of experimental data for  $Sh_1$  with those predicted by equations of Kronig and Brink [18], I and Newman [1] II.

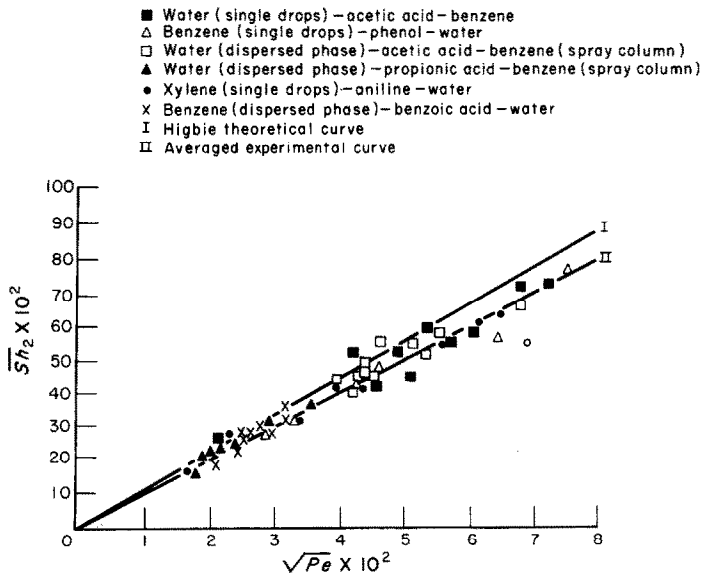


FIG. 10. Comparison of experimental relationship  $Sh_2$  against  $\sqrt{Pe_2}$  with calculations by the Higbie equation.

up to  $Re \approx 200$ . The large discrepancy between experimental data and those predicted by Kronig and Brink is perhaps caused by the fact that in Skelland and Wellek's paper the experimental data are correlated for rather wide ranges of  $Re$  numbers and viscosity ratios.

U Chzhi-Tztuan *et al.* [10], while studying mass transfer to single drops in *n*-butanol and isobutanol, have found that the transfer process is in a good agreement with the data of Kronig and Brink at  $Re = 50-60$ . A number of other works on heat transfer are known which show satisfactory agreement between experimental data and those predicted by Kronig and Brink [2-4]. Thus, according to Calderbank and Korchinski [2] Kronig and Brink's formula is valid for  $Re \leq 200$ .

Kronig and Brink's model has been used in developing the engineering methods for the calculation of spray and plate columns and yielded positive results [27].

For the case of controlling resistance in the continuous phase six systems have been checked [22, 23]. The experimental data showed fine agreement with the values calculated by the Higbie equation,  $Sh_2 = 1.13 \sqrt{(Pe_2)}$ . The comparison of the experimental data with the theoretical curve is presented in Fig. 10. The averaged experimental curve II, Fig. 10 satisfies the equation

$$Sh_2 = 1.02 \sqrt{(Pe_2)} \quad (32)$$

i.e. the deviation of the values predicted by the Higbie equation from the experimental data is on the average 10 per cent.

For the disperse to solid phase ratio  $X < 1$  and  $Re \approx 80$  equation (16) yields results close to the Higbie equation and provides satisfactory agreement with the data of the experiment.

For the case of comparable phase resistances the experimental test of the numerical results was carried out within the range of the parameters  $\alpha = 0.0-0.03$ ;  $10^{-4} < \beta < 1.2$  and Fourier numbers  $\tau \leq 0.016$ . Within the above range the experimental data show satisfactory agreement with the data of calculations [6].

In conclusion the authors express their acknowledgement to V. Ya. Rivkind and B. A. Samokish under whose direction the computations have been carried out on the electronic digital computer M-20 at the Leningrad State University.

#### CONCLUSION

Equations are presented for computation of the rate of mass transfer to a spherical drop for the limiting cases of the resistances present only in the continuous and disperse phases as well as for the general case of comparable phase resistances within the range of Reynolds numbers  $0 < Re \leq 80$ .

The calculation of the mass transfer rate for the controlling resistance in the disperse phase using the approximation of a thin diffusional boundary layer is shown to be incorrect.

It is also shown that within the range  $0 < Re \leq 200$  and the ratio between the viscosities of disperse and continuous phases  $X \leq 2$  the experimental data on the rate of mass and heat transfer for the controlling resistance in the disperse phase and for the case of comparable phase resistances are in satisfactory agreement with the calculated values.

#### REFERENCES

1. B. I. BROUNSHTEIN and A. S. ZHELEZNYAK, *Physico-Chemical Fundamentals of Liquid Extraction (Fiziko-khimicheskie osnovy zhidkostnoi ekstraktsii)*. Izd. Khimiya, Moscow-Leningrad (1966).
2. P. H. CALDERBANK and J. S. O. KORCHINSKI, Circulation in liquid drops, *Chem. Engng Sci.* **6**, 65 (1956).
3. R. V. McDOWELL and J. E. MYERS, Mechanism of heat transfer to liquid drops, *A.I.Ch.E. Jl* **2**, 384 (1956).
4. L. CARWIN and B. D. SMITH, Liquid-liquid spray-tower operation in heat transfer, *Chem. Engng Prog.* **49**, 591 (1953).
5. A. S. ZHELEZNYAK and B. I. BROUNSHTEIN, End effects for disperse phase inlet, *Zh. Prikl. Khim.* **38**, 435 (1965).
6. B. I. BROUNSHTEIN, I. P. GITMAN and A. S. ZHELEZNYAK, Mass transfer to spherical drops, *Dokl. Akad. Nauk SSSR*, **162**, 1336 (1965).
7. V. G. LEVICH, V. S. KRYLOV and V. P. VOROTILIN, Theory of extraction out of a falling drop, *Dokl. Akad. Nauk SSSR* **160**, 1358 (1965).
8. V. P. VOROTILIN, V. S. KRYLOV and V. G. LEVICH, Theory of unsteady diffusion from a moving drop, *Dokl. Akad. Nauk SSSR* **161**, 648 (1965).

9. A. H. P. SKELLAND and R. H. WELLEK, Resistance to mass transfer inside droplets, *A.I.Ch.E. Jl* **10**, 491 (1964).
10. U. CHZHI-TZTUAN *et al.*, Rate of mass transfer to single drops with internal circulation, *Huagong Xuebao* **4**, 237-249 (1965).
11. L. JOHNS and R. BECKMAN, Mechanism of dispersed-phase mass transfer in viscous single-drop extraction-systems, *A.I.Ch.E. Jl* **12**, 10 (1966).
12. E. RUCKENSTEIN, Mass transfer between a single drop and a continuous phase, *Int. J. Heat Mass Transfer* **10**, 1785 (1967).
13. J. HADAMARD, Mouvement permanent lent d'une sphere liquide et visqueuse dans un liquide visqueux, *Compt. rend* **152**, 1735 (1911).
14. W. RYBCZYNSKI, Über die fortschreitende Bewegung einer flüssigen Kugel in einem zähen Medium, *Bull. Acad. Cracovia* **A40** (1911).
15. A. E. HAMIELEC and A. I. JOHNSON, Viscous flow around fluid spheres at intermediate Reynolds numbers, *Can. J. Chem. Engng* **40**(2), 41 (1962).
16. V. G. LEVICH, *Physico-chemical Hydrodynamics (Fiziko-khimicheskaya gidrodinamika)*. Fizmatgiz, Moscow (1959).
17. M. H. I. BAIRD and A. E. HAMIELEC, Forced convection transfer around spheres at intermediate Reynolds numbers, *Can. J. Chem. Engng* **40**(3), 119 (1962).
18. R. KRONIG and I. BRINK, On the theory of extraction from falling droplets, *Appl. Sci. Res.* **A2**, 142 (1950).
19. B. I. BROUNSHTEIN and G. A. FISHBEIN, Unsteady mass and heat transfer to a spherical drop with consideration of internal convection. The questions of the evaporation burning and gas dynamics of the disperse, *Trudy Odesskogo Univer.* **171** (1968).
20. S. G. MIKHLIN, *Numerical Application of Variational Methods (Chislennaya realizatsiya variatsionnykh metodov)*. Izd. Nauka, Moscow (1966).
21. B. I. BROUNSHTEIN and G. A. FISHBEIN, Unsteady mass transfer to a spherical drop, *Teplo-i massoperenos* **2**, 351, Minsk (1968).
22. A. S. ZHELEZNYAK and B. I. BROUNSHTEIN, Mass transfer in single drop extraction, *Zh. Prikl. Khim.* **36**, 2437 (1963).
23. A. S. ZHELEZNYAK, Study of mass transfer mechanism in extraction in perforated plate columns, Cand. Thesis, MITKhT (1965).
24. F. A. KUZNETSOV and N. N. SMIRNOV, Extraction in spray columns, *Trudy LTI* **60**, 190 (1960).
25. F. A. KUZNETSOV, Study of mass transfer in extraction in spray columns, Cand. Thesis, LTI Lensoveta (1961).
26. N. I. KADENSKAYA, A. S. Zheleznyak and B. I. BROUNSHTEIN, Mass transfer in extraction of acetic acid from single ethylacetate drops, *Zh. Prikl. Khim.* **38**, 1156 (65) (65).
27. A. S. ZHELEZNYAK and B. I. BROUNSHTEIN, Study of mass transfer mechanism in perforated plate columns, *Zh. Prikl. Khim.* **40**, 584 (1967).

#### TRANSPORT DE CHALEUR ET DE MASSE POUR L'INTERACTION DE PARTICULES SPHERIQUES ET DE BULLES DE GAZ AVEC UN ECOULEMENT LIQUIDE

**Résumé**—L'article présente un compte-rendu des travaux publiés depuis 1965 sur le transport de chaleur et de masse à une goutte sphérique unique. Les données de l'article actuel sont corrélées. Les cas restreints du transport de masse pour les résistances maximales des phases continue et dispersée à  $0 < Re \leq 80$  et le cas général des résistances de phase commensurables sont étudiés théoriquement. Les résultats des calculs théoriques sont comparés avec les résultats expérimentaux disponibles.

#### WÄRME- UND STOFFÜBERGANG BEI DER WECHSELWIRKUNG VON SPHÄRISCHEN TROPFEN UND GASBLASEN MIT EINEN FLÜSSIGKEITSSTRÖMUNG

**Zusammenfassung**—Der Bericht bietet einen Überblick über die nach 1965 veröffentlichten Arbeiten in der Wärme- und Stoffübertragung an einzelne sphärische Tropfen. Die Messergebnisse in diesem Artikel wurden in Korrelation gebracht. Die begrenzten Fälle des Stofftransportes für die maximalen Widerstände in feinverteilter und in dispersem Zustand bei  $0 < Re \leq 80$  und der allgemeine Fall entsprechender Phasenwiderstände wurden theoretisch untersucht. Die Ergebnisse der theoretischen Kalkulationen wurden mit den verfügbaren experimentellen Daten verglichen.

**Abstract**—The paper presents a review of works on heat and mass transfer to a single spherical drop published after 1965. The data of the present are correlated. The limiting cases of mass transfer for the resistances present in the continuous and disperse phases only at  $0 \leq Re \leq 80$  and the general case of comparable phase resistances were studied theoretically. The results of theoretical calculations are compared with the experimental data available.