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Abstract

Mass transfer to spherical drops or bubbles rising steadily through a liquid at high Re was calculated by using a boundary layer approach for the fluid flow. The result can be written as

$$Nu = \sqrt{\frac{3}{4\pi}}\, I(Re,\mu,\rho) Pe^{\frac{1}{2}}$$

where $I(Re,\mu,\rho)$ is a given function of Re and two physical properties, namely, the viscosity and the density of the system.

This result reduces to the Boussinesq potential solution

$$Nu = 1.128\ Pe^{\frac{1}{2}} ,$$

as $Re \to \infty$.

Comparison of the theoretical result to the experimentally observed behavior of two systems shows satisfactory agreement.

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1. Introduction

Mass transfer to drops or bubbles moving steadily through a liquid is of importance to many chemical engineering operations. However, theoretical calculations have, so far, only been performed for certain limiting cases. Levich[8] studied the case in Stokes flow regime but at high Pe number. Bowman <u>et al</u>[3] extended the calculation to cover the complete range of Pe number in Stokes flow. For ideal fluids where the Pe number is infinite, the exact potential solution was obtained by Boussinesq[1].

In many practical applications, the process occurs at moderately high Pe where these theories fail to describe the system successfully. Consequently, the design of processes involving ascent of bubbles in stagnant fluids depends heavily on experimental correlations.

In this paper the boundary layer approach suggested by Chao[4] and Moore[11] was used to solve the fluid mechanics for steady flow past a drop or bubble at high Re. The resulting velocity was then used to solve the convective diffusion equation. The exact solution thus obtained was compared with available experimental results[5].

2. Fluid Mechanics

At high Re, a boundary layer approach was first suggested by Levich[7]. His method was applied by Moore to the case of gas bubble rising steadily through a liquid. This procedure will be used here for the case of rising liquid drops.

The velocity and pressure are written as perturbations from the potential solution,

$$\underline{v} = \underline{\overline{v}} + \underline{v}^1, \tag{1}$$

and $$p = \overline{p} + p^1 ,\tag{2}$$

where $\overline{\underline{v}}$, $\overline{p}$ are the potential solution and $\underline{v}^1$ and p^1 are the perturbed quantitites.

The potential solution given by Hill[10] are

for $r \leq R$ (inside the drop)

$$\overline{v}_{\theta_i} = -\frac{3}{2} v_\infty \left(1 - \frac{2r^2}{R^2}\right) \sin\theta ,\tag{3}$$

$$\overline{v}_{r_i} = \frac{3}{2} v_\infty \left(1 - \frac{r^2}{R^2}\right) \cos\theta ,\tag{4}$$

and, for $r \geq R$ (outside the drop)

$$\overline{v}_{\theta_o} = v_\infty \left(1 + \frac{1}{2}\frac{R^3}{r^3}\right) \sin\theta ,\tag{5}$$

$$\overline{v}_{r_o} = -v_\infty \left(1 - \frac{R^2}{r^3}\right) \cos\theta .\tag{6}$$

The Navier-Stokes equation of motion and the continuity equation in spherical coordinates are

$$\frac{v_\theta}{r}\frac{\partial v_\theta}{\partial\theta} + v_r \frac{\partial v_\theta}{\partial r} + \frac{v_\theta v_r}{r} = -\frac{1}{\rho r}\frac{\partial p}{\partial\theta} + \nu\left(\nabla^2 v_\theta - \frac{v_\theta}{r^2 \sin^2\theta} + \frac{2}{r^2}\frac{\partial v_r}{\partial\theta}\right) ,\tag{7}$$

$$\frac{v_\theta}{r}\frac{\partial v_r}{\partial\theta} + v_r \frac{\partial v_r}{\partial r} - \frac{v_\theta^2}{r} = -\frac{1}{\rho}\frac{\partial p}{\partial r} + \nu\left(\nabla^2 v_r - \frac{2v_r}{r} - \frac{2\cot\theta}{r^2} v_\theta - \frac{2}{r^2}\frac{\partial v_\theta}{\partial\theta}\right) ,\tag{8}$$

and $$\frac{\partial v_r}{\partial r} + \frac{2v_r}{r} + \frac{1}{r\sin\theta}\frac{\partial}{\partial\theta}(v_\theta \sin\theta) = 0 ,\tag{9}$$

where $$\nabla^2 = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial}{\partial r}\right) + \frac{1}{r^2 \sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta \frac{\partial}{\partial\theta}\right).\tag{10}$$

The boundary conditions are:

1. Far from the interface, potential solution is valid,

   a. as $r\to\infty$, $$v^1_{\theta_o} = v^1_{r_o} = 0 ,\tag{11}$$

   b. at $r=0$, $$v^1_{\theta_i} = v^1_{r_i} = 0 .\tag{12}$$

2. At the interface, there is no radial velocity and the tangential

velocity is continuous,

a. at r=R, $$v^1_{\theta_o} = v^1_{\theta_i} ,\qquad (13)$$

$$v^1_{r_o} = v^1_{r_i} = 0. \qquad (14)$$

At the interface, the tangential shear stress is continuous,

b. at r=R, $$\tau_{r\theta_o} = \tau_{r\theta_i} ; \qquad (15)$$

$$\text{or } \mu_i \left[\frac{1}{r}\frac{\partial}{\partial\theta} v_{r_i} + r\frac{\partial}{\partial r}\left(\frac{v_{\theta_i}}{r}\right)\right] = \mu_o\left[\frac{1}{r}\frac{\partial}{\partial\theta}(v_{r_o}) + r\frac{\partial}{\partial r}\left(\frac{v_{\theta_o}}{r}\right)\right] . \qquad (16)$$

This can be simplified by applying equations (1) and (3-6) to read

$$\mu_o \frac{\partial v^1_{\theta_o}}{\partial r} - \mu_i \frac{\partial v^1_{\theta_i}}{\partial r} = 3\frac{v_\infty}{R} \sin\theta\,(\mu_o + \frac{3}{2}\mu_i) . \qquad (17)$$

Following Moore's technique[11], the magnitudes of each of the terms in equations (7) and (8) are compared. By retaining only terms of order δ where δ is the thickness of the boundary layer, the following boundary layer equation can be obtained,

$$\frac{\bar{v}_\theta}{r}\frac{\partial v^1_\theta}{\partial\theta} + \frac{\bar{v}_\theta}{r}\frac{\partial \bar{v}_\theta}{\partial\theta} + \bar{v}_r \frac{\partial v^1_\theta}{\partial r} \doteq \nu \frac{\partial^2 \bar{v}_\theta}{\partial r^2}. \qquad (18)$$

Substituting expressions $\bar{v}_\theta$ and $\bar{v}_r$ from equations (3-6), changing r to y where y=r-R and retaining only terms of first order in y, equation (18) can be rewritten as

$$v^1_\theta \cos\theta + \sin\theta \frac{\partial v^1_\theta}{\partial\theta} - 2y\cos\theta\frac{\partial v^1_\theta}{\partial y} = \frac{2}{3}\frac{R\nu}{v_\infty}\frac{\partial^2 v^1_\theta}{\partial y^2}. \qquad (19)$$

This result applies both inside and outside the drop.

Equation (19) coupled with the boundary conditions (equations (11-14) and (17)) can be solved by integral transform method. The solutions are

$$v_{\theta_o}^1 = -6v_\infty \left(\frac{\nu_o}{Rv_\infty}\right)^{\frac{1}{2}} \sin\theta\chi^{\frac{1}{2}}(\theta)\Phi(\mu,\rho)f(Y_o/2\chi^{\frac{1}{2}}(\theta)), \tag{20}$$

where

$$\chi(\theta) = \frac{2}{3}\csc^4\theta\left(\frac{2}{3} - \cos\theta + \frac{1}{3}\cos^3\theta\right), \tag{21}$$

$$Y_o = \frac{r-R}{R}\left(\frac{Rv_\infty}{\nu_o}\right)^{\frac{1}{2}}, \tag{22}$$

$$\Phi(\mu,\rho) = \left(1 + \frac{3}{2}\frac{\mu_i}{\mu_o}\right) \Bigg/ \left[1 + \left(\frac{\rho_i\mu_i}{\rho_o\mu_o}\right)^{\frac{1}{2}}\right], \tag{23}$$

$$f(x) = \text{ierfc}\ x, \tag{24}$$

and

$$v_{\theta_i}^1 = -6v_\infty\left(\frac{\nu_o}{Rv_\infty}\right)^{\frac{1}{2}} \sin\theta\ \chi^{\frac{1}{2}}(\theta)\Phi(\mu,\rho)\ f(|Y_i|/2\chi^{\frac{1}{2}}(\theta)), \tag{25}$$

where

$$Y_i = \frac{r-R}{R}\left(\frac{Rv_\infty}{\nu_i}\right)^{\frac{1}{2}}. \tag{26}$$

This is the tangential component of the velocity. The solution reduces to Moore's[11] gas-liquid case when $\mu_i \ll \mu_o$ and $\rho_i \ll \rho_o$.

3. Mass Transfer

For a binary, dilute liquid solution of constant density and diffusivity, the steady convective diffusion equation can be written as

$$\underline{v}\cdot\nabla c = D\nabla^2 c, \tag{27}$$

where c is the concentration of one component in the binary mixture and D is the diffusivity.

The boundary conditions are

1. As $r\to\infty$, $c=c_\infty$, (28)
2. At $r=R$, $c=c_s$, (29)
3. At $\theta=0$, $\frac{\partial c}{\partial\theta} = 0$. (30)

The solution of this problem depends largely on two simplifications.

At large Re, for most practical systems, the Pe will even be larger, consequently a thin mass transfer boundary layer results. This leads to two approximations:

1. The first derivative of concentration can be neglected as compared to the second derivative,

$$\frac{2}{r}\frac{\partial c}{\partial r} \ll \frac{\partial^2 c}{\partial r^2} ,$$

2. The stream function which will be defined later can be approximated by the first two terms in an expansion in powers of y.

The second simplification arises from the ability to reduce the convective diffusion equation to the one-dimensional heat conduction equation with a readily available solution.

Equation (27) may now be written in spherical coordinates,

$$v_r \frac{\partial c}{\partial r} + \frac{v_\theta}{r}\frac{\partial c}{\partial \theta} \doteq D \frac{\partial^2 c}{\partial r^2} . \qquad (31)$$

Here we have omitted from the right side of the equation the angular portion of the Laplacian, $\frac{1}{r^2 \sin\theta}\frac{\partial}{\partial\theta}(\sin\theta \frac{\partial c}{\partial\theta})$, since the derivatives along the surface of the sphere are small compared to the derivatives along the radius vector.

The equation of continuity,

$$\frac{1}{r}\frac{\partial}{\partial r}(r^2 v_r) + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(v_\theta \sin\theta) = 0 , \qquad (32)$$

can be replaced by introducing the stream function ψ where

$$v_\theta = -\frac{1}{r\sin\theta}\frac{\partial\psi}{\partial r} , \quad v_r = \frac{1}{r^2\sin\theta}\frac{\partial\psi}{\partial\theta} . \qquad (33)$$

After changing variable r to ψ in equation (31) and evaluating the equation at $r \approx R$ (or $y \approx 0$), we obtain

$$\frac{\partial c}{\partial\theta} = DR^3 (v_\theta)_{y=0} \sin^2\theta \frac{\partial c}{\partial\psi} . \qquad (34)$$

Using equations (5) and (20) for v_θ and integrating equation (33) for ψ, we obtain

$$(v_\theta)_{y=0} = \frac{3}{2} v_\infty \sin\theta \left[1 - \frac{8}{3}\sqrt{\frac{1}{\pi Re}}\, \Phi(\mu,\rho)\csc^2\theta\,(1-\cos\theta)(2+\cos\theta)^{\frac{1}{2}}\right], \quad (35)$$

and

$$(\psi)_{y<<R} = -\frac{3}{2} v_\infty R y \sin^2\theta\left[1 - \frac{8}{3}\sqrt{\frac{1}{\pi Re}}\, \Phi(\mu,\rho)\csc^2\theta(1-\cos\theta)(2+\cos\theta)^{\frac{1}{2}}\right] \quad (36)$$

The problem can be simplified further by introducing t where

$$t = \frac{3}{2} D v_\infty R^3 \int \sin^3\theta \left[1 - \frac{8}{3}\sqrt{\frac{1}{\pi Re}}\, \Phi(\mu,\rho)\csc^2\theta(1-\cos\theta)(2+\cos\theta)^{\frac{1}{2}}\right] d\theta$$

$$\equiv \frac{3}{2} D v_\infty R^3 \int F(\theta)\, d\theta\ . \quad (37)$$

Combining equations (35) and (37), equation (34) can be transformed into the one-dimensional heat conduction equation,

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial \psi^2}\ . \quad (38)$$

The boundary conditions (equations (28-30)) can be rewritten in terms of the new variables t and ψ:

1. As $\psi\to\infty$, $c=c_\infty$, (39)
2. At $\psi=0$, $c=c_s$, (40)
3. At $t = \frac{3}{2} D v_\infty R^3 \left(\int F(\theta)d\theta\right)_{\theta=0}$, $\frac{\partial c}{\partial t} = 0$. (41)

The solution of equation (38) subjected to these boundary conditions can easily be found by the method of similarity transform,

$$\frac{c-c_s}{c_\infty - c_s} = \text{erf}\ \eta\ , \quad (42)$$

where the similarity variable

$$\eta = \left(\frac{3}{8}\right)^{\frac{1}{2}}\left(\frac{v_\infty}{DR}\right)^{\frac{1}{2}} \frac{y}{\sin\theta}\ \frac{F(\theta)}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}}\ . \quad (43)$$

The diffusion flux j to the surface is found by differentiating equation (42),

$$j = D\left(\frac{\partial c}{\partial y}\right)_{y=0} = \left(\frac{3}{2\pi}\right)^{\frac{1}{2}}\left(\frac{Dv_\infty}{R}\right)^{\frac{1}{2}}(c_\infty - c_s)\frac{F(\theta)}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}} . \qquad (44)$$

The Nernst diffusion layer thickness δ_N defined by

$$\delta_N = D(c_\infty - c_s)/j, \qquad (45)$$

is, therefore, given by

$$\frac{\delta_N}{R} = 2.05\frac{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}}{F(\theta)} Pe^{-\frac{1}{2}} . \qquad (46)$$

As $Pe \to \infty$, $\delta_N \to 0$ and as $\theta \to \pi$, $\delta_N \to \infty$. The latter contradicts the thin diffusion boundary layer model at high Pe. As the rear stagnation point is approached, the magnitude of the tangential and the radial diffusion become equal in importance. The anomaly is caused by neglecting the tangential diffusion term during the derivation of equation (31).

The total mass transfer flux, F, to the drop can be found by integrating j from equation (44) over the total area of the drop,

$$F = 2\pi R^2\int_0^\pi j \sin\theta \, d\theta = \sqrt{6\pi}\, R^2\left(\frac{Dv_\infty}{R}\right)^{\frac{1}{2}}(c_\infty - c_s)\int_0^\pi\frac{F(\theta)\, d\theta}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}} . \qquad (47)$$

The Nusselt number for mass transfer is, therefore,

$$Nu = \frac{2kR}{D} = \frac{2FR}{4\pi R^2(c_\infty - c_s)D} = \sqrt{\frac{3}{4\pi}}\, I(Re,\mu,\rho)\, Pe^{\frac{1}{2}} , \qquad (48)$$

where

$$I(Re,\mu,\rho) \equiv \int_0^\pi\frac{F(\theta)\, d\theta}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}} . \qquad (49)$$

As $Re \to \infty$, the result reduces to the well-known potential solution of Boussinesq[1],

$$Nu = 1.128\, Pe^{\frac{1}{2}} . \qquad (50)$$

Equation (48) is presented in graphical form in Figures 1 to 4. For gas bubbles rising in a liquid, $\Phi(\mu,\rho)$ is taken as unity due to the small viscosity and density of the gas phase as compared to that of the

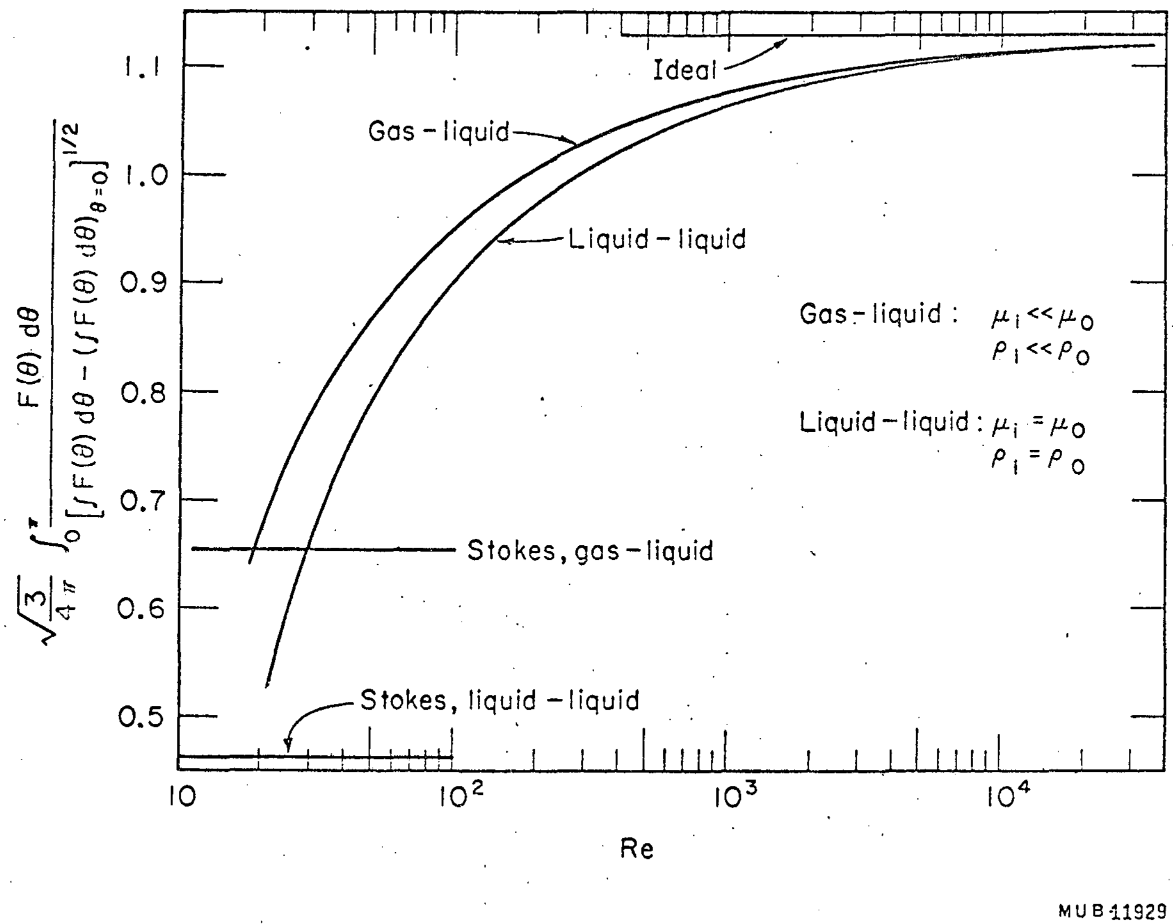

Figure 1. The Function $\sqrt{\frac{3}{4\pi}}\, I(Re,\mu,\rho)$ versus Re.

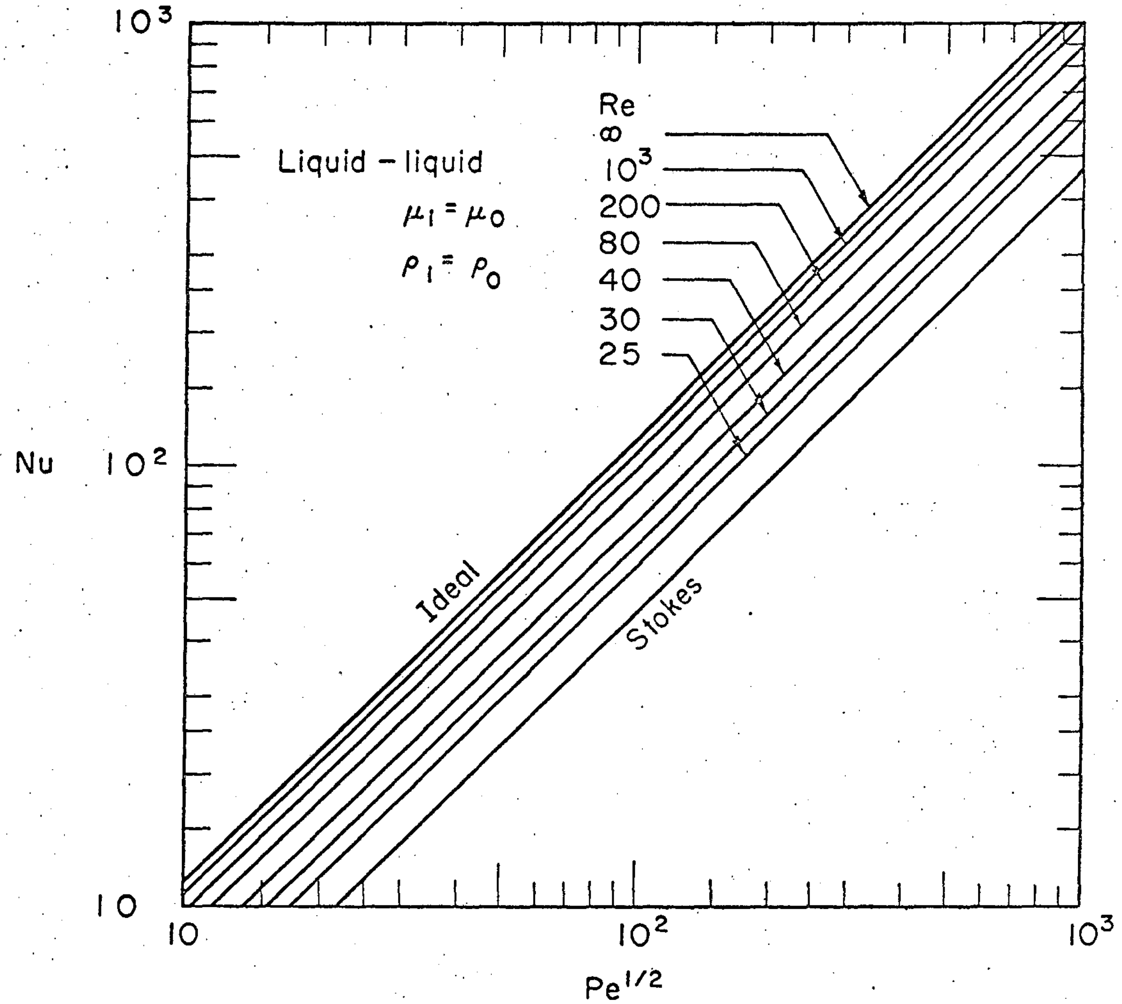

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Figure 2. Mass Transfer to Liquid Drops.

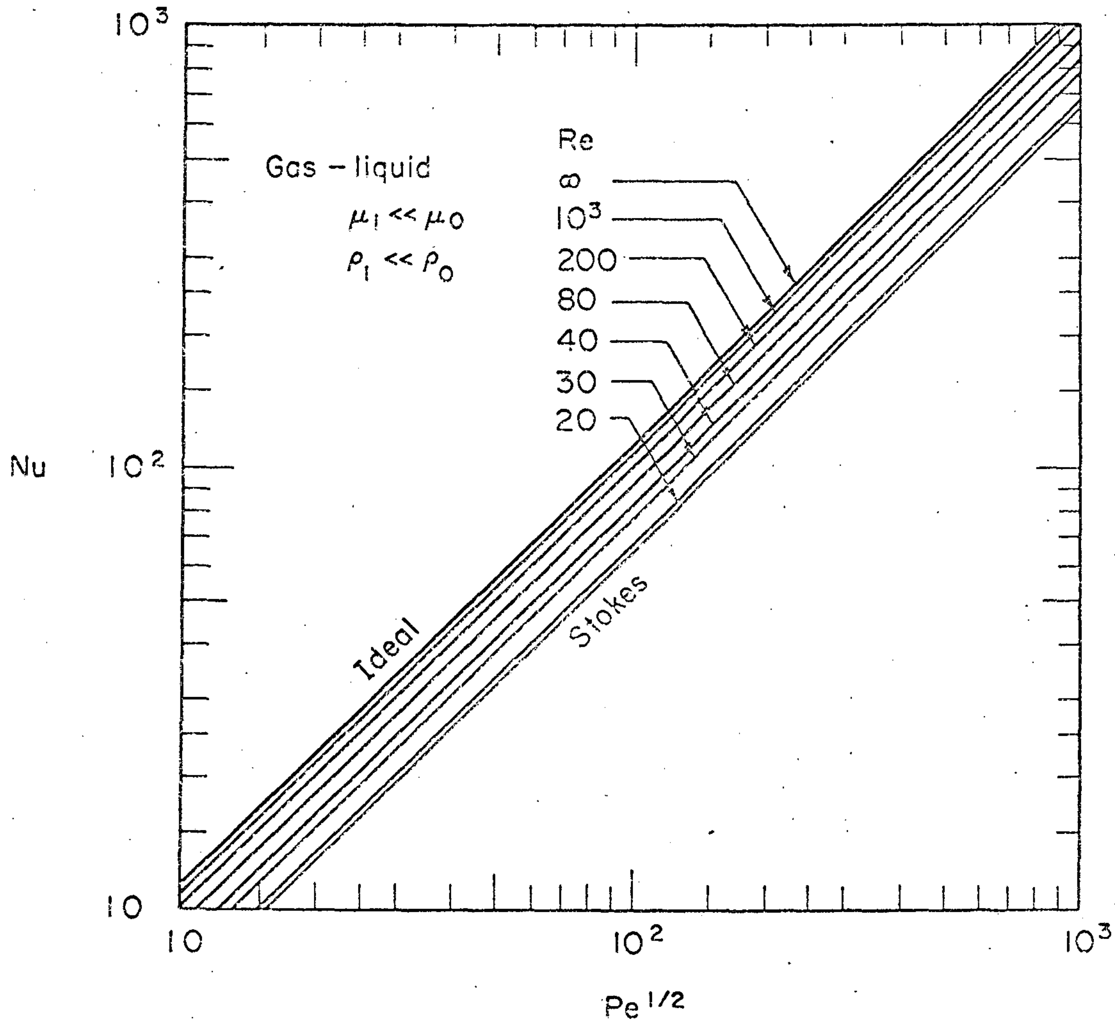

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Figure 3. Mass Transfer to Gas Bubbles.

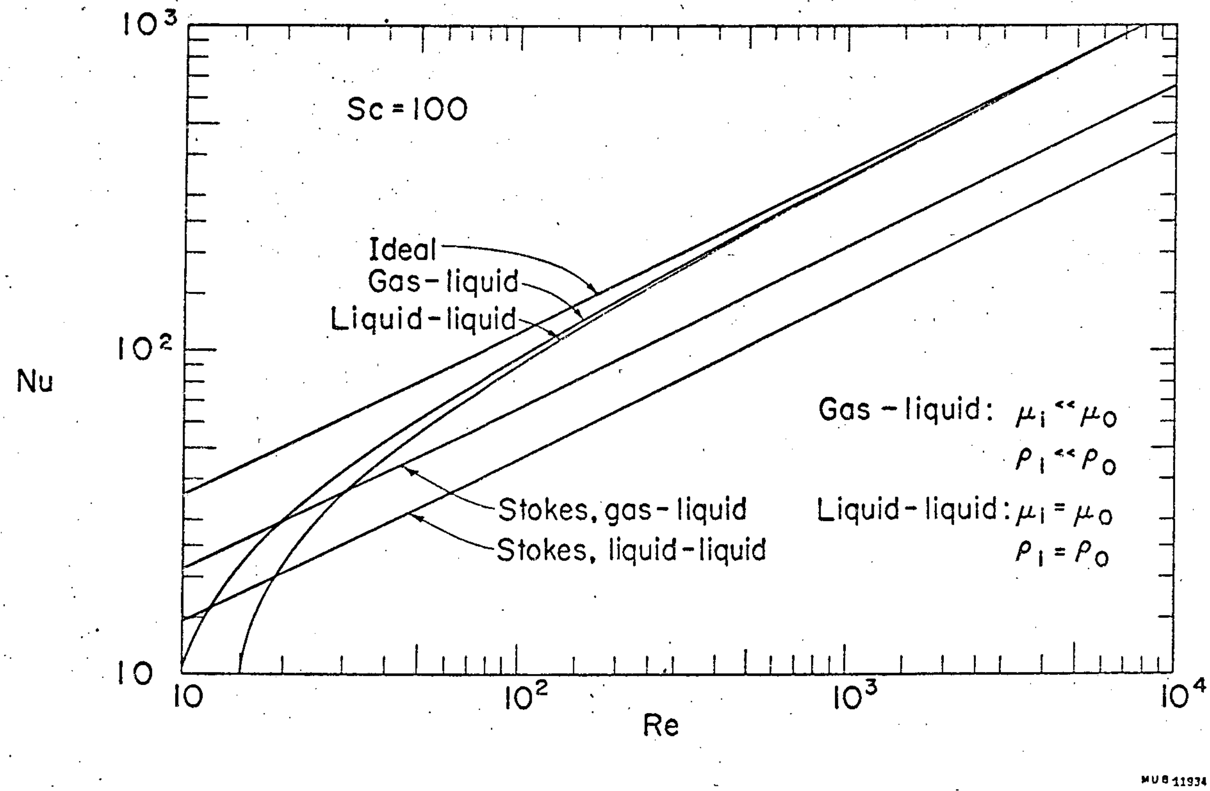

Figure 4. Mass Transfer to Drops and Bubbles
(Nu versus Re)

liquid phase. For liquid drops rising in a liquid medium, the case of $\mu_i = \mu_o$ and $\rho_i = \rho_o$ is taken for illustration. Both cases approach the Boussinesq's potential solution asymptotically as $Re \to \infty$. At lower Re, the mass transfer is smaller for the liquid-liquid case than for the gas-liquid case due to the slower tangential motion at the liquid-liquid interface. The solution does not approach the Stokes solution at very low Re because of the very different approximations used in solving the fluid mechanics for the two cases.

4. Comparison with Available Experimental Data

A. Liquid Drops

Heertjes et al[6] reported mass transfer data between isobutanol and water. Experimental results are plotted in Figures 5 and 6. The present calculation shows a considerable improvement over the calculations based on either the Stokes flow or the ideal fluid model.

B. Gas Bubbles

Bowman and Johnson[2] measured the mass transfer to carbon dioxide bubbles in water. Graphical relationship between the relative velocity of rise, the liquid phase mass transfer coefficient and the bubble volume was presented. The bubbles under their experimental conditions are not spherical. An eccentricity factor not reported was used in their calculation. Fortunately, a correlation of such an eccentricity versus the bubble volume was given by Li et al[9]. Using this information and the diffusivity of 2.07×10^{-5} cm^2/sec [12], we calculated the Nu as a function of Re as shown in Figure 7. The potential solution predicts a Nu approximately 12% high whereas the present boundary layer approach predicts to about 8% high. Levich's solution[8] using Stokes flow is too low.

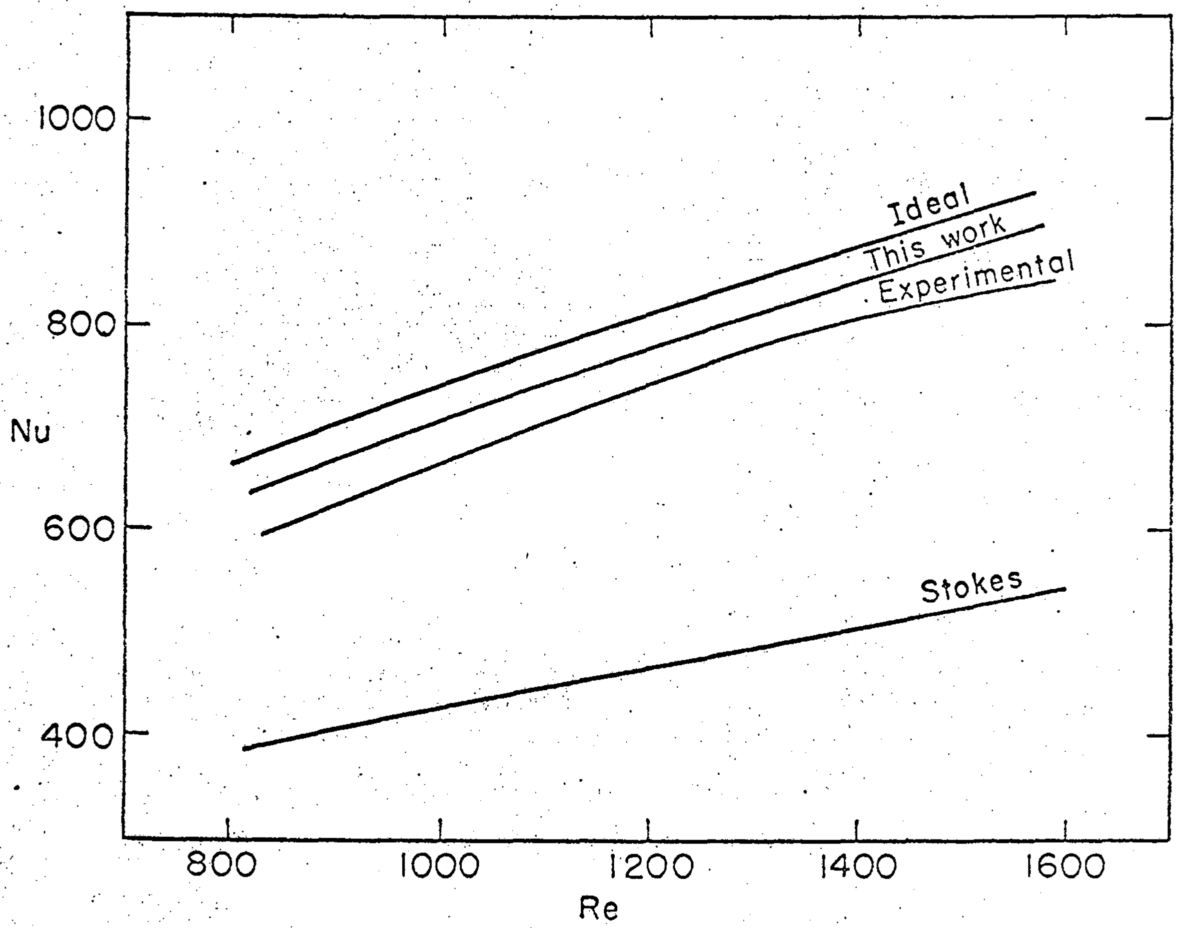

Figure 7. Carbon Dioxide Bubbles in Water.

Considering the uncertainties in these measurements, our agreement with the data is quite satisfactory.

5. Conclusions

Mass transfer to drops or bubbles rising steadily through a liquid at high Re was calculated from a boundary layer solution of the fluid mechanics. Despite the approximations and assumptions used in the derivations, the result compares favorably with the experimental observations. The calculation is especially satisfactory at liquid-liquid interfaces where it should yield useful information to many practical processes.

Acknowledgement

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Nomenclature

c Concentration of a species in a system

d Diameter of a drop or bubble

D Diffusion coefficient of a species in a homogeneous medium

f A shorthand notation for ierfc

F A function of Re, θ, μ and ρ = $\sin^3\theta \left[1 - \frac{8}{3}\sqrt{\frac{1}{\pi Re}}\, \Phi(\mu,\rho)\csc^2\theta(1-\cos\theta)(2+\cos\theta)\right]^{\frac{1}{2}}$

F Total mass transfer flux to a drop or a bubble

I A function of F(θ) = $\int_0^{\pi} \frac{F(\theta)\, d\theta}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}}$

j Local mass transfer flux to the drop or bubble surface

k Mass transfer coefficient in the continuous phase

p Pressure in a system

r Radial coordinate

R Radius of a drop or bubble

t A variable $= \frac{3}{2} Dv_\infty R^3 \int F(\theta)\, d\theta$

v Velocity of a fluid

y A variable = r - R

Y A dimensionless variable $= \frac{r-R}{R}\left(\frac{Rv_\infty}{\nu}\right)^{\frac{1}{2}}$

Nu Nusselt number for mass transfer = kd/D

Pe Peclet number = Re x Sc = $v_\infty d/D$

Re Reynolds number = $v_\infty d/\nu$

Sc Schmidt number = ν/D

δ Thickness of the boundary layer

δ_N Thickness of the Nernst diffusion layer

η A similarity variable $= \left(\frac{3}{8}\right)^{\frac{1}{2}}\left(\frac{v_\infty}{DR}\right)^{\frac{1}{2}} \frac{y}{\sin\theta} \frac{F(\theta)}{\left[\int F(\theta)d\theta - \left(\int F(\theta)d\theta\right)_{\theta=0}\right]^{\frac{1}{2}}}$

θ Angular coordinate

μ Viscosity of a fluid

ν Kinematic viscosity of a fluid

ρ Density of a fluid

τ Magnitude of shear stress at fluid-fluid interface.

Φ A function of μ and ρ $= \left(1 + \frac{3}{2}\frac{\mu_i}{\mu_o}\right) \Big/ \left(1 + \left(\frac{\rho_i \mu_i}{\rho_o \mu_o}\right)^{\frac{1}{2}}\right)$

χ A function of $\theta = \frac{2}{3} \csc^4\theta \left(\frac{2}{3} - \cos\theta + \frac{1}{3}\cos^3\theta\right)$

ψ Stream function of a fluid in motion

Superscripts

\- Refers to the potential solution of ideal fluids

1 Refers to quantitites of first-order perturbation

Subscripts

i Refers to properties inside a drop or bubble

o Refers to properties outside a drop or bubble

s Refers to quantities at equilibrium

∞ Refers to value in the free stream

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