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GAS ABSORPTION INTO TURBULENT LIQUIDS AT INTERMEDIATE CONTACT TIMES

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NOMENCLATURE

- a, eddy diffusivity parameter $[s^{-1}]$;
- c, concentration of diffusing gas $\lceil g \bmod \lfloor \epsilon \rceil \right]$;
- c_0 , initial concentration [g moles/cm³];
- c_s , interfacial concentration [g moles/cm³];
- $d,$ film thickness \lfloor cm \rfloor ;
- D, molecular diffusivity $\lceil \text{cm}^2/\text{s} \rceil$;
- k_c , local mass transfer coefficient

$$
=\frac{-D}{(c_s-c_0)}\frac{\partial c}{\partial y}\bigg|_{y=0}\text{[cm/s];}
$$

 k_{c} , average mass transfer coefficient

$$
= 1/x \int_{0}^{x} k_c dx \, [\text{cm/s}];
$$

-
- L_e , mass transfer entrance length [cm];
 \bar{L}_e , dimensionless mass transfer entra $dimensionless mass transfer entrance length =$ $(L_{e}D)/(ud^{2})$;
- q, volumetric flow rate per unit perimeter $[\text{cm}^2/\text{s}]$;
- Re , Reynolds number = $4q/v$;
- *Sh,* $\log \text{S}$ local Sherwood number = $k_{c}d/D$;
- Sh_m , mean Sherwood number = $\bar{k}_c d/D$;
- u , velocity \lceil cm/s];
- $v'c'$, time averaged value of the fluctuating components of the *v*-velocity and concentration $[g \text{ moles/cm}^3];$ x, distance in direction of flow \lceil cm \rceil ;
- \bar{x} , dimensionless axial distance = $(xD)/(ud^2)$;
- x_1 , limit of applicability of equation (9) [cm];
- y , distance normal to the interface $[cm]$;
- \bar{y} , dimensionless normal distance = y/d .

Greek letters

$$
\beta, \qquad \text{dimensionless diffusion parameter} = (ad^2)/D;
$$

$$
\varepsilon, \qquad \qquad \text{eddy diffusivity} = \frac{\overline{v'c'}}{\mathrm{d}c/\mathrm{d}y} \left[\text{cm}^2/\text{s} \right];
$$

- η , dimensionless $y = y/2(Dx/u)^{1/2}$;
- θ . dimensionless concentration, $(c_s - c)/(c_s - c_0)$;
- θ_{0} concentration function defined by equation (7)

$$
=\frac{2}{\sqrt{\pi}}\int_0^{\eta}e^{-\eta^2}d\eta;
$$

 θ_{1} concentration function defined by equation (7);

V, kinematic viscosity \lfloor cm²/s];

- π , $= 3.1416$;
- ρ . density $\lceil g/cm^3 \rceil$.

UNDER usual operating conditions for gas absorption the controlling mass transfer resistance resides in the liquid phase. The work reported here is concerned with describing liquid phase mass transfer in terms of an eddy diffusivity for gas absorption in turbulent film flow $(4q/v > 1200)$ for the case where the concentration profiles are not fully developed. The motivation for this work is that for many applications, such as liquid flow in packed columns, the film lengths or contact times between complete mixing may not be sufficiently long to achieve fully developed conditions in the liquid. In this case information regarding the entrance region mass transfer coefficient would be of interest.

Liquid phase mass transfer across a free surface has been treated in terms of an eddy diffusivity by Levich [1], Davies [2] and King [3]. Lamourelle and Sandall [4] have experimentally determined the behavior of the eddy diffusivity near a free surface by absorbing four different gases into turbulent water films in a long wetted-wall column. These authors found that the eddy diffusivity varies as the square of the distance from the free surface.

$$
\varepsilon = ay^2. \tag{1}
$$

For water at 25°C, a was found to be

$$
a = 7.90 \times 10^{-5} \text{ Re}^{1.678}.
$$
 (2)

Equation (1) is valid for the region adjacent to the free surface. The major resistance to mass transfer occurs close to the surface because of the large Schmidt numbers usually encountered in gas absorption, and thus it is important to know the eddy diffusivity accurately only for this region.

The differential equation describing diffusion in twodimensional, fully developed flow may be written in terms of an eddy diffusivity as

$$
u(y)\frac{\partial \theta}{\partial x} = \frac{\partial}{\partial y}\left\{ (D + \varepsilon(y))\frac{\partial \theta}{\partial y} \right\}.
$$
 (3)

Equation (3) neglects axial transport and assumes no diffusion-induced velocity. The boundary conditions of interest are expressed as

$$
x = 0, \qquad \theta = 1
$$

(ii)
$$
y = 0.
$$
 $\theta = 0$ (4)

(iii)
$$
y = d
$$
, $\partial \theta / \partial y = 0$.

The first condition expresses the fact that the liquid enters the mass transfer section completely mixed. The second and third conditions state that the concentration remains constant at the gas-liquid interface, and that no transport occurs across the solid boundary.

Two simplifying assumptions are made in solving equations (3) and (4). It is assumed that the velocity, u , may be taken to be constant at its surface value. This assumption is made since the velocity gradient vanishes at the surface for the case of no shear at the interface, and since for the relatively short contact times in this study the absorbing gas will not penetrate very far into the film. It is further assumed that the eddy diffusivity given by equation (1) for the surface region is valid over the complete film thickness. This assumption should not lead to any significant error since the major resistance to mass transfer lies near the surface.

ASYMPTOTIC SOLUTIONS

As pointed out by King [3], asymptotic solutions of equation (3) may be obtained for the case of very short contact times and for very long contact times. The contact time being defined as the film length divided by the surface velocity. In the limit of very short film lengths the absorbing gas does not penetrate far enough into the film for the eddy diffusion term to have an effect on the rate of mass transfer. In this case the Higbie penetration theory [5] describes the rate of absorption and the Sherwood number is given by

$$
Sh = \sqrt{1/\pi \bar{x}}.
$$
 (5)

For long film lengths the concentration profiles become fully-developed, and under these simplifying conditions Lamourelle and Sandall [4] integrate equation (3) to obtain

$$
Sh = 2\sqrt{\beta/\pi}.
$$
 (6)

APPROXIMATE ANALYTICAL SOLUTION

An approximate solution valid for short film lengths may be obtained by considering a solution of the form

$$
\theta = \theta_0(\eta) + x\theta_1(\eta) + \dots \tag{7}
$$

where θ_0 is the penetration theory solution. Substitution of equation (7) into equation (3) and neglecting terms of order x results in an equation for θ_1 . This equation was solved by a fourth-order Runge-Kutta integration procedure $[6]$ using a shooting method. The quantity of interest is the derivative at the free surface which was found to be

$$
\left. \frac{d\theta_1}{d\eta} \right|_{\eta = 0} = 0.5642 \, a/u. \tag{8}
$$

With this value for

$$
\left.\frac{\mathrm{d}\theta_1}{\mathrm{d}\eta}\right|_{\eta=0}.
$$

the Sherwood number is given by

$$
Sh = \frac{1}{2} \sqrt{\frac{1}{x}} \left[\frac{2}{\sqrt{\pi}} + 0.5642 \beta \overline{x} \right].
$$
 (9)

Equation (9)is an approximate analytical expression for the local Sherwood number which is expected to give good results for small \bar{x} . The region of applicability of equation (9), however, can only be determined by comparison with exact results.

1t may be seen by inspection that equation (9) predicts a minimum in the Sherwood number and thus should not be applied beyond this minimum. This limiting value of \tilde{x} is found to be $\bar{x}_1 = 2.00/\beta$.

NUMERICAL SOLUTION

Equation (3) may be written in dimensionless form as

$$
\frac{\partial \theta}{\partial \bar{x}} = (1 + \bar{y}^2 \beta) \frac{\partial^2 \theta}{\partial \bar{y}^2} + 2\beta \bar{y} \frac{\partial \theta}{\partial \psi}.
$$
 (10)

In dimensionless form the boundary conditions become

$$
\bar{x} = 0, \qquad \theta = 1
$$

(ii)
$$
\bar{y} = 0, \qquad \theta = 0 \tag{11}
$$

(iii) $\bar{y} = 1$, $\partial \theta / \partial \bar{y} = 0$.

Equations (10) and (11) were solved numerically by replacing the derivatives with finite difference approximations. The Crank-Nicholson [7] procedure was used for the finite difference representation of equation (10). The resulting simultaneous algebraic equations were solved using the method of Thomas [8].

Figure 1 shows some results of the numerical calculations plotted as Sherwood number vs downstream distance. The curves in Fig. 1 show a smooth transition from the upstream asymptote to the downstream asymptote. For the special case of carbon dioxide absorption in water at $25^{\circ}C$, the range in β shown in Fig. 1 would cover Reynolds numbers from 1100 to 11 000.

Figure 2 shows the approximate analytical formula, equation (9), compared to the exact results for $\beta = 50000$. It is seen that the approximate relationship gives good agreement for small \bar{x} , the deviation increasing to about 15 per cent as the fully-developed Sherwood number is approached. Similar behavior was found for the other values of β for which numerical results were obtained.

It is possible to empirically modify equation (9) to obtain better agreement with the exact results. If the form of equation {9) is retained but the numerical constant in the second term is considered to be an adjustable parameter, then equation (9) may be rewritten as

$$
Sh = 0.564 / \sqrt{\bar{x}} + 0.1975 \beta \sqrt{\bar{x}}.
$$
 (12)

FIG. 1. Development of mass transfer coefficient in entrance region.

FIG. 2. Comparison of approximation formula, equation (14) with exact result for $\beta = 50000$.

Equation (12) predicts the Sherwood number to within a maximum deviation of 2.5 per cent for \bar{x} varying from zero up to the entrance length over the entire range of β investigated (500 < β < 500 000). On the scale of Fig. 2, equation (12) can not be distinguished from the numerical solution.

The mass transfer coefficient given by equation (12) may be integrated over the film length to give an expression for the mean Sherwood number based on the average mass transfer coefficient

$$
Sh_m = 1.128/\sqrt{\bar{x}} + 0.1317 \beta \sqrt{\bar{x}}.
$$
 (13)

Since equation (12) is valid up to the entrance length, it may be used to predict the entrance length. Substituting for the Sherwood number in equation (12) as 1.05 times the fully developed Sherwood number as given by equation (6) and solving for the entrance length gives an equation for the entrance length.

$$
\bar{L}_e = 2.56/\beta. \tag{14}
$$

As was mentioned previously, the eddy diffusivity given by equation (1) is strictly valid only in the surface region; however, it was used over the entire film thickness. The magnitude of the error incurred as a result of this assumption was estimated by carrying out the integration of equation (10) using a more realistic distribution in the bulk liquid. The eddy diffusivity expression of Reichardt $[9]$ was used for this calculation. For the absorption at 25° C of carbon dioxide in a vertical falling film of water with a Reynolds number of 5000 ($\beta = 54,700$) it was found that the maximum difference in the calculated Sherwood numbers was 0.7 per cent. This maximum deviation occurred for the fully developed Sherwood number. Thus it is seen that, as expected, the use of equation (I) in the bulk liquid does not result in any significant error.

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INERT GAS EFFECTS IN THE INCIPIENT BOILING OF ALKALI LIQUID METALS

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POOL-BOILING ARTIFICIAL-CAVITY EXPERIMENTS

INCIPIENT boiling superheats of alkali metals measured by different experimenters, and usually even by the same experimenter, are sensitive to small variations in experi-

mental conditions, such as the concentrations of trace contaminants, the presence or absence of small amounts of entrained or dissolved gas, and the prior history of the system up to the point where the first bubble is produced. Even with considerable precautions, the boiling of alkali metals from natural surfaces at low heat fluxes tends to be quite unstable, with large resulting variations in the incipient

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