t = time for which heater held at given heat flux, min.
t_i = time to increase heat flux from one setting to another, min.
ΔT_{sat} = temperature at metallic surface of heater minus saturation temperature of liquid, °F.
θ = angle from topmost point of bubble to contact point on surface (Figure 4) radians or deg.
ρ = liquid density, $lb._m/cu. ft.$
σ = liquid to vapor surface tension, $lb._f/ft.$

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Manuscript received July 12, 1963; revision received October 25, 1963; revision accepted October 25, 1963. Paper presented at A.I.Ch.E. San Juan meeting.

Gas Absorption Accompanied by a Large Heat Effect and Volume Change of the Liquid Phase

S. H. CHIANG and H. L. TOOR
University of Pittsburgh, Pittsburgh, Pennsylvania

Gas absorption is always accompanied by a heat effect and volume change of the liquid phase. The magnitude of the heat effect and volume change depends largely on the rate of transfer. In the case of absorption of a sparingly soluble gas these effects are usually negligible, and the process may be considered to be isothermal and isometric. On the other hand in many industrially important operations, such as absorption of ammonia by water, the heat effect and volume change are so large that they cannot be ignored. The problem to be solved in such transfer processes is the one involving simultaneous mass and heat transfer with a moving boundary.

In the past several papers (*1, 4, 7*) have appeared dealing with the moving boundary problem for either mass transfer or heat transfer but not for the coupled transfer process. In this paper a solution is obtained for the coupled process, and the results are compared with measurements of the rate of absorption of ammonia by water.

PHYSICAL MODEL AND ASSUMPTIONS

The physical model as shown in Figure 1, consists of a semi-infinite liquid phase in contact with a pure gas phase. As the gas phase consists of only one component and the liquid is assumed to be nonvolatile, no concentration gradient will exist in the gas phase. The concentration $c^*(\theta)$ at the liquid surface is assumed to be always in equilibrium with the gas phase at the liquid surface temperature $t^*(\theta)$. The liquid surface moves at a velocity $v(\theta)$, which is in direct proportion to the amount of gas absorbed.

Other assumptions involved in the present treatment are listed below:

1. The heat of solution (which may also include heats of reaction) is released instantaneously at the interface when the gas hits the liquid surface.
2. Physical properties and transport coefficients are constant at certain average values.
3. The volume change of the liquid phase is directly proportional to the quantity of dissolved gas; that is partial molal volumes are constant.
4. The Dufour and Soret effects are negligible.
5. The heat transfer between the gas and the liquid surface is negligible.

THE RATE EQUATIONS

Based on the physical model and the assumptions stated above the rate equations and boundary conditions may be expressed as follows:

For mass transfer

H. L. Toor is at Carnegie Institute of Technology, Pittsburgh, Pennsylvania.

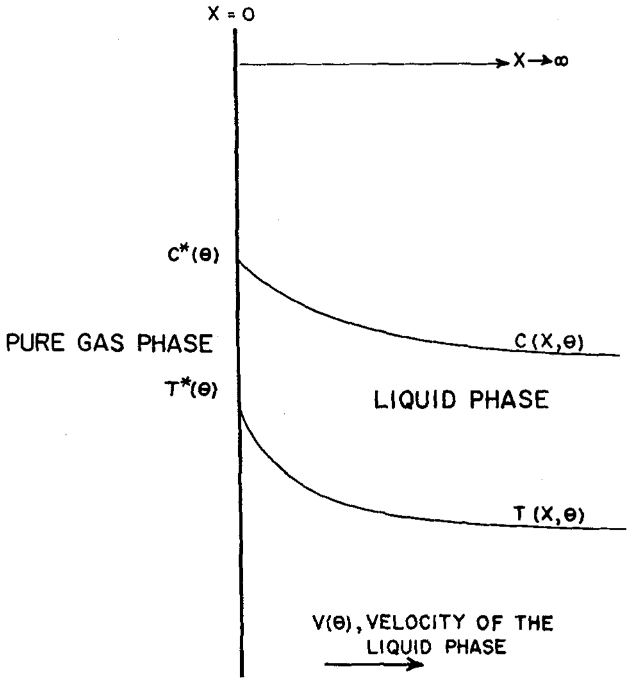

Fig. 1. Physical model.

$$\frac{\partial c}{\partial \theta} + v(\theta)\frac{\partial c}{\partial x} = D\frac{\partial^2 c}{\partial x^2} \tag{1}$$

$$c(0,\theta) = c^*(\theta) \tag{2a}$$

$$c(\infty,\theta) = 0 \tag{2b}$$

$$c(x,0) = 0 \tag{2c}$$

$$v(\theta) = -\frac{D}{\rho_m - c^*(\theta)}\left.\frac{\partial c}{\partial x}\right|_{x=0} \tag{3}$$

For heat transfer

$$\frac{\partial t}{\partial \theta} + v(\theta)\frac{\partial t}{\partial x} = \alpha\frac{\partial^2 t}{\partial x^2} \tag{4}$$

$$t(0,\theta) = t^*(\theta) \tag{5a}$$

$$t(\infty,\theta) = 0 \tag{5b}$$

$$t(x,0) = 0 \tag{5c}$$

$$v(\theta) = -\frac{k}{\Delta H_s\,\rho_m}\left.\frac{\partial t}{\partial x}\right|_{x=0} \tag{3a}$$

Equations (1) and (3) have been derived by using a volume reference frame, since it is in this reference frame that $v = v(\theta)$ when the mixture density is a function of concentration.

Equation (4) is written in the same reference frame and is most easily justified by assuming in addition to the earlier assumptions that the heat capacity times the molal density of the two components do not differ significantly.

The relationship between the surface concentration and surface temperature is assumed to be known. It may be approximated by linear expression over a limited temperature range:

$$c^*(\theta) = a\,t^*(\theta) + c^*(0) \tag{6}$$

$c^*(0)$ represents the surface concentration corresponding to liquid temperature at $\theta = 0$, and the temperature scale is so chosen that the initial liquid temperature is zero at $\theta = 0$. This linear relationship is convenient but not necessary for subsequent work.

The mass and heat flux at the interface are related by the equation

$$\Delta H_s f_m(\theta) = f_h(\theta) \tag{7}$$

or

$$\Delta H_s \rho_m \frac{D}{\rho_m - c^*(\theta)}\left.\frac{\partial c}{\partial x}\right|_{x=0} = k\left.\frac{\partial t}{\partial x}\right|_{x=0} \tag{8}$$

Since this may also be obtained from Equations (3) and (3*a*), it is not an independent condition. Equation (7) however must be satisfied even if $v(\theta)$ is neglected. Similarly Equations (2*a*) and (3) are not independent of each other, and Equations (5*a*) and (3*a*) are not independent of each other.

SOLUTIONS OF RATE EQUATIONS

Equations (1) and (4) have no known closed form solution if the surface concentration and surface temperature are arbitrary functions of time. In order to examine the functional dependency of the surface concentration and temperature on time Equations (1) and (4) will be first solved by dropping the convective terms [terms involving $v(\theta)$]. Equations (3) and (3*a*) then are not used but rather Equations (2*a*) and (5*a*), and the $c^*(\theta)$ and $t^*(\theta)$ must be chosen such that Equation (7) is satisfied.

First Approximation $v(\theta) = 0$

The solutions to Equations (1) and (4), when $v(\theta) = 0$, with boundary conditions (2) and (5) are well known (2). They can be written down immediately with the aid of Duhamel's theorem:

$$c = \frac{1}{\sqrt{D\pi}}\int_0^\theta f_m(\theta-\tau)\exp(-x^2/4D\tau)\frac{d\tau}{\sqrt{\tau}} \tag{9}$$

and

$$t = \frac{\sqrt{\alpha}}{k\sqrt{\pi}}\int_0^\theta f_h(\theta-\tau)\exp(-x^2/4\alpha\tau)\frac{d\tau}{\sqrt{\tau}} \tag{10}$$

From Equations (9) and (10) the surface concentration and temperature may be expressed as

$$c^*(\theta) = \frac{1}{\sqrt{D\pi}}\int_0^\theta f_m(\theta-\tau)\frac{d\tau}{\sqrt{\tau}} \tag{11}$$

and

$$t^*(\theta) = \frac{\sqrt{\alpha}}{k\sqrt{\pi}}\int_0^\theta f_h(\theta-\tau)\frac{d\tau}{\sqrt{\tau}} \tag{12}$$

Combining Equations (6), (7), (11), and (12) one gets

$$\int_0^\theta f_m(\theta-\tau)\frac{d\tau}{\sqrt{\tau}} = K_1 c^*(0)\sqrt{D\pi} \tag{13}$$

where

$$K_1 = \frac{1}{1 - \sqrt{D/c_p\rho k}\;\Delta H_s\,a} \tag{14}$$

Since K_1 is a constant, Equation (13) can easily be solved for the mass flux at the liquid surface

$$f_m(\theta) = K_1\,c^*(0)\sqrt{D/\pi\theta} \tag{15}$$

and from Equation (7)

$$f_h(\theta) = K_1\,\Delta H_s\,c^*(0)\sqrt{D/\pi\theta} \tag{16}$$

Integrate Equations (11) and (12) by using Equations (15) and (16) to give

$$c^*(\theta) = K_1\,c^*(0) \tag{17}$$

and

$$t^*(\theta) = K_1\,c^*(0)\,\Delta H_s\sqrt{D/c_p\rho k} \tag{18}$$

From these equations it is concluded that the surface concentration and surface temperature are constant. However they are different from the concentration and temperature which would be obtained if the process were carried out under isothermal conditions. Thus at the moment of contact the surface conditions are immediately adjusted to the final equilibrium values. This occurs because the rates of mass transfer and heat transfer at the surface are in direct proportion to each other.

Under such conditions the concentration and temperature distribution may be expressed as

$$c = K_1 c^*(0) \operatorname{erfc}(x/2\sqrt{D\theta}) \tag{19}$$

and

$$t = K_1 c^*(0)\, \Delta H_s \sqrt{D/c_p\rho k} \operatorname{erfc}(x/2\sqrt{\alpha\theta}) \tag{20}$$

Second Approximation $v(\theta) \neq 0$

In solving Equations (1) and (4) it is necessary to know the form of $c^*(\theta)$ and $t^*(\theta)$. Based on the result obtained in the first approximation the surface concentration and temperature are assumed to be constant and independent of time but different from the values corresponding to the isothermal case. This assumption now cannot be proven directly, but it will be justified by showing that the final solutions satisfy the physical problem.

With constant surface conditions Equations (1) and (4) can be independently solved by using the following transformation of coordinates:

$$y = \frac{x}{2\sqrt{D\theta}}; \quad y' = \frac{x}{2\sqrt{\alpha\theta}} \tag{21}$$

and let

$$v(\theta) = \beta\sqrt{D/\theta}; \quad v(\theta) = \beta'\sqrt{\alpha/\theta} \tag{22}$$

$$\beta' = \beta\sqrt{D/\alpha} \tag{22a}$$

where β and β' are constants to be determined. The solutions of Equations (1) and (4) with boundary conditions (2) and (5) are

$$c = c^*(\theta)\, \frac{1 + \operatorname{erf}(\beta - y)}{1 + \operatorname{erf}\beta} \tag{23}$$

and

$$t = t^*(\theta)\, \frac{1 + \operatorname{erf}(\beta' - y')}{1 + \operatorname{erf}\beta'} \tag{24}$$

The constants β and β' are related to the surface concentration and surface temperature by the following relationships:

$$c^*(\theta) = \frac{\rho_m \beta\sqrt{\pi} \exp(\beta^2)\, [1 + \operatorname{erf}\beta]}{1 + \beta\sqrt{\pi} \exp(\beta^2)\, [1 + \operatorname{erf}\beta]} \tag{25}$$

and

$$t^*(\theta) = \Delta H_s\, \rho_m\, (\alpha/k)\, \beta'\sqrt{\pi} \exp(\beta'^2)\, [1 + \operatorname{erf}\beta'] \tag{26}$$

Equations (25) and (26) may be solved simultaneously with Equation (22*a*) and the solubility-temperature relationship for β and β'. The trial-and-error computation can be simplified if the value of β' is sufficiently small, for then Equation (26) reduces to

$$\begin{aligned} t^*(\theta) &= \Delta H_s\, \rho_m\, \beta' \sqrt{\pi}\, (\alpha/k) \\ &= \Delta H_s\, \rho_m\, \beta \sqrt{\pi} \sqrt{D/C_p\, \rho\, k} \end{aligned} \tag{26a}$$

Using Equations (25), (26*a*), and (6) one obtains the following relationship:

$$\frac{\beta\sqrt{\pi} \exp(\beta^2)\, [1 + \operatorname{erf}\beta]}{1 + \beta\sqrt{\pi} \exp(\beta^2)\, [1 + \operatorname{erf}\beta]} = a\, \beta\, \Delta H_s \sqrt{\frac{\pi D}{c_p\, \rho\, k}} + \frac{c^*(0)}{\rho_m} \tag{27}$$

This equation can be used for the evaluation of β and β' for systems having small values of β' (say β' less than 0.05).

By differentiating Equations (23) and (24) the concentration and temperature gradient at the liquid surface are found to be

$$-\frac{\partial c}{\partial x}\bigg|_{x=0} = \frac{1}{\exp(\beta^2)\, [1 + \operatorname{erf}\beta]}\, \frac{c^*(\theta)}{\sqrt{D\theta\pi}} \tag{28}$$

and

$$-\frac{\partial t}{\partial x}\bigg|_{x=0} = \frac{1}{\exp(\beta'^2)\, [1 + \operatorname{erf}\beta']}\, \frac{t^*(\theta)}{\sqrt{\alpha\theta\pi}} \tag{29}$$

When one knows the concentration and temperature gradient, expressions for the mass flux and for the heat flux at the liquid surface may be obtained:

$$\begin{aligned} f_m(\theta) &= -\frac{D\rho_m}{\rho_m - c^*(\theta)}\, \frac{\partial c}{\partial x}\bigg|_{x=0} \\ &= \frac{c^*(\theta)\sqrt{D/\pi\theta}}{[1 + \operatorname{erf}\beta][1 - c^*(\theta)/\rho_m] \exp(\beta^2)} \end{aligned} \tag{30}$$

and

$$f_h(\theta) = -k\frac{\partial t}{\partial x}\bigg|_{x=0} = \frac{t^*(\theta)\sqrt{k\, c_p\, \rho/\pi\theta}}{[1 + \operatorname{erf}\beta'] \exp(\beta'^2)} \tag{31}$$

It can be easily shown that the solutions obtained above satisfy Equations (1), (2*b*), (2*c*), (3), (4), (5*b*), (5*c*), and (3*a*). This in fact shows that the solutions satisfy the physical problem and justifies the initial assumption of constant surface conditions.

DISCUSSION

The solutions given in the first approximation are essentially the same as that obtained by Dankwerts (5). The basic difference however lies in the initial assumptions. Danckwerts assumed that the surface concentration is a constant value corresponding to the initial temperature, while the present development allows the surface concentration to change. Even though the result of the present derivation gives a constant surface temperature and constant surface concentration, the values are different from the initial surface conditions.

Thus in the first approximation the rate of heat transfer and the temperature profiles follow the usual equations for conduction into a quiescent medium with a constant surface temperature of $t^*(\theta)$ [Equations (16), (18), and (20)], while the rate of mass transfer and the concentration profiles follow the usual equations for diffusion into a quiescent medium with a constant surface concentration of $c^*(\theta)$ [Equations (15), (17), and (19)]. As compared with the isothermal diffusion equations

$$[f_m(\theta)]_{\text{Isothermal}} = c^*(0)\sqrt{D/\pi\theta} \tag{32a}$$

and

$$[c]_{\text{Isothermal}} = c^*(0) \operatorname{erfc}(x/2\sqrt{D\theta}) \tag{32b}$$

the mass transfer rate and concentration profiles are modified by the factor K_1. Since K_1 is usually less than 1 [Equation (14)], the heat of solution accompanying absorption reduces the surface concentration and the rate of mass transfer.

The rate of heat transfer obtained in the second approximation [Equation (31)] is K_2 times greater than the rate which would be obtained if the interfacial temperature were $t^*(\theta)$ and there were no flow where

$$K_2 = [(1 + \operatorname{erf}\beta') \exp(\beta'^2)]^{-1} \tag{33}$$

This factor is a function of β' which is directly proportional to the velocity of the liquid phase or to the rate of

Table 1. Physical Constants

D, 22.2°C.	2.30×10^{-5} sq. cm./sec.
D, 40°C.	3.55×10^{-5} sq. cm./sec.
ΔH_s	7.63×10^3 cal./g.-mole NH_3
k (water)	1.5×10^{-3} cal./°C.-cm.-sec.
c_p (water)	1 cal./g.-°C.
ρ (water)	1 g./cc.
ρ_m (ammonia)	0.04 g.-mole/cc.
α (water)	1.5×10^{-3} sq. cm./sec.

volume change of the liquid phase. It is equivalent to the solution obtained by Knuth (7) for the case of a pure condensing vapor and is less than 1.

The rate of mass transfer obtained in the second approximation [Equation (30)] is K_3 times the rate which would be obtained if the interfacial concentration were $c^*(0)$ and there were no flow where

$$K_3 = \left[\frac{c^*(\theta)}{c^*(0)}\right]\left[\frac{1}{[1+\operatorname{erf}\beta]\exp(\beta^2)}\right]\left[\frac{1}{1-c^*(\theta)/\rho_m}\right] \quad (34)$$

K_3 is made up of three factors. The first factor $c^*(\theta)/c^*(0)$ represents the heat effect and can be evaluated from a rearrangement of Equation (27):

$$\frac{c^*(\theta)}{c^*(0)} = 1 + a\,\Delta H_s\,\beta\sqrt{\frac{\pi D}{c_p\,\rho\,k}}\,\frac{\rho_m}{c^*(0)} \quad (27a)$$

From this equation it can be shown that the ratio $c^*(\theta)/c^*(0)$ is usually less than unity because the quantity "$a\Delta H_s$" usually takes on negative values. Once again it is found that the heat effect tends to decrease the surface concentration and the rate of transfer.

The second factor is similar to K_2 which has been seen to arise out of the flow or volume change. It is less than 1 because the flow flattens the profiles and decreases the transfer rates.

The third factor also depends upon the flow, but it only modifies the diffusion [It also indirectly affects the heat transfer through its effect on $t^*(\theta)$.] It may be considered to arise from the fact that the dissolved gas is diffusing through a liquid which is immobile at the interface. Since this factor is greater than 1, it increases the rate of mass transfer.

The product of the second and third factors is analogous to the factor obtained by Arnold (1) for isothermal diffusion of one gas into a second gas which is immobile at the interface. Since both factors in this product depend upon the flow, the product may be considered to represent the net convective effect which is inseparable from the volume change of the liquid phase. The product may be written as follows:

$$\left[\frac{1}{[1+\operatorname{erf}\beta]\exp(\beta^2)}\right]\left[\frac{1}{1-c^*(\theta)/\rho_m}\right] = \frac{1}{[1+\operatorname{erf}\beta]\exp(\beta^2)} + \beta\sqrt{\pi} \quad (35)$$

It is greater than unity for all positive values of β, so the net effect of the convection or volume change is to increase the rate of mass transfer which is opposite to the heat effect. Because of this no general inference can be drawn as to the value of K_3 without knowledge of the relative magnitudes of the various quantities. Hence the overall effect on the rate of mass transfer due to the heat of solution and the volume change of the liquid phase can only be evaluated for individual cases when physical properties and transport coefficients of given systems are known.

ABSORPTION OF AMMONIA BY WATER

As an illustration the above results are applied to the absorption of ammonia by water.

Experimental data (3) for the absorption of ammonia gas saturated with water by a laminar water jet at 22.2°C. and 740 mm. Hg are used. These data were obtained at contact times ranging from 0.0009 to 0.02 sec. and are corrected for length effects (3). The penetration of the gas into the liquid phase was very shallow. Consequently the liquid phase may be considered to be semi-infinite and the jet diameter constant.

The heat of solution data of this system are available (6). The reported data are believed to include the heat of reaction as the ammonia will form ammonium hydroxide and dissociate in aqueous solution. There is no data available for the rate of reaction and dissociation, so it is necessary to consider the heat of solution and the heat of reaction as a whole and to assume that they are released at the liquid surface as an instantaneous heat source, as was assumed in the earlier derivations.

The constants in Equation (6), which relates the surface concentration and surface temperature, are determined from the data reported by Sherwood (8):

$$c^*(\theta) = 1.78 \times 10^{-2} - 3.0 \times 10^{-4}\,t^*(\theta) \quad (6a)$$

where

$$c^*(0) = 1.78 \times 10^{-2} \text{ g.-mole/cc.}$$

$$a = -3.0 \times 10^{-4} \text{ g.-mole/cc.} - {}^\circ\text{C.}$$

$$t^*(\theta) = ({}^\circ\text{C.} - 22.2^\circ\text{C.})$$

Other physical constants used in the calculation are listed in Table 1. It is seen that the values of k, ρ, and c_p are that of pure water. If k, ρ, and c_p are taken as the mixture values at the surface temperature and concentration, there is no significant change in the results. The diffusivity data given in the table are extrapolated values based on low temperature experimental data and Wilke's generalized correlation (9).

The mean rate of absorption has been calculated for three cases:

1. Absorption under isothermal conditions at 22.2°C. with Equation (32) used.
2. Absorption accompanied by heat of solution with Equations (15), (17), and (18) used.
3. Absorption accompanied by both heat of solution and volume change of the liquid phase with Equations (25), (27), and (30) used.

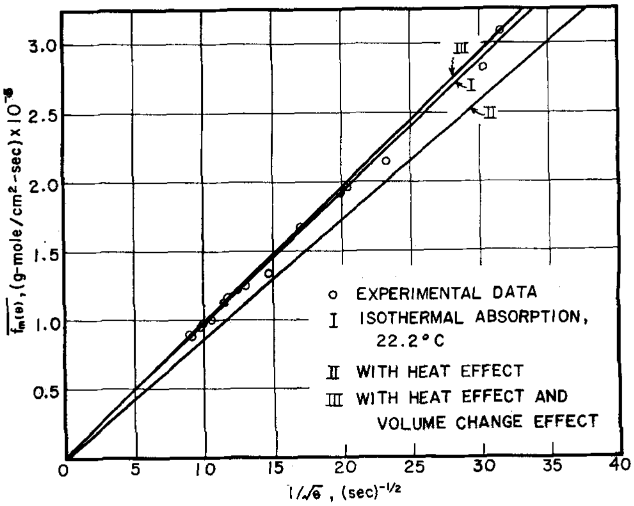

Fig. 2. Rate of absorption of ammonia into water.

Table 2. Calculated Results

	$t^*(\theta)$ (°C. − 22.2)	$c^*(\theta)$ $\frac{\text{g.-mole NH}_3}{\text{cc. soln.}}$	$f_m(\theta)\sqrt{\theta}$ $\frac{\text{g.-mole NH}_3}{\text{sq. cm.} - \sqrt{\text{sec.}}}$	$\overline{f_m(\theta)}\sqrt{\theta}$† $\frac{\text{g.-mole NH}_3}{\text{sq. cm.} - \sqrt{\text{sec.}}}$
Case I Isothermal, 22.2°C. $D = 2.3 \times 10^{-5}$ sq. cm./sec.	0	1.78×10^{-2}	4.83×10^{-5}	9.65×10^{-5}
Case II With heat effect $D = 3.3 \times 10^{-5}$ sq. cm./sec. $K_1 = 0.746$	15.0	1.33×10^{-2}	4.30×10^{-5}	8.60×10^{-5}
Case III With heat effect and volume change effect $D = 3.5 \times 10^{-5}$ sq. cm./sec. $\beta = 0.208$ $\beta' = 0.0316$	17.7	1.25×10^{-2}	4.90×10^{-5}	9.80×10^{-5}

† $\overline{f_m(\theta)} = \left[\int_0^\theta f_m(\theta)d\theta\right] / \theta = 2f_m(\theta)$.

The calculated results are tabulated in Table 2 and are also shown in Figure 2 together with experimental data.

In Figure 2 it is seen that the experimental data are seemingly in good agreement with both curve I and curve III, while curve II lies about 12% below the data. The agreement between curve I, representing the isothermal case, and the experimental result is certainly unexpected. It is a well-known fact that the absorption of ammonia into water will not proceed under isothermal condition because the process is highly exothermic in nature. In fact the calculations show that the surface temperature is more than 17°C. greater than the initial temperature.* The reason that the experimental data lie close to the isothermal curve is simply because two opposing factors, the heat effect and the volume change effect, almost exactly cancel each other as indicated by curve III. Thus the apparent agreement between curve I and the data must be considered fortuitous. The experimental results should actually be represented by curve III which accounts for both the heat effect and the volume change effect.

The agreement between the experimental data and the theoretical prediction demonstrated the usefulness of the second approximation. It is also important to point out some of the implications of the analysis. First from the practical point of view the ammonia data seem to indicate that the isothermal rate equation could be used to advantage in estimating the rate of transfer even for systems with a large heat effect, because such a heat effect is likely to be compensated to a large extent by the accompanying convective effect. Secondly from the theoretical point of view the present analysis has shown that results based on rate measurements alone can be misleading, especially if they are used in interpreting the mechanism of the transfer process. The temperature of the interface may be many degrees above the bulk temperature, yet the rate of transfer may remain practically unchanged from the isothermal value. Phenomena such as this will be rather difficult to detect by the usual rate measurements.

ACKNOWLEDGMENT

This work was supported in part by the National Science Foundation.

NOTATION

a = constant
c = concentration
$c^*(0)$ = surface concentration, at $\theta = 0$
$c^*(\theta)$ = surface concentration, at $\theta = \theta$
c_p = specific heat
D = diffusivity measured in volume reference frame
$f_h(\theta)$ = rate of heat transfer per unit area at interface
$f_m(\theta)$ = rate of mass transfer per unit area at interface
ΔH_s = heat of solution
k = thermal conductivity
K_1 = constant
K_2 = constant
K_3 = constant
t = temperature
$t^*(\theta)$ = surface temperature, at $\theta = \theta$
$v(\theta)$ = volumetric average velocity of the liquid phase
x = distance in the direction of transfer
y = dimensionless distance, $y = x/2\sqrt{D\theta}$
y' = dimensionless distance, $y' = x/2\sqrt{\alpha\theta}$

Greek Letters

α = thermal diffusivity
β = dimensionless velocity, $\beta = v(\theta)/\sqrt{D/\theta}$
β' = dimensionless velocity, $\beta' = v(\theta)/\sqrt{\alpha/\theta}$
θ = time
ρ = density of liquid
ρ_m = molal density of dissolved gas

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Manuscript received April 1, 1963; revision received November 1, 1963; paper accepted November 4, 1963. Paper presented at A.I.Ch.E. San Juan meeting.

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* A recent experimental study of the temperature of the liquid surface for the ammonia-water system has confirmed the predicted surface temperature rise of about 17°C. The details of such a study will be reported in a future paper.