Gas Absorption with Exothermic Bimolecular (1,1 Order) Reaction

Gas absorption with a nonisothermal, bimolecular (1,1 order), and irreversible reaction are analyzed and approximate film theory solutions are obtained for interfacial temperature rise and enhancement factor. The model of Mann and Moyes is extended to incorporate depletion of liquid reactant concentration at the gas-liquid interface and also the variation of dissolved gas concentration in the bulk liquid, leading to a generalized solution independent of the regime of absorption. The effect of heat generation, heat dissipation, and reaction rate parameters on enhancement factor and interfacial temperature rise is discussed. The system showed multiplicity behavior under certain conditions, but at higher values of heat dissipation parameters and for some combinations of heat generation and reaction rate parameters, unique solutions were obtained. However, even the unique solutions of enhancement factor are substantially different from the values obtained under isothermal conditions. Since depletion of liquid reactant concentration is incorporated, asymptotic values of enhancement factor (at large $\sqrt{M}$) were observed to be the same as those obtained in the instantaneous reaction regime of isothermal absorption and reaction process. The model presented does not require *a priori* assumption of the regime of absorption and hence can be useful in modeling nonisothermal reactors.

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Introduction

Gas absorption with exothermic reaction in a liquid is encountered in several practical cases, some examples of which are found in chlorination of organic compounds (Ding et al., 1974; Mann and Clegg, 1975), reaction of sulfur trioxide with dodecylbenzene (Mann and Moyes, 1977), and liquid phase catalytic oxidation of hydrocarbons (Carra and Santacesaria, 1980). In these problems, heat is released due to dissolution of the gas as well as to the exothermic chemical reaction, leading to an increase in the temperature at the gas-liquid interface. This in turn has an influence on the rate of absorption. In the analysis of such a problem, heat and mass transfer with chemical reaction must be considered simultaneously.

Danckwerts (1951) presented approximate analytical expressions for the interfacial temperature rise based on the film, penetration, and surface renewal theories. Shah (1972) analyzed the same problem based on the penetration theory with an exponential dependence of the physical, transport, and reaction properties on temperature. Mann and Moyes (1977) reported film theory solutions for the interfacial temperature rise and enhancement factor for the case of a nonisothermal pseudofirst-order reaction and compared the results with experiments on the SO_3-dodecylbenzene system. Further, Allan and Mann (1982) have analyzed the steady-state multiplicity of the enhancement factor and interfacial temperature rise for the same case. White and Johns (1985), using the model of Allan and Mann, have shown that five solutions for interfacial temperature can exist for some parameter combinations. Nielsen and Villadsen (1983, 1985) have analyzed a case of nonisothermal gas absorption accompanied with a pseudofirst-order reaction in a falling film column. They have mainly discussed the multiplicity aspects of the temperature rise at the gas-liquid interface. The problem of nonisothermal gas absorption followed by an instantaneous reaction has been analyzed by Asai et al. (1985). In general, it is well understood that the increase in interfacial temperature

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reduces the solubility but increases the rate of chemical reaction, and hence the rate of absorption is a result of the combination of these two effects. However, in most of the earlier reports a fast pseudofirst-order reaction regime of absorption has been assumed. But the increase in interfacial temperature is likely to result in depletion of the liquid phase reactant concentration at the gas-liquid interface, under certain conditions. This important aspect has not been incorporated in any of the previous works.

It is the objective of this paper to present a general solution for the interfacial temperature rise and enhancement factor for gas absorption with a bimolecular (1,1 order) reaction, incorporating depletion of the liquid phase reactant concentration at the gas-liquid interface. This analysis is based on the model of Mann and Moyes (1977) but does not require *a priori* assumption of the regime of absorption. The effect of various parameters on the enhancement factor, interfacial temperature rise, and the multiplicity behavior of the system is discussed.

Theory

Let us consider a problem of absorption of a gaseous reactant A, followed by a 1,1 order irreversible and exothermic chemical reaction with the liquid phase reactant B, according to the following stoichiometry: $A(g) + zB(L) \longrightarrow$ Products.

In order to obtain solutions for the interfacial temperature rise and the enhancement factor, the following assumptions have been made:

1. Two-film theory is applicable
2. Gas-side mass transfer resistance is negligible
3. The liquid phase reactant and solvent are nonvolatile
4. Physical properties of the liquid and gas such as diffusivity, density, heat capacity, and thermal conductivity are unaffected by the changes in temperature and composition of the liquid phase
5. Volume change due to absorption, Dufour, and Soret effects is negligible
6. Semibatch operation with no bulk liquid flow is assumed
7. The temperature dependence of the solubility and reaction rate constant can be expressed as:

$$A^*(T^*) = A^*(T_b) \exp\left[-\frac{\Delta H_S}{R}\left(\frac{1}{T^*} - \frac{1}{T_b}\right)\right] \tag{1}$$

and

$$k_2(T^*) = k_2(T_b) \exp\left[-\frac{E}{R}\left(\frac{1}{T^*} - \frac{1}{T_b}\right)\right] \tag{2}$$

A diagram of the film model with concentration and temperature profiles is shown in Figure 1. In what follows, the model of nonisothermal gas absorption in a reactive liquid, as proposed by Mann and Moyes (1977), is extended and generalized to a case that involves an exothermic bimolecular reaction. Referring to Figure 1, we see that there is as usual a mass transfer film of thickness σ_M over which concentration gradients exist. However, due to a large difference in the magnitudes of mass and thermal diffusivities ($\alpha = 100\, D$), one can conceive of a so-called hypothetical heat transfer film thickness, σ_H, over which the temperature gradient would exist, and for obvious reasons $\sigma_H \gg \sigma_M$. Thus the temperature over the mass transfer film can be assumed to be constant. This well-understood approximation to the physical reality of the problem thus allows us to uncouple the mass and heat balance equations for the mass transfer film and solve the following isothermal mass balance equations at a tem-

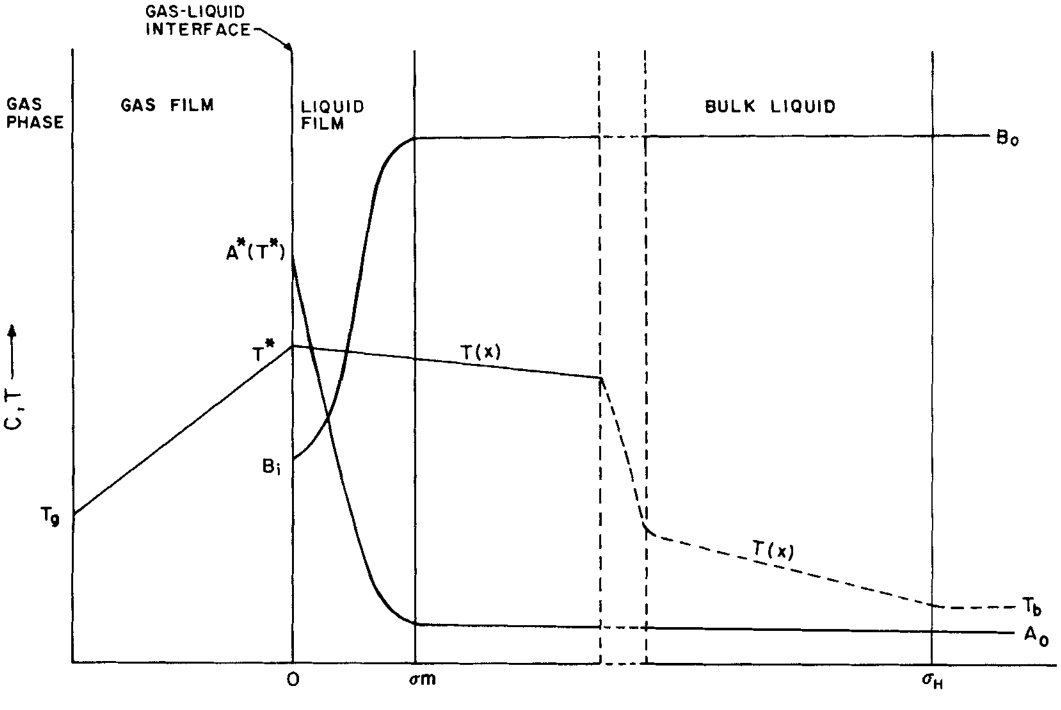

Figure 1. Diagram of film model for nonisothermal gas absorption with reaction.

perature corresponding to that at the gas-liquid interface.

$$D_A \frac{d^2A}{dx^2} = k_2(T^*)AB \tag{3}$$

$$D_B \frac{d^2B}{dx^2} = zk_2(T^*)AB \tag{4}$$

where T^* is the unknown interfacial temperature and is to be determined. The boundary conditions at the gas-liquid interface can be written as:

$$x = 0, \quad A = A^*(T^*), \frac{dB}{dx} = 0 \tag{5}$$

It is in the postulation of the boundary conditions at $x = \sigma_M$ and the interpretation of the underlying physical situation that our model deviates from and, indeed, generalizes the model proposed by Mann and Moyes. The analysis here has been applied to a semibatch operation for simplicity. However, it can easily be extended to a continuous stirred-tank reactor or any other continuous reactor. The boundary condition at $x = \sigma_M$ can, in general, be written as:

$$x = \sigma_M, \quad A = A_o, \quad B = B_o \tag{6}$$

in which B_o is known but A_o, in general, is not. In all the previous studies, including that of Mann and Moyes (1977), $A_o = 0$ was *a priori* assumed. In our opinion this can be done only in specific cases where the regime of absorption even at the datum temperature condition is known to be fast. It is more common, however, to encounter situations where the regime of absorption at the datum conditions is slow, but depending upon the nonisothermal effects due to absorption and reaction, the effective regime of absorption may or may not shift to the fast regime. Finally, in a nonisothermal absorption-reaction system multiple steady states are known to occur; therefore, by arbitrarily fixing the bulk concentration $A_o = 0$, one may be led to suppression of some of the possible steady states.

In order to avoid *a priori* assignment of a value to A_o, one normally writes a bulk-side mass balance (such as written by Hofmann et al., 1975) which for the case of a semibatch reactor, as considered here, is given by:

$$-aD_A \frac{dA}{dx}\bigg|_{x=\sigma_M} = r_A(T, A_o, B_o) \tag{7}$$

There is a minor difficulty associated with evaluation of the rate expression on the righthand side of Eq. 7. According to the film model used in this work, beyond $x = \sigma_M$, while the concentrations are at their bulk values, a temperature gradient exists between σ_M and σ_H. However, the physical reality of the situation allows us a reasonable approximation for r_A, the rate of reaction in the bulk liquid beyond σ_M. It is obvious that the volume of the zone between σ_M and σ_H in which the temperature gradient may exist is a very small fraction of the total bulk liquid volume. Therefore, the reaction rate in the entire bulk liquid beyond σ_M can be approximated as $(\epsilon_L - a\sigma_M)k_2(T_b)A_oB_o$. It is reasonable to assume that the contribution of the temperature gradient in the zone between σ_M and σ_H to the total rate in the bulk liquid beyond σ_M would be negligible. Also, when $T^* \gg T_b$, the concentration of A (the limiting reactant in this case) in the bulk, i.e., A_o, would tend to zero and the contribution of the bulk reaction rate beyond σ_M to the overall rate of absorption would be negligible. On the other hand, it should be noted that the contribution of the boundary condition, Eq. 7, to the overall rate of absorption would be significant only when A_o is finite. But when A_o is finite, normally the interfacial temperature rise is rather small and hence the above approximation in the boundary condition is justified under such conditions. The boundary condition, Eq. 7, can then be approximated as:

$$x = \sigma_M, \quad -aD_A \frac{dA}{dx} = (\epsilon_L - a\sigma_M)k_2(T_b)A_oB_o \tag{8}$$

The interfacial temperature T^* can be related to the bulk gas and liquid phase temperatures, T_g and T_b, via an overall heat balance as:

$$(-\Delta H_S)\left(-D_A \frac{dA}{dx}\bigg|_{x=0}\right) + (-\Delta H_R)\left[-D_A \frac{dA}{dx}\bigg|_{x=0} - \left(-D_A \frac{dA}{dx}\bigg|_{x=\sigma_M}\right)\right] = h_L(T^* - T_b) + h_g(T^* - T_g) \tag{9}$$

The basis of the above heat balance equation is that the entire heat evolved due to both dissolution of A and the reaction within the mass transfer film is considered to be dissipated to the bulk gas and liquid phases. The heat dissipation processes are characterized through lumped heat transfer coefficients.

The complete formulation of the problem then consists of differential Eqs. 3 and 4 subject to the boundary conditions given by Eqs. 5, 6, and 8, with the unknown interfacial temperature being determined from Eq. 9. The model equations can be solved using the approximation suggested by Hikita and Asai (1964), in which, in Eqs. 3 and 4, B_i has been substituted for B. Using Eqs. 3 and 4, and the boundary conditions of Eqs. 5 and 6, the following relationship between the interfacial concentration of the liquid phase reactant b_i $(=B_i/B_o)$, the enhancement factor E, and θ^* $(=T^*/T_b)$ can be obtained:

$$b_i = 1 + \frac{1}{sq}[(a^* - a_o) - E_1] \tag{10}$$

From solution of Eq. 3 using boundary conditions 5, 6, and 8, the following expressions can be derived:

$$-D_A \frac{dA}{dx}\bigg|_{x=0} = \frac{k_LA^*(T_b)\sqrt{M_1}}{\tanh\sqrt{M_1}}\left(a^* - \frac{a_o}{\cosh\sqrt{M_1}}\right) \tag{11}$$

and

$$-D_A \frac{dA}{dx}\bigg|_{x=\sigma_M} = \frac{k_LA^*(T_b)\sqrt{M_1}}{\tanh\sqrt{M_1}}\left(\frac{a^*}{\cosh\sqrt{M_1}} - a_o\right) \tag{12}$$

Using Eq. 12 and the boundary condition of Eq. 8 and rearranging, we obtain

$$a_o = \frac{a^* \sqrt{M_1}}{\sinh \sqrt{M_1} \left(\frac{M}{\beta} + \frac{\sqrt{M_1}}{\tanh \sqrt{M_1}} \right)} \tag{13}$$

Thus, the bulk liquid concentration of A need not be *a priori* assumed, as has been done in all the previous studies (Mann and Moyes, 1977; White and Johns, 1985) and can be evaluated by Eq. 13 within the computational procedure developed in this work. Substituting for the fluxes of A at $x = 0$ and $x = \sigma_M$ in Eq. 9, from Eqs. 11 and 12 we obtain the relationship of θ^* in terms of other parameters:

$$\theta^* = \frac{(G_S + G_R)E_1 - G_R E_2 + G_1 + G_2 \theta_g}{(G_1 + G_2)} \tag{14}$$

The calculation procedure involves Eqs. 10–14, which compute b_i, a_o, E_1, E_2, and θ^* in a sequence of trial and error until convergence. This iterative algorithm is easily programmed on a digital computer.

Since the present model is known to provide multiple steady-state solutions for θ^* as well as for E_1, we have made careful computations, for identical parameter sets, with different initial guess values of θ^*. This has allowed us to obtain multiple steady-state solutions wherever they can be expected to exist.

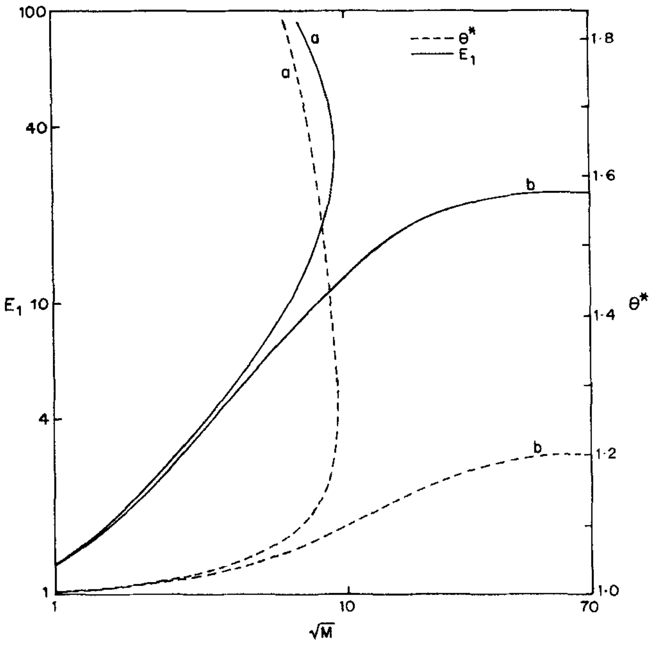

Figure 2. Comparison of E_1 vs. $\sqrt{M}$ and θ^* vs. $\sqrt{M}$ results.

a. Assuming $Bi = B_o$ and $A_o = 0$
b. No prior assumption
$G_E = 42$; $G_R = 38$; $G_S = 15$; $G_1 = 6{,}550$; $G_2 = 0$

Results and Discussion

Using the numerical procedure described above, extensive computations for interfacial temperature rise and enhancement factor were made. The results have been presented in Figures 2–6 as E_i vs. $\sqrt{M}$ and θ^* vs. $\sqrt{M}$ plots in order to demonstrate the effect of parameters G_R, G_E, G_S, and G_1. These are the main parameters describing the nonisothermal effects in gas absorption with exothermic reaction.

The E_1 vs. $\sqrt{M}$ and θ^* vs. $\sqrt{M}$ behavior in the case of a nonisothermal gas absorption with reaction, is controlled by the combined influence of a decrease in the solubility and an increase in the reaction rate with increase in temperature. These trends are further modulated depending on the heat generated (controlled by G_R and G_S) and heat dissipated either to the liquid bulk or to the gas phase, or to both sides (controlled by G_1 and/or G_2). It should be also noted that for gas absorption with a bimolecular reaction, increase in interfacial temperature can lead to a depletion of liquid phase reactant concentration at the gas-liquid interface. Similarly, under certain conditions the bulk concentration of the gaseous reactant can be finite. Therefore, *a priori* assumption of a pseudofirst-order reaction ($b_i = 1$) as well as zero bulk concentration of $A (A_o = 0)$ can in general lead to erroneous results. The generalized analysis presented in this paper allows, for the first time, prediction of nonisothermal effects on the enhancement factor and interfacial temperature rise for the case of a bimolecular gas-liquid reaction incorporating all the above parameters and without making *a priori* assumption of the regime of absorption.

The results of θ^* vs. $\sqrt{M}$ and E_1 vs. $\sqrt{M}$ are shown in Figure 2 for two cases: in case *a* we assume $B_i = B_o$ and $A_o = 0$; in case *b* no such assumption is made. Here, a remarkable difference was observed between the two cases. Under these conditions, the increase in the interfacial temperature leads to substantial depletion of the liquid phase reactant (B) concentration at the gas-liquid interface, and both the enhancement factor and the interfacial temperature rise approach their asymptotic values. These results clearly show that an arbitrary assumption (as implied by case *a*) can lead to wrong results not only quantitatively but also qualitatively. The above comparisons have demonstrated a need for incorporating the depletion of liquid reactant concentration at the gas-liquid interface in evaluating the enhancement factor and interfacial temperature rise for a nonisothermal gas-liquid reaction.

In order to understand the effect of parameters G_E and G_S and the general features of the nonisothermal gas-liquid reactions, E_1 vs. $\sqrt{M}$ and θ^* vs. $\sqrt{M}$ plots were generated, using the procedure described earlier, for $G_1 = 6.55 \times 10^3$ and $G_2 = 0$. The results are shown in Figures 3 and 4. The parameter $G_2 = 0$ implies no heat dissipation to the gas phase, and therefore the θ^* values in Figure 4 represent the upper bound possible, for the given set of parameters. The following important observations can be made from these results.

1. The E_1 vs. $\sqrt{M}$ plot tends to reach an asymptotic value that is approximately the same as the isothermal asymptote. This region indicates an instantaneous reaction regime. It can be shown as follows that the asymptotic value of E_1 (as defined in this work) for the nonisothermal case would be the same as that for isothermal case for $q \gg 1$.

In the instantaneous regime we have,

$$R_A = k_L A^*(T^*) \left(1 + \frac{B_o}{z A^*(T^*)} \right), \quad \text{for } D_A = D_B \tag{15}$$

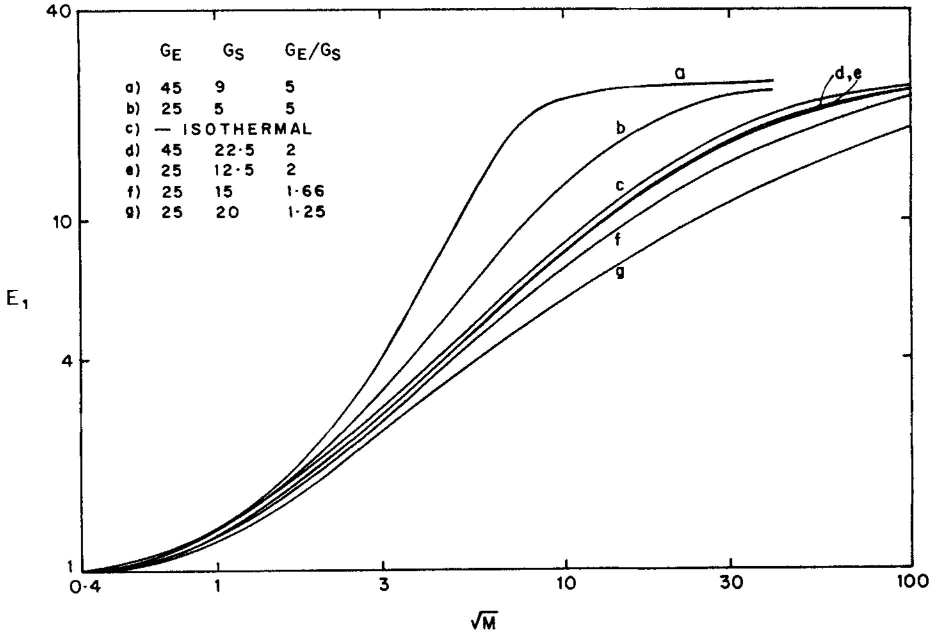

Figure 3. Effect of $G_E - G_S$ combinations on E_1 vs. $\sqrt{M}$ plots.
$G_R = 38$; $G_1 = 6{,}550$; $G_2 = 0$

Then,

$$E_1 = \frac{R_A}{k_L A^*(T_b)} = \frac{A^*(T^*)}{A^*(T_b)}\left[1 + \frac{B_o}{zA^*(T^*)}\right] \tag{16}$$

$$\simeq \frac{B_o}{zA^*(T_b)} = q \tag{17}$$

2. For $G_E/G_S = 2$, the E_1 vs. $\sqrt{M}$ plot is in close agreement with that of an isothermal case, Figure 3. This can also be realized from Eq. 11, which indicates that in this situation the decrease in solubility and the increase in rate constant with increase in the interfacial temperature mutually compensate each other, although the corresponding θ^* vs. $\sqrt{M}$ plot, Figure 4, indicates a substantial increase in interfacial temperature.

3. For G_E/G_S values other than 2, the nonisothermal E_1 vs. $\sqrt{M}$ results are distinctly different in comparison with the isothermal case except for the asymptotic region. Also, the results in Figures 3 and 4 show that the particular combination of G_E and G_S (not the G_E/G_S ratio) decides the E_1 and θ^* values. At lower G_E/G_S values, for a certain combination of G_E and G_S—

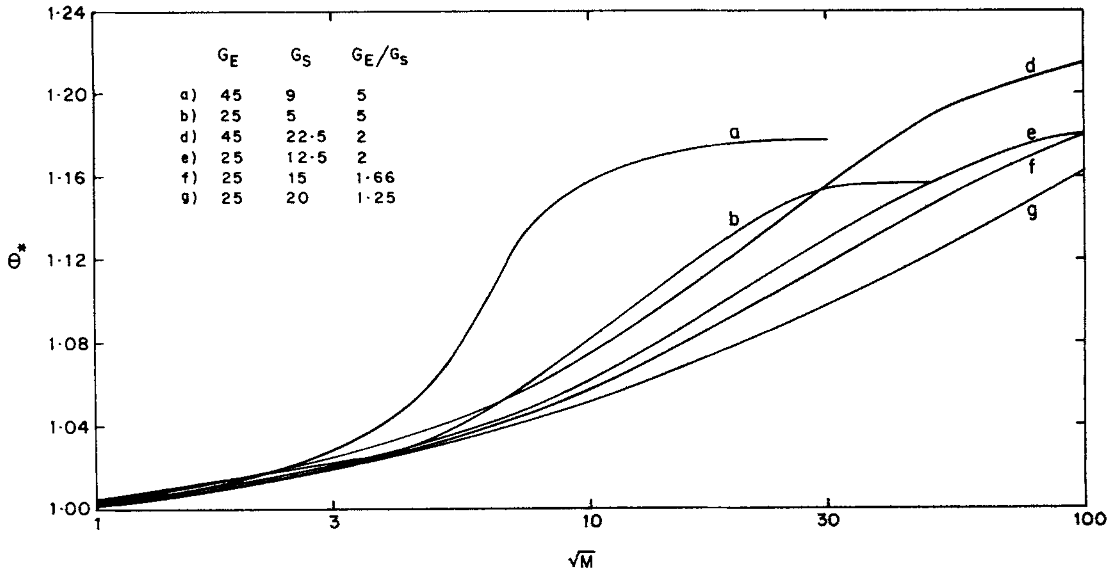

Figure 4. Effect of $G_E - G_S$ combinations on θ^* vs. $\sqrt{M}$ plots.
$G_R = 38$; $G_1 = 6{,}550$; $G_2 = 0$

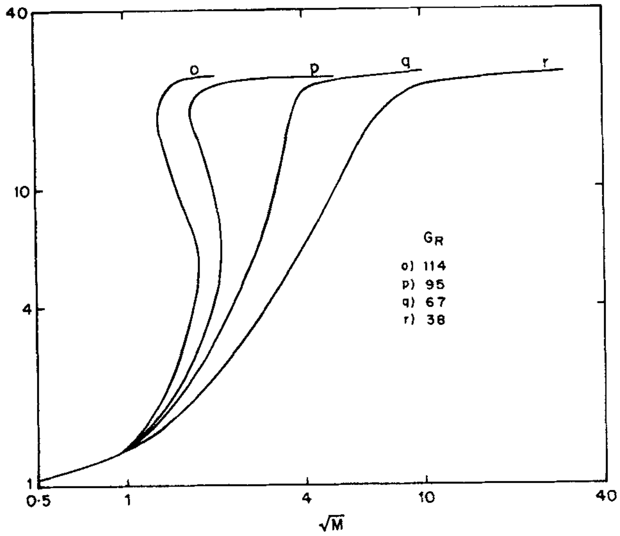

Figure 5. Effect of G_R on E_1 vs. $\sqrt{M}$ plots.
$G_E = 45; G_S = 9; G_1 = 6{,}550; G_2 = 0$

curves *f* and *g* in Figure 3—the E_1 values are even lower than for the isothermal case. In this situation, the reduction in solubility due to interfacial temperature rise is more significant compared to the increase in the rate of reaction. On the other hand, the results shown in curves *a* and *b*, Figure 3, represent situations where increase in rate constant is more significant compared to the solubility reduction.

The effect of parameter G_R on E_1 vs. $\sqrt{M}$ behavior is shown in Figure 5. A plot on the effect of G_R on $\theta^* - \sqrt{M}$ behavior is presented as supplementary material. An interesting feature observed in this case is the multiplicity of steady-state values of E_1 as well as θ^* at higher G_R values. The range of G_R covered in these calculations is based on the practical range of $(-\Delta H_R)$ values for some important gas-liquid reactions, such as oxidation of hydrocarbons in the liquid phase. For such a case, in which increase in the rate constant predominated over decrease in the solubility with increase in the interfacial temperature, multiplicity behavior is shown for the first time in this paper. Here, the depletion of the liquid phase reactant (B) concentration at the gas-liquid interface is significant and the earlier models based on a fast pseudofirst-order reaction regime cannot be used. It may also be noted that despite the observed turning points in $E_1 - \sqrt{M}$ and $\theta^* - \sqrt{M}$ plots, the correct asymptotic values of E_1 and θ^* at large $\sqrt{M}$ are always reached.

The parameter G_1 controls the extent of heat dissipation from mass transfer film to the bulk liquid. At lower G_1 values, the heat evolved due to dissolution and reaction would tend to accumulate within the film, resulting in an increase in θ^*. The effect of G_1 on E_1 vs. $\sqrt{M}$ is shown in Figure 6. (A plot showing the effect of G_1 on θ^* vs. $\sqrt{M}$ is presented as supplementary material.) As observed in other cases, here too either unique or multiple solutions of θ^* and E_1 were obtained, depending on the parameter combinations. As G_1 is increased, θ^* is reduced and unique solutions for E_1 and θ^* are obtained. At low G_1 values, θ^* observed are too high; in most practical cases this situation will not exist. However, the model proposed here does not incorporate vaporization of the solvent or liquid reactant; hence the results at low G_1 are mainly indicative of the trend in which G_1 will influence θ^* and E_1. It may, however, be noted that in all cases the asymptotic E_1 values obtained are the same as would be expected.

Conclusions

A problem of gas absorption with nonisothermal bimolecular (1,1 order) reaction has been analyzed and a procedure for calculation of interfacial temperature rise and enhancement factor has been discussed. For the first time, a general case independent of regime of absorption has been analyzed incorporating

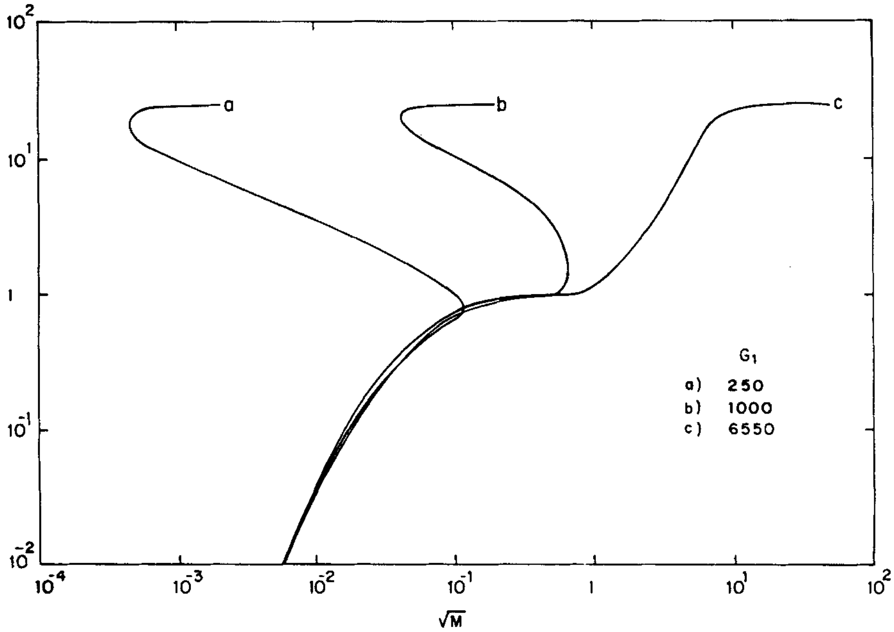

Figure 6. Effect of G_1 on E_1 vs. $\sqrt{M}$ plots.
$G_E = 45; G_R = 38; G_S = 7.8; G_2 = 0$

the depletion of liquid phase reactant concentration at the gas-liquid interface as well as the variation in the bulk liquid concentration of dissolved gas. Multiple solutions for θ^* and E_1 are observed for certain conditions, while unique solutions prevail under other conditions.

The multiplicity behavior was found to disappear with increase in heat dissipation parameter G_1. Similarly, the multiplicity pattern was also found to be sensitive to parameters G_2, G_S, G_R, and G_E. An important implication of the multiplicity behavior is that, depending on the initial conditions, the system can attain different steady states which may be in altogether different regimes of absorption. In each case, however, the asymptotic value of E_1 (at large $\sqrt{M}$) is the same as that obtained in the instantaneous reaction regime of the isothermal absorption process irrespective of the set of parameters used.

Notation

A = concentration of dissolved gas A
A_o = concentration of A in bulk liquid
$A^*(T^*)$ = interface concentration of A at temperature T^*
$A^*(T_b)$ = interface concentration of A at temperature T_b
a = interfacial area per unit volume
a_o = concentration of dissolved gas, $A_o/A^*(T_b)$
a^* = interface concentration of A, $A^*(T^*)/A^*(T_b)$
B = concentration of reactant B
B_o = concentration of reactant B in bulk liquid
B_i = interface concentration of reactant B
b_i = interface concentration of $B, B_i/B_o$
C_p = specific heat of liquid
D_A = diffusivity of A
D_B = diffusivity of B
E = activation energy
$E_1 = [-D_A(dA/dx)_{x=0}]/k_LA^*(T_b)$
$E_2 = [-D_A(dA/dx)_{x=\sigma_M}]/k_LA^*(T_b)$
G_1 = parameter, $h_L/Rk_LA^*(T_b)$
G_2 = parameter, $h_g/Rk_LA^*(T_b)$
G_E = activation energy parameter, E/RT_b
G_R = heat of reaction parameter, $-\Delta H_R/RT_b$
G_S = heat of solution parameter, $-\Delta H_S/RT_b$
h_g = gas phase heat transfer coefficient
h_L = liquid phase heat transfer coefficient
K = reaction rate constant, $K_2(T^*)/k_2(T_b)$
$k_2(T)$ = reaction rate constant at temperature T
$k_2(T^*)$ = reaction rate constant at temperature T^*
$k_2(T_b)$ = reaction rate constant at temperature T_b
k_L = liquid phase mass transfer coefficient
M = parameter, $k_2(T_b)D_AB_o/k_L^2$
M_1 = parameter, $k_2(T^*)D_AB_i/k_L^2$
q = concentration ratio, $B_o/zA^*(T_b)$
R = gas constant
r_A = bulk reaction rate
s = diffusivity ratio, D_B/D_A
T = temperature
T_b = bulk liquid temperature
T_g = bulk gas temperature
T^* = interface temperature
x = distance from interface into liquid
z = stoichiometric coefficient of B

Greek letters

α = thermal diffusivity
β = parameter, $a\sigma_M/(\epsilon_L - a\sigma_M)$
σ_H = thickness of heat transfer film
σ_M = thickness of mass transfer film
$-\Delta H_R$ = heat of reaction
$-\Delta H_S$ = heat of solution
ϵ_L = liquid holdup
θ^* = interface temperature, T^*/T_b
θ_g = gas phase temperature, T_g/T_b

Subscripts

A = species A
B = species B
b = bulk
E = activation
g = gas phase
i = at interface
L = liquid phase
R = reaction
S = solution

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