

Penetration Theory for Gas Absorption Accompanied by a Second Order Chemical Reaction

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The penetration-theory solution for the effect of a second-order irreversible liquid-phase chemical reaction on the rate of gas absorption has been computed numerically on an IBM-704 computer. A linearized, time-centered, implicit finite-difference method was used to solve the nonlinear partial-differential equations. The method was very effective, permitting the solution of the equations for a wide range of the parameters of interest. The penetration-theory results are compared with the film-theory solution, and it is shown that the solutions to the two theories agree within 16% if they are compared for conditions which produce the same asymptotic solution for an infinitely rapid chemical reaction. A simplified equation and some correction charts are presented which permit a rapid, accurate estimation of the penetration-theory solution over a wide range of variables.

The problem of predicting the effect of a simultaneous liquid-phase chemical reaction on the rate of gas absorption has often been approached by adopting a simplified model of the liquid flow pattern which could then be treated mathematically. In the years since Hatta (11) first used the film-theory model to analyze the effect of an infinitely rapid, irreversible bi-molecular reaction, a number of types of chemical reaction have been treated from the viewpoints of the film-theory and the penetration-theory models. The use of these two models to analyze the effect of a simultaneous chemical reaction on the rate of gas absorption is reviewed by Sherwood and Pigford (18).

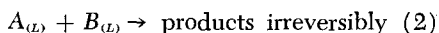
One interesting aspect of this problem is that it is often found that the predicted answer is surprisingly insensitive to the liquid flow pattern model that is chosen. This was pointed out by Peaceman (13), who compared the film- and penetration-theory solutions for several kinds of chemical reactions and found that they were in close agreement. Furthermore several recent publications have considered the effect of an infinitely rapid bimolecular reaction on the rate of mass transfer from a solid surface to a fluid stream, and results for laminar and turbulent boundary-layer models show surprising agreement with film and penetration theory results (3, 8, 15, 19).

It is the purpose of this paper to present a solution to the penetration-theory equations for gas absorption accompanied by a second-order chemical reaction of finite rate and to com-

pare this solution with the film-theory solution.

THE SECOND-ORDER REACTION

The problem to be considered is that in which a gaseous species A dissolves into the liquid phase [Equation (1)] and then reacts irreversibly with species B according to Equation (2):



Species B is a nonvolatile solute which has been dissolved into the liquid phase prior to its introduction into the gas absorber. It is assumed that gas-phase resistance to absorption is negligible, and thus the concentration of species A at the gas-liquid interface A_i corresponds to equilibrium with the partial pressure of species A in the bulk gas phase. The chemical reaction Equation (2) is assumed to be second-order, with the kinetic equation

$$= k_1 A B \quad (3)$$

PENETRATION THEORY

In the penetration-theory model of the liquid flow pattern it is assumed that the liquid flows down over a piece of absorber packing in slug, laminar flow. Absorption takes place by molecular diffusion and accumulation within a slug of liquid as it flows down the packing and is exposed to the gas phase for a given contact-time interval. The liquid is completely and instantaneously mixed as it goes from one piece of packing to the next; thus each slug begins each contact-time interval with flat concentration profiles for all components. The contact time between successive mixing points is so

short that the absorbing species never penetrates deeply enough to approach the wall of the piece of packing, and so the liquid depth is assumed to be infinite for mathematical simplicity.

When one uses this model and the second-order chemical reaction system described by Equations (1), (2), and (3), the following differential equations can be derived:

$$\frac{\partial^2 a}{\partial z^2} - \frac{\partial a}{\partial \theta} = ab \quad (4)$$

$$rq \frac{\partial^2 b}{\partial z^2} - q \frac{\partial b}{\partial \theta} = ab \quad (5)$$

The boundary conditions to be imposed on the simultaneous solution of these differential equations are

$$\begin{aligned} \text{At } \theta = 0, \text{ any } z > 0 \quad a &= 0 \\ b &= 1 \end{aligned} \quad (6)$$

$$\begin{aligned} \text{At } z = 0, \text{ any } \theta > 0 \quad a &= 1 \\ \frac{\partial b}{\partial z} &= 0 \end{aligned} \quad (7)$$

$$\begin{aligned} \text{At } z \rightarrow \infty, \text{ any } \theta \quad a &= 0 \\ b &= 1 \end{aligned} \quad (8)$$

These boundary conditions contain the assumption that chemical equilibrium prevails at the beginning of the contact-time interval; thus $a = 0$. Peaceman (13) has shown that the liquid holdup in a gas-absorption column is sufficient to insure that this assumption is a good one for cases of practical importance.

It can be seen by Equations (4) through (8) that if numerical values are assigned to the parameters r and q , then a and b are specified as unique functions of θ and z . The result desired in this study is

$$\phi \equiv k_L/k_L^*$$

Both of these gas absorption coefficients are based on the average absorption rate over the contact-time interval. The physical absorption coefficient is related to the contact time by

$$k_L^* = 2 \sqrt{\frac{D_A}{\pi t}} \quad (9)$$

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Once a and b have been determined as functions of θ and z , the variable ϕ can be determined for a particular contact time θ , by the following equation:

$$\phi = \sqrt{\frac{\pi}{4\theta}} \int_0^\infty a \, dz + \sqrt{\frac{\pi}{4\theta}} \int_0^\infty q(1-b) \, dz \quad (10)$$

The first integral in Equation (10) represents the total amount of species A in the liquid phase after a contact-time interval θ ; the second integral represents the disappearance of species B and thus the amount of species A which has been removed by chemical reaction.

Careful study of Equations (4) through (10) reveals that ϕ is uniquely specified when numerical values are assigned to r , q , and the dimensionless contact time θ .

While it is to be expected that penetration-theory solutions will generally be expressed in terms of the contact time t , it is often desirable to eliminate the contact time in favor of the physical absorption coefficient by use of Equation (9). This permits a more general expression of penetration-theory results which facilitates their comparison with experimental data and with the predictions of other theories, such as the film theory. If Equation (9) is used to eliminate t from the definition of θ , the result is

$$\theta \equiv k_i B_o t = \frac{4}{\pi} \left[\frac{k_i B_o D_A}{(k_L^*)^2} \right] \equiv \frac{4}{\pi} [M] \quad (11)$$

The more general variable M can be interpreted as a measure of how rapid the chemical reaction is, relative to physical absorption in an absorber with the physical absorption coefficient k_L^* .

INFINITELY RAPID CHEMICAL REACTION

For given values of r and q the variable ϕ approaches an asymptotic value at large values of θ (or M). This asymptote ϕ_a corresponds to the solution for an infinitely rapid chemical reaction. The mathematical solution for this case has been published by Danckwerts (6) and by Sherwood and Pigford (18). The result can be represented by the parametric equations

$$\phi_a = \frac{1/\text{erf}(\sigma)}{1 - \text{erf}(\sigma/\sqrt{r})} \\ q\sqrt{r} = \frac{\text{erf}(\sigma) \exp[\sigma^2(1-1/r)]}{\text{erf}(\sigma)} \quad (12)$$

Figure 1 shows a convenient graphical representation of Equation (12).

It can be seen from Figure 1 that

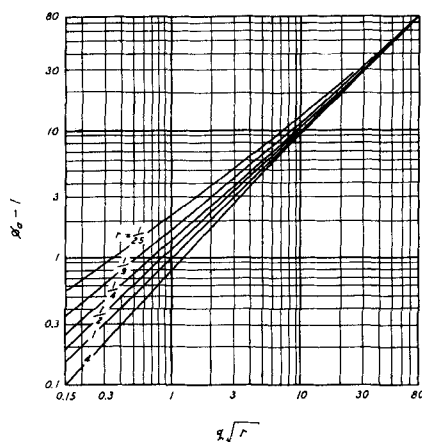


Fig. 1. Penetration-theory solution for absorption with an infinitely rapid irreversible reaction.

the penetration-theory solution can be expressed as

$$\phi_a = 1 + q \quad \text{when } r = 1 \quad (13)$$

$$\phi_a \approx 1 + \sqrt{r} q \quad \text{when } r \neq 1 \quad (14)$$

the approximation of Equation (14) being better at high values of ϕ_a and at values of r near 1.0. The film-theory solution for an infinitely rapid chemical reaction is (18)

$$\phi_a = 1 + rq \quad (15)$$

Thus, for the infinitely rapid chemical reaction, the film and penetration theories agree when the diffusivity ratio is equal to unity; they disagree if r is different from unity.

PSEUDO FIRST-ORDER REACTION

If the concentration of species B is constant throughout the liquid phase at the value B_o , species A undergoes a pseudo first-order reaction with species B. The analytical solution to this problem has been published by Danckwerts (5) and by Sherwood and Pigford (18), and Peaceman (13) has shown that the film and penetration theories agree within 7% for this case.

SOLUTION OF PERRY AND PIGFORD

Except for the limiting conditions of the infinitely rapid reaction and the pseudo first-order reaction, no analytical solutions to Equations (4) and (5) have been achieved. A numerical solution, obtained on a digital computer, has been presented by Perry and Pigford (14). These authors presented results for values of q equal to 1, 2, and 20, with r equal to unity for each case. The range of variation of θ was limited to values up to about 5 or 6. The results were presented in terms of the instantaneous absorption rate, but

Brian (1) has graphically integrated them to obtain ϕ vs. θ and has compared them with the film-theory solution. Van Krevelen and Hoftijzer (20) presented an approximate solution to the film theory

$$\phi = \frac{\sqrt{M} \sqrt{1 - \frac{\phi - 1}{rq}}}{\tanh \left[\sqrt{M} \sqrt{1 - \frac{\phi - 1}{rq}} \right]} \quad (16)$$

and Peaceman (13) proved that this approximation deviates from the true film-theory solution by no more than 8%. For a graphical representation of Equation (16) and a discussion of the various limiting regions reference (9) may be consulted. When Equation (16) was compared (1) with the integrated results of Perry and Pigford, the maximum deviation in the ϕ values was found to be 9%. The solution of Perry and Pigford was limited to values of \sqrt{M} less than 2.4 and to the restricted case of r equal to unity (for which the two theories predict the same value of ϕ_a), but nevertheless the comparison led to the speculation (1, 9) that the penetration theory solution could be approximated by

$$\phi = \frac{\sqrt{M} \sqrt{1 - \frac{\phi - 1}{\phi_a - 1}}}{\tanh \left[\sqrt{M} \sqrt{1 - \frac{\phi - 1}{\phi_a - 1}} \right]} \quad (17)$$

for which ϕ_a is obtained from Equations (12) or Figure 1. This approximation simply assumes that the penetration-theory solution approaches a given value of ϕ_a in the same way that Van Krevelen and Hoftijzer's approximate film-theory equation (20) approaches this same value of ϕ_a , even though the values of r and q may be different for the two theories.

This study was undertaken in order to obtain the penetration-theory solution over wide ranges of the variables M and r . These results are needed to permit a critical comparison of the film- and penetration-theory solutions and to evaluate the adequacy of Equation (17) in approximating the penetration-theory solution.

SOLUTION OF THE EQUATIONS

Finite-Difference Equations

The differential equations [Equations (4) and (5)] were approximated by the following time-centered, implicit finite-difference equations analogous to the equation of Crank and Nicholson (4, 16):

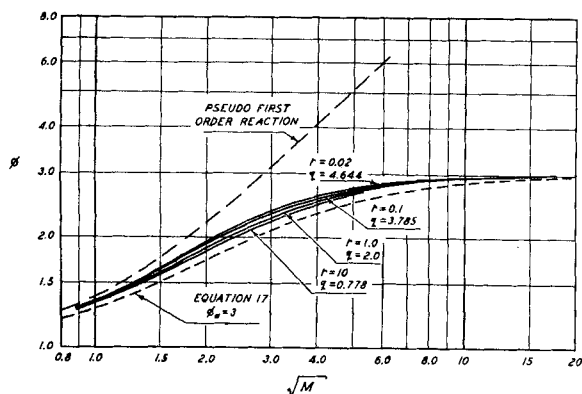


Fig. 2. Penetration-theory solution, $\phi_a = 3$.

$$\begin{aligned} & \left[\frac{\delta^2(a_j^{n+1}) + \delta^2(a_j^n)}{2 \Delta z^2} \right] \\ & - \left[\frac{a_j^{n+1} - a_j^n}{\Delta \theta} \right] \\ & = \left[\frac{a_j^{n+1} + a_j^n}{2} \right] (b_j^{n+1/2}) \\ & \quad (18) \\ & r q \left[\frac{\delta^2(b_j^{n+1}) + \delta^2(b_j^n)}{2 \Delta z^2} \right] \\ & - q \left[\frac{b_j^{n+1} - b_j^n}{\Delta \theta} \right] \\ & = \left[\frac{b_j^{n+1} + b_j^n}{2} \right] (a_j^{n+1/2}) \end{aligned} \quad (19)$$

The solution of these implicit equations was greatly simplified by linearizing according to the method of Douglas (7). The method of solution was as follows: when one assumes that all of the values a_j^n and b_j^n are known at some time-step n for $j = 0, 1, 2, \dots, J$.

1. The values $b_j^{n+1/2}$ ($j = 1, 2, \dots, J$) were calculated by using the explicit finite-difference equation

$$(r q) \frac{\delta^2(b_j^n)}{\Delta z^2} - q \left(\frac{b_j^{n+1/2} - b_j^n}{(\Delta \theta/2)} \right) = a_j^n b_j^n \quad (20)$$

2. Inserting these values of $b_j^{n+1/2}$ into Equation (18) one obtains a sys-

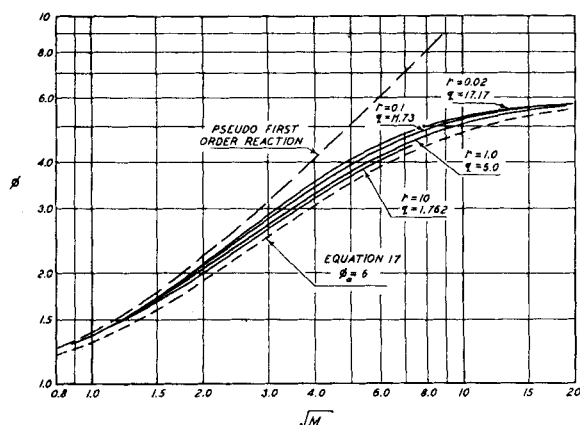


Fig. 3. Penetration-theory solution, $\phi_a = 6$.

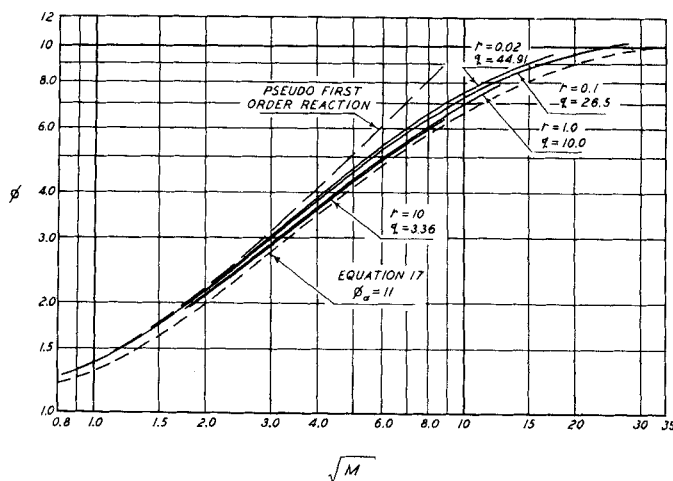


Fig. 4. Penetration-theory solution, $\phi_a = 11$.

tem of J simultaneous linear equations which, together with the boundary conditions to be discussed later, was readily solved for the values a_j^{n+1} ($j = 1, 2, \dots, J$) by the method of solving tridiagonal equations presented in references (2, 17).

3. It was then possible to calculate the values $a_j^{n+1/2}$ as

$$a_j^{n+1/2} = \frac{a_j^{n+1} + a_j^n}{2} \quad (j = 1, 2, \dots, J) \quad (21)$$

with the late-time values a_j^{n+1} just obtained.

4. Inserting these values of $a_j^{n+1/2}$ into Equation (19) one obtains a system of J simultaneous linear equations which, together with the boundary conditions, was solved by the same method as in step 2 to yield the late-time values b_j^{n+1} ($j = 0, 1, 2, \dots, J$).

Thus the complete set of future-time concentrations a_j^{n+1} and b_j^{n+1} was obtained. In the calculations for the next time-step, the order in which Equations (18) and (19) were used was reversed:

5. The values $a_j^{n+1/2}$ were calculated by the explicit finite-difference equation

$$\frac{\delta^2(a_j^n)}{\Delta z^2} - \frac{a_j^{n+1/2} - a_j^n}{(\Delta \theta/2)} = a_j^n b_j^n \quad (22)$$

6. These were inserted into Equation (19) from which were obtained the late-time values b_j^{n+1} .

7. Then the values $b_j^{n+1/2}$ were obtained as

$$b_j^{n+1/2} = \frac{b_j^{n+1} + b_j^n}{2} \quad (23)$$

8. Inserting these values into Equation (18) one obtains the late-time values a_j^{n+1} .

These two schemes were used alternately from cycle to cycle as the calculation proceeded from one time-step to another.

This linearized, implicit finite-difference method was chosen in order to avoid the severe stability limitations encountered when an explicit method is used (2, 7, 12, 16). Perry and Fig-

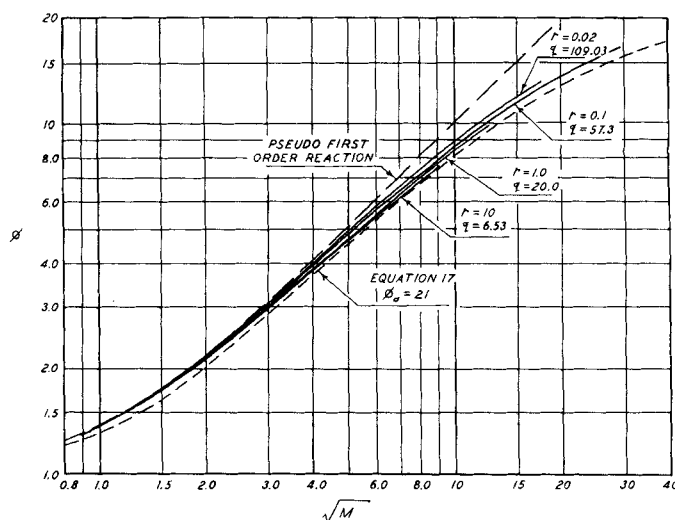


Fig. 5. Penetration-theory solution, $\phi_a = 21$.

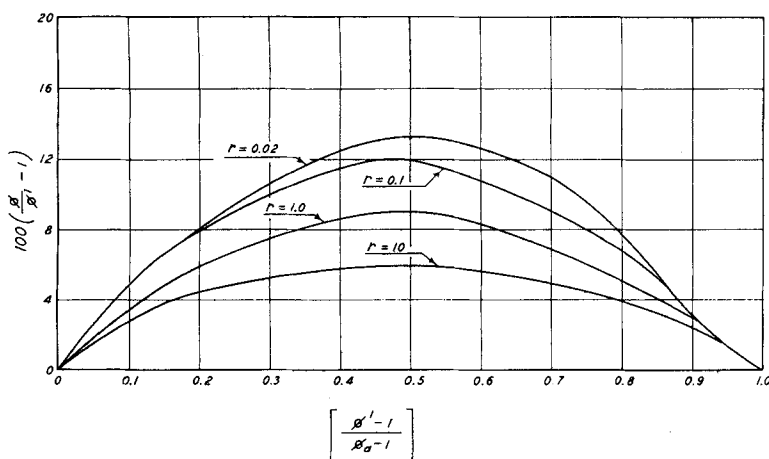


Fig. 6. Correction to Equation (17), $\phi_a = 3$.

ford (14), who used an explicit method, reported that an exceedingly small net size was required, and a very large amount of machine time was consumed in obtaining their results. It was clear that a prohibitively large amount of computer time would be required if an explicit method were used to cover the wide range of variables desired in this study.

Boundary Conditions

The boundary conditions for the difference equations are:

from Equations (6),

$$\left. \begin{array}{l} a_j^0 = 0 \\ b_j^0 = 1 \end{array} \right\} (j = 0, 1, 2, \dots, J) \quad (24)$$

from Equations (7),

$$\left. \begin{array}{l} a_n^n = 1 \\ b_n^n = \frac{18 b_1^n - 9 b_2^n + 2 b_3^n}{11} \end{array} \right\} (n = 1, 2, 3, \dots) \quad (25)$$

The second condition of Equation (25) results from assuming b_n^n to lie on a cubic polynomial in z passing through the values b_1^n , b_2^n , and b_3^n and having a slope of zero at z equal to zero. The form of this boundary condition required an alteration in the procedure for solving tridiagonal equations (2, 17). The method used to solve Equation (19) was a simple extension of the procedure presented in the references. Reference (10) presents the details of the algorithm used.

The boundary condition of Equation (8) was replaced by

$$\left. \begin{array}{l} a_{j-1}^n = a_j^n \\ b_{j-1}^n = b_j^n \end{array} \right\} (n = 1, 2, 3, \dots) \quad (26)$$

with the additional requirement that J must be chosen so large that increasing

it will not affect the resulting values of ϕ .

Computation

The IBM-704 digital computer was programed to solve the difference equations as outlined. For any values of r and q introduced, the computer developed a and b as functions of z and θ ; at selected values of θ , ϕ was evaluated by Equation (10) and Simpson's rule, and the results were printed out as ϕ vs. θ .

A more detailed description of the method of solution and the Fortran program appears in reference (10).

RESULTS AND DISCUSSION

Stability and Convergence

The finite-difference method did not become unstable until the parameter $\Delta\theta/\Delta z^2$ became greater than two hundred times the stability limit for a simple explicit method. Based upon convergence tests made by decreasing $\Delta\theta$ and Δz and upon the agreement of the results with the known asymptotes and with the results of Perry and Pigford (14), it is believed that the results presented here deviate from the

true solution to the differential equations by less than 2%.

Results

The solution, expressed as ϕ vs. θ , was obtained for sixteen combinations of the parameters r and q . The first four combinations are for the following conditions, respectively:

$$r = 0.02, q = 4.644$$

$$r = 0.10, q = 3.785$$

$$r = 1.0, q = 2.000$$

$$r = 10.0, q = 0.778$$

Reference to Figure 1 or Equations (12) reveals that each of these combinations of r and q results in an asymptotic ϕ value of 3.0. The results for these four cases are plotted in Figure 2 as ϕ vs. \sqrt{M} , Equation (11) having been used to substitute M for θ . The pseudo first-order curve (5, 18) is the upper dotted curve. The lower dotted curve is Equation (17) with ϕ_a equal to 3; this of course is also Equation (16) with rq equal to 2. The four penetration-theory curves agree well with each other. While they represent a 500-fold variation in r , when the q values are chosen so that each curve has an asymptote of 3, the ϕ values are found to agree with each other within 7%. The maximum deviation from Equation (17) is 13%.

Figures 3, 4, and 5 contain similar comparisons for the runs with asymptotic ϕ values of 6, 11, and 21, respectively. The comparisons are quite similar to those shown in Figure 2; the deviations between the curves are slightly larger in Figure 3 and somewhat smaller in Figure 5.

It should be noted in Figures 2 and 3 that ϕ approaches its asymptotic value from below. This is in contrast to the ratio of the instantaneous absorption coefficient with chemical reaction to the instantaneous coefficient without chemical reaction; Perry and Pigford (14) showed that this ratio crosses its asymptote and approaches it from above.

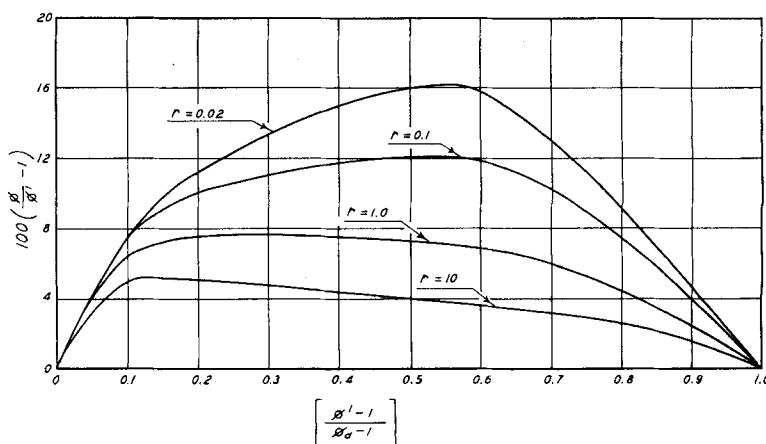


Fig. 7. Correction to Equation (17), $\phi_a = 6$.

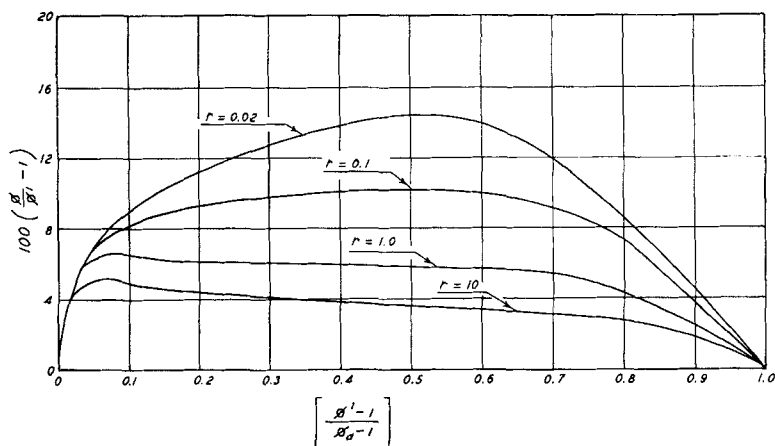


Fig. 8. Correction to Equation (17), $\phi_a = 11$.

Simplified Approximation to Penetration Theory Solution

The results of this study reveal that while Equation (17) is a fair approximation to the penetration theory solution, a correction to this approximate solution is required. Yet the simplicity of Equation (17) suggests that it be retained as a basis for correlating deviations; thus the results presented in Figures 2 through 5 were correlated as deviations from Equation (17) in Figures 6 through 9. Some extrapolation was necessary in preparing these graphs as they appear.

If the penetration theory solution is desired for given values of r , q , and \sqrt{M} , the following procedure is recommended:

1. For the values of r and q in question the asymptote ϕ_a is determined from Equations (12) or Figure 1.
2. For this value of ϕ_a and for the value of \sqrt{M} in question the first approximation to ϕ is obtained from Equation (17). This approximate value is called ϕ' .
3. When one uses this value ϕ' together with ϕ_a and r , the second approximation ϕ can be obtained by interpolation from Figures 6 through 9. This second approximation should be within 3% of the true penetration-theory solution if r , ϕ_a , and \sqrt{M} are within the ranges covered in this study.

CONCLUSIONS

The linearized, time-centered, implicit finite-difference method employed for integrating the nonlinear partial-differential equations proved to be quite effective. This permitted the computation of the penetration-theory solution to values of \sqrt{M} as high as 28, for some cases, with the expenditure of a tolerable amount of computer time. While these results consumed approximately 20 hr. of computer time, it is estimated that more than one

thousand computer hours would be required if an explicit finite-difference method were used.

When the diffusivity ratio r is different from unity, the film and penetration theories predict different asymptotic values of ϕ for an infinitely rapid chemical reaction. However if the two theories are compared, not at the same value of q but at values of q such that the asymptotes are the same, the curves of ϕ vs. \sqrt{M} for the two theories agree surprisingly well. This fact formed the basis of the use of Figures 6 through 9 with Equation (17) for a simplified method of obtaining the penetration theory solution over a wide range of variables.

ACKNOWLEDGMENT

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NOTATION

- A = concentration of species A, the gas being absorbed, moles/cc.
 a = A/A_i
 a_j^n = the value of a at $\theta = n\Delta\theta$, $z = j\Delta z$
 B = concentration of species B, the nonvolatile solute, moles/cc.
 b = B/B_o
 b_j^n = the value of b at $\theta = n\Delta\theta$, $z = j\Delta z$
 D = liquid-phase diffusivity, sq. cm./sec.
 $\text{erf}(\sigma) = (2/\sqrt{\pi}) \int_0^\sigma e^{-y^2} dy$
 $\exp(\sigma) = e^\sigma$
 J = the number of distance increments considered in the finite-difference equations.
 k_1 = reaction rate constant, cc./ (mole) (sec.)
 k_L = liquid-phase absorption coefficient, cm./sec., based on average absorption rate for penetration theory
 $M = kD_A B_o / (k_L^*)^2$ in general
 $= (\pi/4)\theta$ for penetration theory
 $q = B_o/A_i$
 $r = D_B/D_A$
 t = time, sec.
 $\tanh(\sigma) = (e^\sigma - e^{-\sigma}) / (e^\sigma + e^{-\sigma})$
 x = distance into the liquid phase, cm.
 $z = \sqrt{\frac{k_1 B_o}{D_A}} (x)$

Greek Letters

- Δz = increment in z for finite-difference equations.
 $\delta^2(a^n_j) = a^n_{j-1} - 2a^n_j + a^n_{j+1}$, the second distance difference
 $\theta = k_1 B_o t$, dimensionless time of contact
 $\Delta\theta$ = increment in θ for finite-difference equations
 σ = parameter in parametric equations
 $\phi = k_1/k_L^*$

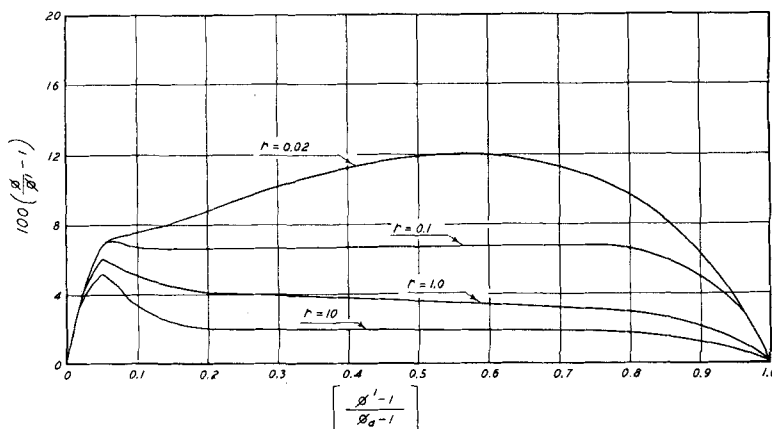


Fig. 9. Correction to Equation (17), $\phi_a = 21$.

ϕ_a = asymptotic value of ϕ at large values of θ (or \sqrt{M})

Subscripts

A = species A
B = species B
G = gas phase
i = to gas-liquid interface
L = to liquid phase
o = in film theory, the bulk liquid core; in penetration theory, the liquid at the start of a contact time interval

Superscripts

* = physical absorption, in which no chemical reaction occurs

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Two-Phase Concurrent Flow in Packed Beds

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Pressure drop and liquid saturation accompanying two-phase concurrent flow have been studied in a variety of packings and with gas-liquid systems having a wide range of fluid properties. Two basic flow patterns were observed with nonfoaming systems. Correlations of pressure-drop and liquid-saturation data were obtained in terms of the single-phase friction losses for the liquid and the gas when each flows alone in the bed. Deviations from the correlation with foaming systems are discussed and illustrated with sample data.

To design reaction vessels having concurrent two-phase flow of liquid and gas through a catalyst bed a knowledge of certain characteristics of the flow is required. The trickle bed reactor is one such area in which predictions of two-phase pressure drop and liquid saturation, the fraction of void space occupied by liquid, are important.

The purpose of this investigation was to obtain data on two-phase flow in packed beds and to establish a suitable correlation for the prediction of the pressure drop and liquid saturation. To narrow the scope somewhat this discussion will be limited to the correlation of simultaneous downflow of a liquid and a gas and to unconsolidated beds of sufficient pore diameter

that capillary forces are not a major effect. Although some foaming systems will be presented, the main portion of the data is further limited to systems which do not foam.

Piret, Mann, and Wall (9) was the only reference found for two-phase concurrent flow in packed beds. The data for four runs indicated that pressure drops for a given liquid rate and with a gas velocity of 0.46 ft./sec. were different by a factor of 2 for the countercurrent and concurrent modes of flow. The concurrent pressure drop was observed to be the lower drop.

TWO-PHASE FLOW IN PIPES

While data on two-phase concurrent flow in packed beds are quite scarce, there has been a substantial interest in pressure-drop data for two-phase flow in open pipes for as long as

engineers have been designing equipment to vaporize liquids or to condense vapors inside tubes. An extensive bibliography of the early investigations may be found in reference 7.

The first important correlation for two-phase flow in open pipes was proposed by Martinelli, Boelter, Taylor, Thomsen, and Morrin (7) and was based upon previously reported data as well as data obtained in horizontal pipes of 1-in. diam. and smaller. Martinelli, Putnam, and Lockhart (8) extended the base of the proposed correlation with additional data in the viscous region. The final paper in the series by Lockhart and Martinelli (6) presents the completed correlation in terms of a single curve for each of the four flow mechanisms observed. These mechanisms are as follows: turbulent flow in the gas phase with turbulent flow in the liquid phase; turbulent flow in the gas phase with viscous flow in the liquid phase; viscous flow in the gas phase with turbulent flow in the liquid phase; and viscous flow in the gas phase with viscous flow in the liq-

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