

# Chapter 7

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## Rate Principles

### INTRODUCTION

The rate transfer process can be described by the product of three terms (see Chapter 5):

- 1 the area available for transfer
- 2 the driving force for transfer
- 3 the (reciprocal of the) resistance to the transfer process

In effect, the rate process in equation form is

$$\text{rate} = \frac{(\text{area})(\text{driving force})}{(\text{resistance})} \quad (7.1)$$

For mass transfer (MT) applications, Equation (7.1) becomes

$$(\text{rate of MT}) = \frac{(\text{area available for MT})(\text{driving force for MT})}{(\text{resistance to MT})} \quad (7.2)$$

Rate principles may be applied at molecular, microscopic, or macroscopic levels. These three approaches were previously discussed in a generic sense in Chapter 2. For mass transfer operations, the molecular rate process will employ the diffusivity in Fick's Law,<sup>(1)</sup> while the microscopic approach will employ overall and individual mass transfer coefficients.

Both the molecular and microscopic approaches are reviewed in this chapter and every attempt has been made to relate the molecular and microscopic approaches to each other. Relating the microscopic treatment of mass transfer operations to the macroscopic treatment is not as simple, but information is available in the literature.<sup>(2,3)</sup> As noted in Chapter 2, macroscopic approaches often produce algebraic equations that have withstood the test of time. For example, the macroscopic equation for calculating the height of an absorber (see Chapter 10) is simply given by the

product of two terms:  $H_{OG}$  and  $N_{OG}$ .  $H_{OG}$  is related to the rate process while  $N_{OG}$  is related to equilibrium.

## THE OPERATING LINE

The  $\text{NH}_3$ –air– $\text{H}_2\text{O}$  discussion presented in Chapter 6 is now revisited in an attempt to shed some light on rate considerations. Refer first to Figure 6.8 in the previous chapter. The initial state and final (equilibrium) state of the system pictured in Figure 7.1 are designated with a square point and a circular point, respectively. If  $V$  and  $L$  represent the moles of flowing air and water, respectively, and  $y$  and  $x$  the mole fraction of  $\text{NH}_3$  in the air and  $\text{H}_2\text{O}$ , respectively, an  $\text{NH}_3$  mole balance written between the initial (0) and final (1) states gives

$$Lx_0 + Vy_0 = Lx_1 + Vy_1 \quad (7.3)$$

(The reader should note that the terms  $V$  and  $G$  are used interchangeably in this text as well as in the literature.) This assumes that if the mole fraction of  $\text{NH}_3$  is small in both phases ( $L$  and  $V$  are essentially constant), this equation may be rearranged to give

$$L(x_1 - x_0) = V(y_0 - y_1)$$

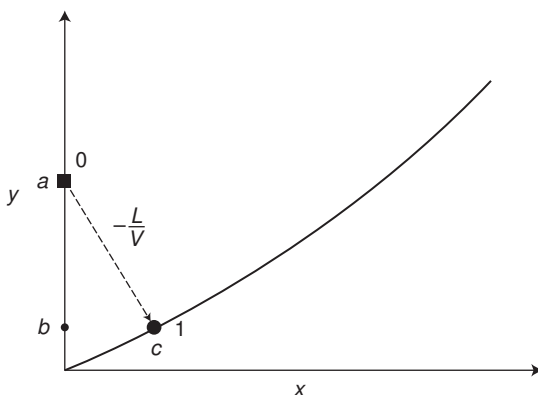
$$\frac{L}{V} = \frac{y_0 - y_1}{x_1 - x_0} \quad (7.4)$$

or

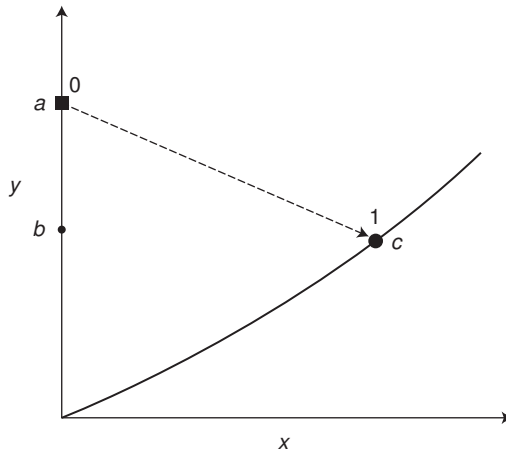
$$-\frac{L}{V} = \frac{y_1 - y_0}{x_1 - x_0} \quad (7.5)$$

There are two important points to be made regarding Figure 7.1.

- 1 The dashed line may be thought of as an “operating line” since it describes the operating  $x$ ,  $y$  values during the system’s transition from point 0 to 1.



**Figure 7.1** Equilibrium-operating line plot.



**Figure 7.2** Equilibrium-operating line plot.

- 2 The vertical displacement of any point from the equilibrium line provides a direct measure of the driving force (and thus the rate) of the system's attempt to achieve equilibrium. In effect, the driving force is maximum at point 0 and zero at point 1.

Equation (7.4) plots out as a straight line as shown with dashes in Figure 7.1. If  $ab/bc = 2.0$ , the mole ratio of liquid to gas is correspondingly 2.0. If the initial state is as shown in Figure 7.2 and  $ab/bc = 0.5$ , then the liquid to gas ratio is also 0.5.

The important point made in the above analysis is that the rate of the  $\text{NH}_3$  transfer process is linearly related to the vertical displacement of the point representing the state of the system from the equilibrium point directly below and, as indicated above, the rate ultimately becomes zero when the operating point reaches the equilibrium point. While this type of macroscopic rate/equilibrium approach receives treatment in Part II—Applications, what follows keys on the aforementioned molecular and microscopic considerations.

## FICK'S LAW

Molecular diffusion results from the motion of molecules. At any instant, the individual molecules in a fluid are moving in random directions at speeds varying from low to high values. The molecules move at random, frequently colliding with one another. Because of the frequent collisions, the molecular velocities are continually changing in both direction and magnitude. Diffusion is more rapid at higher temperatures due to greater molecular velocities. For gases, it is more rapid at low pressures because the average distance between the molecules is greater and the collisions are less frequent.

If a solution is not uniform in concentration, the solution is gradually brought into uniformity by diffusion; the molecules move from an area of high concentration to one

of low concentration. The rate at which a solute travels depends on the concentration gradient which exists in the solution. This gradient applies across adjacent regions of high and low concentrations. However, a quantitative measure of rate is needed to describe what is occurring.

The rate of diffusion can be described in terms of a molar flux term, with units of moles/(area)(time), and with the area being measured as that which the solute diffuses through. In a nonuniform solution containing only two components, both must diffuse if uniformity is to occur. This leads to the use of two fluxes to describe the motion of one of the components  $N$ , the flux relative to a fixed location and  $J$ , the flux of a component relative to the average molar velocity of all components. The first of these is of importance in the design of equipment, but the second is more characteristic of the nature of the component. For example, the rate at which a fish swims upstream against the flowing current is analogous to  $N$ , while the velocity of the fish relative to the stream is more characteristic of the swimming ability of the fish and is analogous to  $J$ .

The diffusivity, or diffusion coefficient,  $D_{AB}$ , of component  $A$  in solution  $B$ , which is a measure of its diffusive mobility, is defined as the ratio of its flux,  $J_A$ , to its concentration gradient and is given by

$$J_A = -D_{AB} \frac{\partial C_A}{\partial z} \quad (7.6)$$

This is Fick's first law<sup>(1)</sup> written for the  $z$  direction. The concentration gradient term represents the variation of the concentration,  $C_A$ , in the  $z$  direction. The negative sign accounts for diffusion occurring from high to low concentrations. The diffusivity is a characteristic of the component and its environment (temperature, pressure, concentration, etc.). This equation is analogous to the flux equations<sup>(2,3)</sup> defined for momentum transfer (in terms of the previously defined viscosity) and for heat transfer (in terms of the thermal conductivity). The diffusivity is usually expressed with units of (length)<sup>2</sup>/time or moles/time · area. This coefficient, as well as Fick's law, will receive additional treatment later in this chapter.

### ILLUSTRATIVE EXAMPLE 7.1

Express the diffusivity in English units.

**SOLUTION:** Based on its definition, the units of the diffusivity may be expressed as either

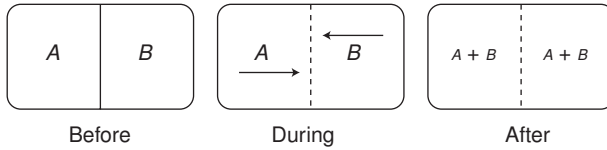
$$\text{ft}^2/\text{hr}$$

or

$$\text{lbmol}/\text{hr} \cdot \text{ft}$$

The latter units are derived by simply multiplying  $\text{ft}^2/\text{hr}$  by the molar density, i.e.,  $\text{lbmol}/\text{ft}^3$ . ■

Imagine if two fluids are placed side by side in a container separated by a partition.<sup>(2)</sup> As pictured in Figure 7.3, fluid  $A$  is on the left-hand side and fluid  $B$  is on



**Figure 7.3** Diffusion process.

the right-hand side. When the partition is removed, the two fluids begin to diffuse ( $A$  towards  $B$  and  $B$  towards  $A$ ). The diffusion process occurs because of a finite concentration driving force, which is the concentration gradient between the two containers. Diffusion stops when the concentration is uniform throughout the total mixture, i.e., there is no concentration gradient or driving force. However, imagine that there has been a net mass movement to the right. If the direction to the right is taken as positive, the flux of  $A$  (noted as  $N_A$ ), relative to a fixed position is positive, while the flux of  $B$ ,  $N_B$ , is negative. At steady state, the net flux is

$$N_A + N_B = N \quad (7.7)$$

The movement of  $A$  is made up of two parts, namely, that resulting from the bulk movement of  $A$  in  $N$  (i.e.,  $x_A N$ ), and that resulting from the diffusion of  $A$  through  $B$ . This latter effect is defined as  $J_A$ . The sum of these two effects is then

$$N_A = x_A N + J_A \quad (7.8)$$

Employing Fick's first law leads to

$$N_A = x_A(N_A + N_B) - D_{AB} \frac{\partial C_A}{\partial z} \quad (7.9)$$

The next sub-section presents three different cases of steady-state molecular diffusion in gases.<sup>(2,3)</sup>

## Diffusion in Gases

**1 Diffusion of  $A$  through non-diffusing  $B$ .** This can be illustrated by the example of ammonia ( $A$ ) being absorbed from air ( $B$ ) into water. Since air does not dissolve appreciably in water, only ammonia diffuses. For this case,  $N_B = 0$  and  $N_A$  is constant and the following relationship can be written

$$\frac{N_A}{N_A + N_B} = 1 \quad (7.10)$$

It can be shown that for a system where component  $A$  diffuses through non-diffusing  $B$ , the diffusion rate can be calculated from

$$N_A = \left[ \frac{D_{G,AB} P}{RTz \bar{p}_{B,M}} \right] (p_{A1} - p_{A2}) \quad (7.11)$$

where  $\bar{p}_{B,M} = \frac{p_{B2} - p_{B1}}{\ln(p_{B2}/p_{B1})}$  is the log mean partial pressure difference driving force of component  $B$  and  $p_{B1} = P - p_{A1}$ ,  $p_{B2} = P - p_{A1}$ ,  $p_{B1}$  is the partial pressure of component  $B$  at the liquid–vapor interface,  $p_{B2}$  is the partial pressure of component  $B$  at distance  $z$  from the interface,  $p_{A1}$  is the partial pressure of component  $A$  at the interface,  $p_{A2}$  is the partial pressure of component  $A$  at distance  $z$  from the interface,  $P$  is the total system pressure,  $D_{G,AB}$  is the diffusivity of component  $A$  through  $B$ ,  $T$  is the system absolute temperature,  $z$  is the distance from the interface for state 2, and  $R$  is the ideal gas constant.

For addition analyses of these systems, the interested reader is referred to the original works of Treybal<sup>(2)</sup> and Geankopolis.<sup>(4)</sup>

- 2 Steady state equimolar counter-diffusion.** For this case, both  $A$  and  $B$  are diffusing, as occurs in distillation operations. Again, a detailed analysis of this system ultimately leads to

$$N_A = \left[ \frac{D_{G,AB}}{RTz} \right] (p_{A1} - p_{A2}) \quad (7.12)$$

- 3 Steady-state diffusion in multicomponent mixtures.** An example of this is the diffusion of oxygen in a non-diffusing mixture of methane and hydrogen. The estimation of diffusion in multicomponent systems is very complicated, but it can usually be handled by defining an effective diffusivity,  $D_{AM}$ . The effective diffusivity of one component can be calculated based on its diffusivity with each of the other constituents. It can be shown that  $D_{AM}$  can be represented by the following equation

$$D_{AM} = \frac{1}{\sum_{i=B}^i \frac{y_i}{D_{Ai}}} \quad (7.13)$$

where  $D_{Ai}$  is the binary diffusivity of component  $A$  in each of the components present in the system and  $y_i$  is the mole fraction of component  $i$  on an  $A$  free basis, e.g., for a system consisting of components  $A$ ,  $B$ , and  $C$ ,  $y_B$  is the mole fraction of  $B$  based on  $B$  and  $C$  only. Thus, the calculation of the effective diffusivity of oxygen in a mixture of methane and hydrogen is dependent on the diffusivity of oxygen in methane and oxygen in hydrogen.

### ILLUSTRATIVE EXAMPLE 7.2

Calculate the rate of diffusion of oxygen ( $A$ ) in a non-diffusing mixture of carbon monoxide ( $B$ ) and carbon dioxide ( $C$ ), which has a volume ratio of 3:1, respectively. The system temperature and pressure are 25°C and  $1 \times 10^5 \text{ N/m}^2$  at the interface, respectively. The oxygen partial pressure is 15,000  $\text{N/m}^2$  at the interface and 7500  $\text{N/m}^2$  at a distance of 3 mm from the interface. It can be assumed that the partial pressure of the non-diffusing mixture is the difference between the total pressure and the oxygen partial pressure at each location. In addition,  $D_{AB} = 0.185 \text{ cm}^2/\text{s}$  and  $D_{AC} = 0.139 \text{ cm}^2/\text{s}$ .

**SOLUTION:** The describing equation for the diffusivity for this system is given by Equation (7.13)

$$D_{AM} = \frac{1}{\sum_{i=B}^i \frac{y_i}{D_{Ai}}}$$

Write the mole fraction of each non-diffusing component on an *A*-free basis

$$y_B = 0.75$$

$$y_C = 0.25$$

Calculate the effective diffusivity for the mixture

$$D_{AM} = \frac{1}{\sum_{i=B}^i \frac{y_i}{D_{Ai}}} = \frac{1}{\left(\frac{y_B}{D_{AB}}\right) + \left(\frac{y_C}{D_{AC}}\right)} \frac{1}{\frac{0.75}{0.185} + \frac{0.25}{0.139}} = 0.171 \text{ cm}^2/\text{s}$$

Obtain the log mean pressure difference for the mixture

$$\bar{p}_{B,M} = \frac{p_{B2} - p_{B1}}{\ln\left(\frac{p_{B2}}{p_{B1}}\right)} = \frac{(10^5 - 7500) - (10^5 - 15,000)}{\ln\left(\frac{(10^5 - 7500)}{(10^5 - 15,000)}\right)} = 8.87 \times 10^4 \text{ N/m}^2$$

Calculate the diffusion of oxygen in the mixture using Equation (7.11), being careful to maintain consistent units

$$N_A = \left[ \frac{D_{G,AB}P}{RTz\bar{p}_{B,M}} \right] (p_{A1} - p_{A2}) = \frac{(0.171/1000)(10^5)}{(8.314)(298)(0.003)(8.87 \times 10^4)} (15,000 - 7500) \\ = 1.95 \times 10^{-2} \text{ gmol/m}^2 \cdot \text{s} \quad \blacksquare$$

### ILLUSTRATIVE EXAMPLE 7.3

Ethylene is diffusing at a constant rate through a 2 mm-thick stagnant layer of nitrogen. Conditions are such that at one boundary of the stagnant layer, the gas contains 60% by volume ethylene. The ethylene concentration at the other boundary can be considered negligible. The total pressure is one atmosphere and the temperature 25°C. The diffusivity for the mixture is 0.163 cm<sup>2</sup>/s. Determine the rate of diffusion of ethylene through the nitrogen layer.

**SOLUTION:** This is another case of steady-state diffusion through a second non-diffusing gas; hence, Equation (7.11) is again applicable.

$$N_A = \left[ \frac{D_{G,AB}P}{RTz\bar{p}_{B,M}} \right] (p_{A1} - p_{A2})$$

Pertinent data are now rewritten (*A* = ethylene, *B* = nitrogen)

$$D_{AB} = 0.163 \text{ cm}^2/\text{s}$$

$$P = 1.0 \text{ atm}$$

$$R = 82.07 \text{ cm}^3 \cdot \text{atm}/\text{gmol} \cdot \text{K}$$

$$T = 298 \text{ K}$$

$$p_{A1} = (0.6)(1.0) = 0.6 \text{ atm}$$

$$p_{A2} = 0.0 \text{ atm}$$

$$p_{B1} = 1.0 - 0.6 = 0.4 \text{ atm}$$

$$p_{B2} = 1.0 \text{ atm}$$

$$z = 2 \text{ mm} = 0.2 \text{ cm}$$

Substituting yields

$$\bar{p}_{B,M} = \frac{p_{B2} - p_{B1}}{\ln\left(\frac{p_{B2}}{p_{B1}}\right)} = \frac{1 - 0.4}{\ln\left(\frac{1}{0.4}\right)} = 0.655 \text{ atm}$$

Substitute into Equation (7.11).

$$\begin{aligned} N_A &= \left[ \frac{D_{G,AB}P}{RTz\bar{p}_{B,M}} \right] (p_{A1} - p_{A2}) \\ &= \left[ \frac{(0.163)(1)}{(82.07)(298)(0.2)(0.655)} \right] (0.6) \\ &= 3.047 \times 10^{-5} \text{ gmol/cm}^2 \cdot \text{s} \end{aligned}$$

The diffusivity, or diffusion coefficient,  $D$ , was defined previously as the proportionality constant in the rate equation for mass transfer (Fick's law) and it is a property of the system that is dependent on temperature, pressure, and the nature of the components. Reliable diffusion data is difficult to obtain, particularly over a wide range of temperatures. Table 7.1 lists diffusion coefficients for a few pairs of gases that have been investigated. The diffusion coefficient,  $D_G$  ( $\text{cm}^2/\text{s}$ ), at a temperature  $T_C$  ( $^\circ\text{C}$ ) and pressure  $P$  (atm), may be determined from the data in Table 7.1 at state "0" and the following equation

$$D_G = D_{G0} \left( \frac{T_C + 273.2}{T_{C0} + 273.2} \right)^b \frac{1}{P} \quad (7.14)$$

Additional values for diffusivities may be found in the literature.<sup>(5)</sup> When experimentally determined diffusivity data is not readily available, several estimation

**Table 7.1** Diffusion Coefficients

Gas	Air	CO <sub>2</sub>	H <sub>2</sub>	N <sub>2</sub>	O <sub>2</sub>	$T_{C0}$ , $^\circ\text{C}$	$b$
Air	–	–	0.611	–	0.178	0	1.75
CO	–	0.137	0.651	0.192	0.185	0	1.75
CO <sub>2</sub>	0.138	–	–	–	0.139	0	2.00
H <sub>2</sub>	0.611	0.550	–	0.674	0.697 <sup>a</sup>	0	1.75
N <sub>2</sub>	–	0.144	0.674	–	0.181	0	1.75

<sup>a</sup>Temperature ( $T_{C0}$ ) for H<sub>2</sub> in O<sub>2</sub> is 20 $^\circ\text{C}$ .



techniques based on the kinetic theory of gases are available. However, the gas diffusivity may be calculated directly by using the following expression:

$$D_G = \frac{B'T^{1.5}\sqrt{MW}}{P\sigma_{AB}^2\Omega} \quad (7.15)$$

where  $D_G$  = gas diffusivity,  $\text{cm}^2/\text{s}$ ,  $B'$  = molecular weight parameter, dimensionless,  $\overline{MW}$  = weighted average molecular weight, dimensionless,  $\sigma_{AB}$  = molar volume parameter, dimensionless, and  $\Omega$  = parameter, dimensionless.

The term  $B'$  can be found using the equation:

$$B' = 0.00217 - 0.00050\sqrt{\frac{1}{MW_A} + \frac{1}{MW_B}} \quad (7.16)$$

where  $MW_A$  = molecular weight of  $A$  and  $MW_B$  = molecular weight of  $B$ .

## Diffusion in Liquids

No purely theoretical generalized correlation of liquid phase diffusivities has yet been found, but certain empirical equations are available. This probably reflects the inadequacy of kinetic theory when applied to liquids. It is therefore preferable to use experimental data for liquid phase diffusivities. Table 7.2 provides a number of typical values for liquid phase diffusivities. Additional liquid diffusivities are available in the literature.<sup>(2,5)</sup>

Investigations on diffusion in liquids are not as extensive as those on diffusion in gases, with less experimental data available. The rate of diffusion in liquids may take a long time to reach equilibrium unless agitated. This is, in part, explained by the fact that there is a much closer spacing of the molecules in a liquid, thereby retarding the movement of solute. Thus, molecular attractions become more important. Also note that diffusivity values in liquids are therefore smaller than in gases.

In the absence of an adequate theory for diffusion in liquids, it is usually assumed that Fick's law is obeyed and that the equations developed in the subsection for diffusion in gases can also be applied to diffusion in liquids. Two situations are considered below.

**Table 7.2** Liquid Diffusivities at Atmospheric Pressure

Solute	Solvent	Temperature, °C	Diffusivity, $\text{m}^2/\text{s} \times 10^9$
NH <sub>3</sub>	Water	5	1.24
NaCl	Water	18	1.26
Ethanol	Water	10	0.5
Acetic Acid	Water	12.5	0.82

**1 Steady-state diffusion of A through non-diffusing B**

$$N_A = \frac{D_{L,AB}}{z\bar{x}_{B,M}} \left( \frac{\rho}{MW} \right)_{\text{avg}} (x_{A1} - x_{A2}) \quad (7.17)$$

**2 Steady-state equimolar counterdiffusion**

$$N_A = \frac{D_{L,AB}}{z} \left( \frac{\rho}{MW} \right)_{\text{avg}} (x_{A1} - x_{A2}) \quad (7.18)$$

where  $\rho$  is the mass density of the solution, and  $MW$  is the molecular weight of the solution.

When no experimental data is available, an estimation of the liquid diffusivity,  $D_{L,AB}$ , can be obtained using various empirical approaches, one of which is the Wilke–Chang equation<sup>(6)</sup>

$$D_{L,AB} = \frac{117.3 \times 10^{-18} (\varphi MW_B)^{0.5}}{\mu v_A^{0.6}} \quad (7.19)$$

where  $D_{L,AB}$  is the diffusivity of A in a very dilute solution in solvent B ( $\text{m}^2/\text{s}$ ),  $MW_B$  is the molecular weight of solvent (g/gmol),  $T$  is the temperature (K),  $\mu$  is the solution viscosity ( $\text{kg}/\text{m} \cdot \text{s}$ ),  $v_A$  is the solute molar volume at the normal boiling point ( $\text{m}^3/\text{kmol}$ ), and  $\varphi$  is the association factor for the solvent ( $\varphi = 2.26$  for water, 1.5 for ethanol, and 1.0 for solvents like benzene and ethyl ether).

A modified form of Equation (7.19) that can be used to obtain the diffusivity is

$$D_L = \frac{13.26 \times 10^{-5}}{\mu^{1.14} v_A^{0.589}} \quad (7.20)$$

where  $D_L$  = liquid diffusivity,  $\text{cm}^2/\text{s}$

$v_A$  = molar volume of solute,  $\text{cm}^3/\text{mol}$

**MASS TRANSFER COEFFICIENTS**

In the discussion of diffusion in the previous section, the emphasis was placed on the molecular transport in fluids that were stagnant or in laminar flow. However, in many cases, these diffusion processes are too slow, and more rapid diffusion or transport is required. Quite often, to speed up this diffusion, the fluid velocity is increased so that turbulent transport occurs.

When a fluid flows past a surface under such conditions that the fluid is in turbulent flow, the actual velocity of small parcels or lumps of fluid cannot be described as simply as in laminar flow. Since fluid flows in smooth streamlines in laminar flow, its behavior can usually be described mathematically. However, there are no orderly streamlines or equations to describe fluid behavior in turbulent motion. However, there are large eddies or “chunks” of fluid which move rapidly in a seemingly random fashion. This eddy transfer, or turbulent diffusion, is very fast in comparison

to the relatively slow process of molecular diffusion, where each solute molecule must move by random motion through the fluid.

When a fluid flows past a surface under conditions such that turbulence generally prevails, a thin laminar-type sublayer film exists adjacent to the surface. The mass transfer in this region occurs by molecular diffusion since little or no eddies are present. Since this is a slow process, a large concentration gradient or decrease in concentration across this laminar film occurs. Adjacent to this is the transition or buffer region. Here, some eddy activity exists and the transfer occurs by the sum of molecular and turbulent diffusion. In this region, there is a gradual and non-abrupt transition from the total transfer occurring by almost pure molecular diffusion at one end to mainly turbulent at the other end. The concentration decrease is much less in this region. Although most of the transfer is by turbulent or eddy diffusion, molecular diffusion still occurs, but it contributes little to the overall transfer. The concentration decrease is very small here since the rapid eddy movement evens out any gradients tending to exist.

Many approaches to the turbulent (convective) mass transfer problem exist: film theory, combined film-surface-renewal theory, boundary layer theory, empirical approaches, etc. One former theory has somehow managed to survive the test of time, having been successful in interpreting the results of most two-phase mass transfer operations of industrial importance.

Film theory<sup>(7)</sup> (as applied by Whitman) postulates the existence of an imaginary stagnant film next to the interface whose resistance to mass transfer is equal to the total mass transfer resistance of the system. The difficulty with this theory is in the calculation of the effective film thickness. Other theories are briefly detailed below.

- 1 Surface-renewal theory assumes that a clump of fluid far from the interface:
  - (a) moves to the interface without transferring mass,
  - (b) sits there stagnant, transferring mass by molecular diffusion for a time short enough such that little change in the concentration profile occurs in the clump, and
  - (c) then moves away from the interface without transferring mass en route and mixes with the bulk fluid instantly.

This theory is somewhat more satisfactory in general than film theory.

- 2 Boundary layer theory rests on the solution of a set of simplified differential equations which are approximations to a more nearly correct set of differential equations.<sup>(8)</sup>
- 3 Empirical approaches, which are merely data correlations, serve for specific cases, but give little information about extrapolation.

Further details regarding any of these approaches are available in the literature.<sup>(8)</sup>

## Individual Mass Transfer Coefficients

In the mass transfer operation of gas absorption, two insoluble phases are brought into contact in order to permit the transfer of a solute from one phase to the other (e.g., and as discussed earlier, ammonia can be absorbed from an air-ammonia mixture into

a water stream without air dissolving appreciably in the water). Concern for this application is with the simultaneous application of the diffusion mechanism for each phase to the combined system.

It has already been shown that the rate of diffusion within each phase is dependent on the concentration gradient existing within it. At the same time, the concentration gradients of a two-phase system are indicative of the departure from equilibrium which exists between the phases. Since this departure from equilibrium provides the driving force for diffusion, the rates of diffusion in terms of the driving forces may now be studied.

In view of Whitman's two-film theory, it is assumed that at the gas-liquid interface the principal diffusion resistances occur in a thin film of gas and a thin film of liquid. The rates of diffusion in these two films will describe the mass transfer operation. The diffusion coefficient,  $D$ , in Fick's law is inversely proportional to the concentration of the inert material,  $c_B$ , in the liquid film through which material A must diffuse. Replacing  $D$  with  $(k/c_B)$  yields

$$N_A = \left[ \frac{k}{z c_{BM}} \right] (c_{AI} - c_{AL}) \quad (7.21)$$

where  $c_{BM}$  is the log mean concentration difference of the inert material across the film,  $k$  is a proportionality constant, and  $N$  is the amount of material transferred per unit area per unit time, or mols/area · time. The practical application of Equation (7.21) is based on the assumption that  $z$ , the film thickness, is a constant that represents an effective average value throughout the length of the contact path. Also,  $c_{BM}$  is considered to be constant since many mass transfer processes usually involve fairly dilute mixtures and solutions. Equation (7.21) can therefore be written as

$$N_A = k_L (c_{AI} - c_{AL}) \quad (7.22)$$

or

$$N_A = k_G (p_{AG} - p_{AI}) \quad (7.23)$$

where  $k_L$  is the liquid mass transfer coefficient based on concentration,  $k_G$  is the gas mass transfer coefficient based on partial pressure,  $c_{AI}$  is the interfacial (surface) concentration of component A,  $c_{AL}$  is the bulk liquid concentration of component A,  $p_{AI}$  is the interfacial partial pressure of component A,  $p_{AG}$  is the partial pressure of component A.

Equation (7.22) expresses the transfer of  $N$  molecules of solute (A) through the liquid film under a concentration driving force, and Equation (7.23), the transfer of the same number of molecules of solute through the gas film under a partial pressure driving force.

For certain simplified cases of molecular diffusion, equations can be derived to determine precisely the rate at which mass is being transferred. For example, the equation below was presented in the previous section

$$N_A = \left[ \frac{D_{G,AB} P}{RTz \bar{p}_{B,M}} \right] (p_{A1} - p_{A2}) \quad (7.11)$$

The bracketed term above is an exact definition for the individual mass transfer coefficient corresponding to the steady-state situation of one component diffusing through a nondiffusing second component. In principle, it is then not necessary to calculate any other mass transfer coefficient for laminar flow since molecular diffusion prevails and exact equations are available. However, in general, obtaining such analytically-derived expressions are difficult; in most cases encountered in practice, it is impossible since turbulent mass transfer, which becomes quite complex, usually prevails.

Only the more frequently encountered situations of diffusion will now be discussed, namely, equimolar counterdiffusion and diffusion of one component through another non-diffusing component.<sup>(2,4)</sup>

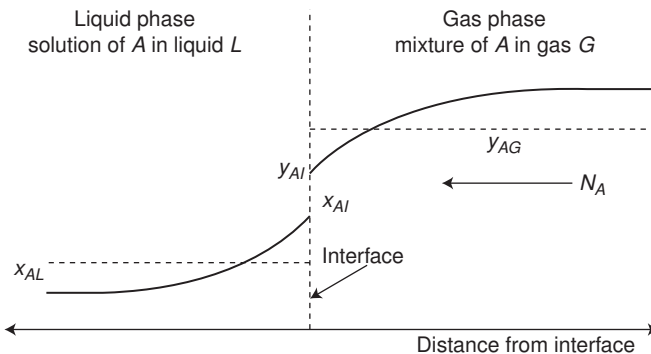
## Equimolar Counterdiffusion

In absorption operations, the absorbing medium may evaporate into the gas being treated, resulting in the simultaneous diffusion of both gases in opposite directions. The diffusion of each gas is affected by the presence of the molecules of the other gas and hindered if the gases are diffusing in opposite directions. When such a situation exists, the diffusion of equal moles per unit area per unit time occurs in opposite directions. This is referred to as equimolar counterdiffusion.

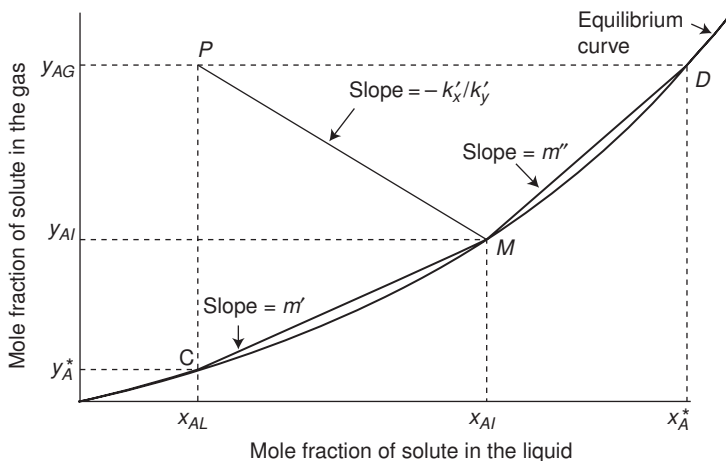
For the case of equimolar counterdiffusion, the concentration profile shown in Figure 7.4 can be plotted graphically as in Figure 7.5. In this latter figure, point *P* represents the bulk phase compositions  $y_{AG}$  and  $x_{AL}$ , and point *M* represents the concentrations  $y_{AI}$  and  $x_{AI}$  at the interface. The equations for the flux of component *A*, when *A* is diffusing from a gas to a liquid and there is equimolar counterdiffusion of component *B* from the liquid, are given by

$$N_A = k'_y(y_{AG} - y_{AI}) = k'_x(x_{AI} - x_{AL}) \quad (7.24)$$

where  $k'_x$  is the liquid mass transfer coefficient based on mole fraction,  $k'_y$  is the gas mass transfer coefficient based on mole fraction,  $x_{AI}$  and  $y_{AI}$  are the bulk liquid and gas interfacial mole fraction, respectively, and  $x_{AL}$  and  $y_{AG}$  are the bulk liquid and



**Figure 7.4** Concentration (mole fraction) profile of solute *A* diffusing from one phase to another.



**Figure 7.5** Overall concentration differences.

gas mole fractions, respectively. Note that the prime with the individual mass transfer coefficient, i.e.,  $k'_y$ , is a reminder that the transfer process involves equimolar counterdiffusion.

The values  $(y_{AG} - y_{AI})$  and  $(x_{AI} - x_{AL})$  are the differences in concentration, or driving forces, in each phase. For example,  $(y_{AG} - y_{AI})$  is the driving force in the vapor phase since  $y_{AG}$  represents the average vapor concentration at a distance from the liquid vapor interface which has a composition  $y_{AI}$ . Rearranging Equation (7.24) gives

$$-\frac{k'_x}{k'_y} = \frac{(y_{AG} - y_{AI})}{(x_{AL} - x_{AI})} \quad (7.25)$$

Hence, the slope of  $PM$  in Figure 7.5 is  $-(k'_x/k'_y)$ . This means that if the two mass transfer coefficients are known, then the interfacial compositions can be determined by the line  $PM$ . While the bulk concentrations  $y_{AG}$  and  $x_{AL}$  can ordinarily be determined experimentally (from equilibrium relationships), the concentrations at the interface cannot, and Equation (7.25) can instead be used.

## Diffusion of Component A Through Non-diffusing Component B

In the case of component A diffusing through nondiffusing component B, the concentrations can also be plotted as in Figure 7.5 where  $P$  represents the bulk phase compositions and  $M$ , the interface. The equation for A diffusing through a stagnant gas and through a stagnant liquid can be shown to be:

$$N_A = \frac{k'_y}{(1 - y_A)_{IM}} (y_{AG} - y_{AI}) = \frac{k'_x}{(1 - x_A)_{IM}} (x_{AI} - x_{AL}) \quad (7.26)$$

where the subscript “*IM*” represents the bulk flow correction factor for nondilute liquid and gas phases. The correction terms can be calculated from the following two equations:

$$(1 - y_A)_{IM} = \frac{(1 - y_{AI}) - (1 - y_{AG})}{\ln \frac{(1 - y_{AI})}{(1 - y_{AG})}} \quad (7.27)$$

or,

$$(1 - x_A)_{IM} = \frac{(1 - x_{AL}) - (1 - x_{AI})}{\ln \frac{(1 - x_{AL})}{(1 - x_{AI})}} \quad (7.28)$$

Equation (7.26) may be combined with Equations (7.27) and (7.28) to give:

$$-\frac{k'_x/(1 - x_A)_{IM}}{k'_y/(1 - y_A)_{IM}} = \frac{(y_{AG} - y_{AI})}{(x_{AL} - x_{AI})} \quad (7.29)$$

The slope of the line *PM* for the case of *A* diffusing through stagnant *B* is given by the left-hand side of Equation (7.29). The slope of Equation (7.29) differs from that of Equation (7.25) for equimolar counterdiffusion by the bulk flow correction terms,  $(1 - y_A)_{IM}$  and  $(1 - x_A)_{IM}$ . When *A* is diffusing through nondiffusing *B* and the solutions are dilute, the bulk flow correction terms are approximately unity, and Equation (7.25) can be used instead of Equation (7.29). It is for this reason that Equation (7.25) is often employed even if the transfer process involves *A* diffusing through nondiffusing *B*. Also note that the subscripts *L* and *G*, e.g.,  $k_L$  and  $k_G$ , are employed when the rate is expressed in terms of the concentration and partial pressure, respectively.

The use of Equation (7.29) to obtain the slope is, by necessity, trial-and-error because the left-hand side contains the interfacial concentrations  $y_{AI}$  and  $x_{AI}$  which are being sought. A first trial estimate can be obtained using Equation (7.25). With these estimates for  $y_{AI}$  and  $x_{AI}$ , a value of the left-hand side of Equation (7.29) is computed and a new slope drawn to obtain new values of  $y_{AI}$  and  $x_{AI}$  (read off the equilibrium line). The second trial is repeated until the values of  $y_{AI}$  and  $x_{AI}$  do not change significantly with successive trials. Three trials will usually suffice.

#### ILLUSTRATIVE EXAMPLE 7.4

An air–ammonia mixture is being treated with a water stream in a 10 ft pilot scale absorber in a lab test under atmospheric conditions (70°F and 1 atm). The absorber was set-up in such a way to measure the partial pressure of each component at various locations in the column. It was found that at a distance of 5 feet up the column the liquid contained 0.4 wt% ammonia while the partial pressure of the ammonia was 10 mm Hg. Estimate the interfacial gas and liquid phase concentrations. Based on previous experiments, the ratio of the liquid mass transfer coefficient ( $-k_L/k_G$ ) was found to be  $-1.0 \text{ atm/lbmol} \cdot \text{ft}^3$ . Solubility data for this system are presented in Table 7.3.

**Table 7.3** Ammonia Data

NH <sub>3</sub> partial pressure, atm	Liquid concentration, lbmol NH <sub>3</sub> /ft <sup>3</sup>
0.0045	0.0180
0.0097	0.0367
0.0119	0.0440
0.0158	0.0586
0.0201	0.0733
0.0255	0.0916
0.0309	0.1100

**SOLUTION:** Write the describing equation for this system. See Equation (7.24).

$$N_A = k'_y(y_{AG} - y_{AI}) = k'_x(x_{AI} - x_{AL})$$

Construct an equilibrium diagram from the data in Table 7.3, as shown in Figure 7.6.

Based on the data provided

$$y_{\text{NH}_3} = 10/760 = 0.0136; \quad p_{\text{NH}_3} = 0.0136 \text{ atm}$$

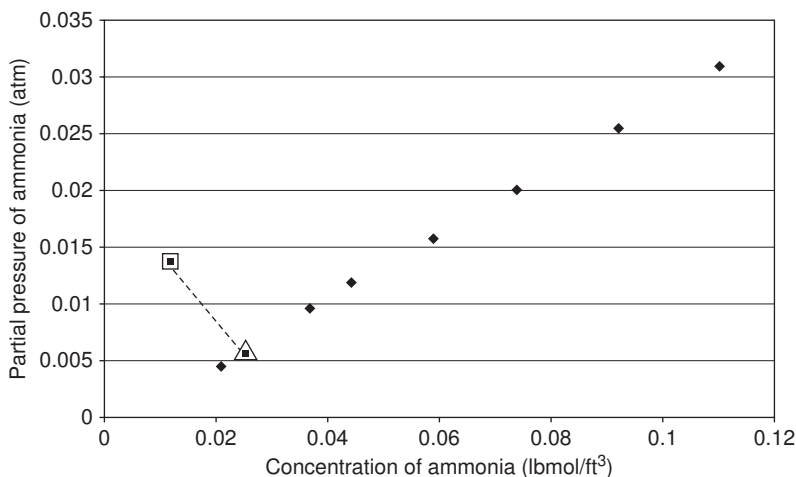
The corresponding liquid concentration is

$$c_{\text{NH}_3} = (0.004/17)62.4 = 0.0147 \text{ lbmol/ft}^3$$

These two conditions are represented as a square point on Figure 7.6. A line extended from that point with a slope of  $-1.0$  intersects the equilibrium line (see triangle point) at approximately:

$$p_{AI} = 0.0056 \text{ atm}$$

$$c_{AI} = 0.023 \text{ lbmol/ft}^3$$



**Figure 7.6** Equilibrium plot for ammonia–water.



**ILLUSTRATIVE EXAMPLE 7.5**

Referring to Illustrative Example 7.4, if the mass transfer coefficients ( $k_G$ ) is  $12.5 \text{ lbmol/h} \cdot \text{ft}^2 \cdot \text{atm}$ , estimate the molar flux of the ammonia.

**SOLUTION:** Apply Equation (7.24). First note that

$$p_{AI} = y_{AI}P$$

so that,

$$y_{AI} = \frac{p_{AI}}{P}$$

Since  $P = 1 \text{ atm}$  and the solution is dilute, one may write

$$\begin{aligned} N_A &= k_y(y_{AG} - y_{AI}) = k_G(p_{AG} - p_{AI}) \\ &= 12.5(0.01316 - 0.0056) = 0.0945 \text{ lbmol/h} \cdot \text{ft}^2 \end{aligned}$$

**OVERALL MASS TRANSFER COEFFICIENT**

It is generally more convenient to utilize an overall coefficient for the gas and liquid phases rather than the individual coefficients since it is not possible to measure the partial pressure and concentration at the interface ( $p_{AI}$  or  $y_{AI}$ , and  $c_{AI}$  or  $x_{AI}$ , respectively). The preferred procedure is to express the overall coefficient in terms of the individual coefficients. For this approach, it is common to employ overall mass transfer coefficients based on the overall driving force between  $p_{AG}$  (or  $y_{AG}$ ) and  $c_{AL}$  (or  $x_{AL}$ ). The overall coefficients may be defined on the basis of the gas film,  $K_G$ , or the liquid film  $K_L$ , by the equations

$$N_A = K_G(p_{AG} - p_A^*) = K_y(y_{AG} - y_A^*) \quad (7.30)$$

$$N_A = K_L(c_A^* - c_{AL}) = K_x(x_A^* - x_{AL}) \quad (7.31)$$

where  $p_A^*$  is the partial pressure in equilibrium with  $c_{AL}$ , and  $c_A^*$  is the concentration in equilibrium with  $p_{AG}$ .

Consider again the situation shown in Figures 7.4 and 7.5 using the concentration (mole fraction) driving force. The equilibrium-distribution curve for the system is unique at a fixed temperature and pressure. Then,  $y_A^*$ , since it is in equilibrium with  $x_{AL}$ , is as good a measure of  $x_{AL}$  as  $x_{AL}$  itself, and, moreover, it is on the same basis as  $y_{AG}$ . In this situation, the entire two-phase mass transfer effect may then be determined from expressions such as those given in Equations (7.30) and (7.31). In this manner, the ratio of the resistance of either phase to the total resistance is given by the ratio of the driving force through that phase to the "total" driving force across both films. In effect,

$$\frac{1/k'_y}{1/K_y} = \frac{y_{AG} - y_{AI}}{y_{AG} - y_A^*} \quad (7.32)$$

with a similar equation on the liquid side. This analysis is further expanded in the next three subsections.

## Equimolar Counterdiffusion and/or Diffusion in Dilute Solutions

When equimolar counterdiffusion is occurring, or when the solutions are quite dilute, the following equation applies:

$$N_A = k'_y(y_{AG} - y_{AI}) = k'_x(x_{AI} - x_{AL}) \quad (7.33)$$

From the geometry of Figure 7.5,

$$(y_{AG} - y_A^*) = (y_{AG} - y_{AI}) + (y_{AI} - y_A^*) \quad (7.34)$$

$$(y_{AG} - y_A^*) = (y_{AG} - y_{AI}) + m'(x_{AI} - x_{AL}) \quad (7.35)$$

where  $m'$  is the slope of the chord  $CM$ . Substituting for the concentration differences, one obtains

$$\frac{N_A}{K'_y} = \frac{N_A}{k'_y} + \frac{N_A m'}{k'_x} \quad (7.36)$$

or

$$\frac{1}{K'_y} = \frac{1}{k'_y} + \frac{m'}{k'_x} \quad (7.37)$$

Equation (7.37) demonstrates the relationship between the individual mass transfer coefficients and the overall mass transfer coefficient. The left-hand side of Equation (7.37) can be looked upon as the total resistance based on the overall gas driving force, which is equal to the sum of the gas film resistance ( $1/k'_y$ ) and the liquid film resistance ( $m'/k'_x$ ).

In a similar manner, from the geometry of Figure 7.5,

$$(x_A^* - x_{AL}) = (x_A^* - x_{AI}) + (x_{AI} - x_{AL}) \quad (7.38)$$

The slope between the points  $M$  and  $D$  is

$$m'' = \frac{y_{AG} - y_{AI}}{x_A^* - x_{AI}} \quad (7.39)$$

Then,

$$\frac{y_{AG} - y_{AI}}{m''} = x_A^* - x_{AI} \quad (7.40)$$

and it can be readily shown that

$$\frac{1}{K'_x} = \frac{1}{m''k'_y} + \frac{1}{k'_x} \quad (7.41)$$

As before, the left-hand side of Equation (7.41) is the total resistance and is equal to the sum of the individual resistances.

## Gas Phase Resistance Controlling

Assuming that the numerical values of  $k'_x$  and  $k'_y$  are roughly equal, the importance of the slope of the equilibrium curve chords can readily be demonstrated. However, if  $m'$  is very small, so that the equilibrium curve in Figure 7.5 is nearly flat, then only a very small concentration of  $y_A$  in the gas will give a relatively large value of  $x_A$  in equilibrium with the liquid. This indicates that gas solute  $A$  is very soluble in the liquid phase, and hence, the term  $m'/k'_x$  in Equation (7.37) becomes very small or negligible. Then

$$\frac{1}{K'_y} \approx \frac{1}{k'_y} \quad (7.42)$$

and the major resistance is said to be in the gas phase, or the “gas phase is controlling.” Also,

$$y_{AG} - y_A^* \approx y_{AG} - y_{AI} \quad (7.43)$$

Under such circumstances, even fairly large percentage changes in  $k'_x$  will not significantly affect  $K'_y$ , and efforts to increase the rate of mass transfer would best be directed toward decreasing the gas-phase resistance, e.g., by increasing the gas phase turbulence or using equipment that specifically will have a high turbulence in the gas phase.

## Liquid Phase Resistance Controlling

In a similar manner, when  $m''$  is very large or the solute  $A$  is very insoluble in the liquid, with  $k'_x$  and  $k'_y$  again very roughly equal, then the term  $1/(m''k'_y)$  becomes very small and

$$\frac{1}{K'_x} \approx \frac{1}{k'_x} \quad (7.44)$$

The major resistance to mass transfer is then in the liquid, the “liquid phase is controlling,” and

$$x_A^* - x_{AL} \approx x_{AI} - x_{AL} \quad (7.45)$$

In such cases, efforts to affect large changes in the rate of mass transfer are best directed toward conditions influencing the liquid coefficient,  $k'_x$ , i.e., increasing the turbulence in the liquid phase.

For cases where  $k'_x$  and  $k'_y$  are not nearly equal, Figure 7.5 shows that it will be the relative size of the ratio  $k'_x/k'_y$  and of  $m'$  (or  $m''$ ) which will determine the location of the controlling mass transfer resistance.

Table 7.4 lists some cases of specific films controlling a particular mass transfer operation.<sup>(9,10)</sup>

**Table 7.4** Controlling Films for Various Systems**Gas film**

1. Absorption of ammonia in water
2. Absorption of ammonia in aqueous ammonia
3. Stripping of ammonia from aqueous ammonia
4. Absorption of water vapor in strong acids
5. Absorption of sulfur trioxide in strong sulfuric acid
6. Absorption of hydrogen chloride in water
7. Absorption of hydrogen chloride in weak hydrochloric acid
8. Absorption of 5 vol% ammonia in acids
9. Absorption of sulfur dioxide in alkali solutions
10. Absorption of sulfur dioxide in ammonia solutions
11. Absorption of hydrogen sulfide in weak caustic
12. Evaporation of liquids
13. Condensation of liquids

**Liquid film**

1. Absorption of carbon dioxide in water
2. Absorption of oxygen in water
3. Absorption of hydrogen in water
4. Absorption of carbon dioxide in weak alkali
5. Absorption of chlorine in water

**Both gas and liquid film**

1. Absorption of sulfur dioxide in water
2. Absorption of acetone in water
3. Absorption of nitrogen oxide in strong sulfuric acid

## Experimental Mass Transfer Coefficients

In many instances, investigators have found that their data correlate well on the basis of some empirical relationship quite different from the foregoing formalized treatment, and that these relationships are useful in design work for systems to be reviewed in Part Two. Examples of such relationships are provided in the literature along with other important data pertaining to the reported experimental studies.<sup>(11)</sup>

Experimental data are often correlated in terms of dimensionless numbers such as the Schmidt number ( $Sc = \mu/\rho D_{AB}$ ) and the Reynolds's number ( $Re = Lv\rho/\mu$ ). In the absence of experimental mass transfer data, many correlations are available which may be used to estimate the mass transfer coefficient for the system being studied. In practice, when choosing a correlation, one should make every effort to match as closely as possible the system conditions under which the correlation was formulated. Some of the correlations, primarily applicable in gas absorption mass transfer processes, are available in the literature.<sup>(2,4,5)</sup> In general, they apply to absorption columns loaded with various types of packing. These various types of absorption columns and the different packings utilized will be discussed in Chapter 10.

**ILLUSTRATIVE EXAMPLE 7.6**

The absorption of ammonia into water from an air–ammonia mixture was studied at 40°F and at a total pressure of 2.0 atm. The average value of  $k'_y$  was estimated to be 0.40 lbmol/h · ft<sup>2</sup> · mol fraction and that of  $k'_L$  to be 1.10 lbmol/h · ft<sup>2</sup> · mol/ft<sup>3</sup>. The equilibrium partial pressure of ammonia was approximated by Henry's law to be

$$p = 0.246c$$

where  $p$  is the partial pressure of ammonia (mm Hg) and  $c$  is the molar concentration of ammonia in the liquid (lbmol/ft<sup>3</sup>). Estimate the overall gas mass transfer coefficient in lbmol/h · ft<sup>2</sup> · atm.

**SOLUTION:** First, convert  $k'_y$  to a pressure basis in atm. From Equations (7.23) and (7.24), and noting that  $p_i = y_i P$

$$k'_G = \frac{k'_y}{P} = \frac{0.40}{2.0} = 0.20 \text{ lbmol/h} \cdot \text{ft}^2 \cdot \text{atm}$$

One may now modify and apply Equation (7.37) to obtain  $K'_G$ . Note the equilibrium coefficient 0.246 essentially represents Henry's constant,  $m'$ .

$$\frac{1}{K'_G} = \frac{1}{k'_G} + \frac{m'}{k'_L} = \frac{1}{0.20} + \frac{0.246}{1.10} = 5.224$$

Therefore,

$$K'_G = \frac{1}{5.224} = 0.1914 \text{ lbmol/h} \cdot \text{ft}^2 \cdot \text{atm} \quad \blacksquare$$

**ILLUSTRATIVE EXAMPLE 7.7**

Refer to Illustrative Example 7.6 and determine the relative magnitude of the resistances of the gas and liquid phases.

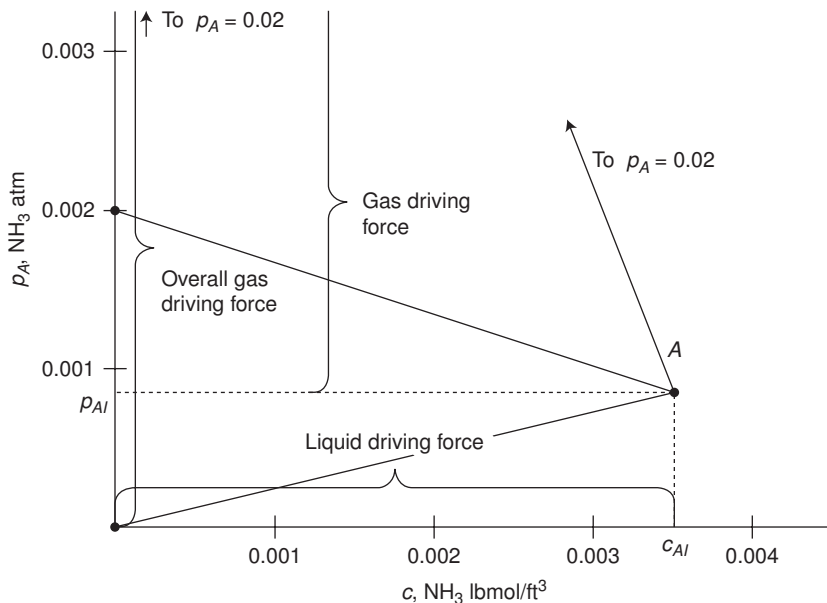
**SOLUTION:** Since  $1/k'_G = 5.0$ , the relative magnitude of the gas phase resistance is the overall resistance,  $R$ , is

$$\frac{1/k'_G}{1/K'_G} = R = \frac{5.0}{5.224} = 0.957 = 95.7\%$$

The relative resistance of the liquid is therefore 4.3%. ■

**ILLUSTRATIVE EXAMPLE 7.8**

In Illustrative Example 7.6, the average gas concentration at one point in the equipment is 1% ammonia by volume, and the water is pure. For this condition, calculate the interfacial composition for both phases, draw to scale a graph of  $p$  vs  $c$ , plot the equilibrium curve, the point representing the gas and liquid compositions, and the point representing the interface compositions. Mark the driving forces in each phase and the overall gas phase driving force. Compute the rate of absorption, in lbmol NH<sub>3</sub>/h · ft<sup>2</sup> using  $k_G$ ,  $k_L$ , and  $K_G$ .



**Figure 7.7** Figure for Illustrative Example 7.8.

**SOLUTION:** Start with Equations (7.22) and (7.23), noting that for a dilute solution  $k_G = k'_G$  and  $k_L = k'_L$

$$N_A = k_G(p_{AG} - p_{AI}) = k_L(c_{AI} - c_{AL})$$

with  $p_{AG} = 0.01(2.0) = 0.02$  atm and  $c_{AL} = 0$ : substituting yields

$$0.20(0.02 - p_{AI}) = 1.10(c_{AI} - 0)$$

In addition, at the interface (see previous Illustrative Example)

$$p_{AI} = 0.246c_{AI}$$

The solution of these two equations, or the intersection (point A) of the two lines in Figure 7.7, leads to

$$p_{AI} = 0.000856 \text{ atm}$$

From the equilibrium relation,

$$c_{AI} = \frac{p_{AI}}{0.246} = \frac{0.000856}{0.246} = 0.00348 \text{ lbmol/ft}^3$$

The graph is provided in Figure 7.7.

The rate of absorption may be calculated from any one of three equations:

$$N_A = k_G(p_{AG} - p_{AI}) = 0.20(0.0191) = 0.00383 \text{ lbmol/h} \cdot \text{ft}^2$$

$$N_A = k_L(c_{AI} - c_{AL}) = 1.10(0.00348) = 0.00383 \text{ lbmol/h} \cdot \text{ft}^2$$

$$N_A = K_G(p_{AG} - p_A^*) = 0.19(0.02) = 0.00383 \text{ lbmol/h} \cdot \text{ft}^2$$

■

**ILLUSTRATIVE EXAMPLE 7.9**

Compare the results generated for the flux from the three equations.

**SOLUTION:** As can be seen, and as expected, there is excellent agreement. ■

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