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Mass Transfer with Chemical Reaction in Liquid Foam Reactors

Mass transfer studies were conducted in a stable liquid foam reactor under various operating conditions to evaluate gas holdup, effective interfacial area, liquid-phase mass transfer coefficient and a modified interfacial mass transfer coefficient to include the surface-active agents employed. Gas holdup and effective interfacial area were evaluated experimentally. The interfacial mass transfer coefficient was evaluated semitheoretically, by considering the interfacial region as a separate phase and using the experimental data developed for mass transfer accompanied by a fast first-order chemical reaction. The liquid-phase mass transfer coefficient was also evaluated semitheoretically, using Danckwert's theory for the liquid phase and the experimental data on mass transfer accompanied by a slow pseudofirst-order chemical reaction.

An experimental unit was set up to provide a stable flowing foam column, simulating the foam reactor. Mass transfer rates were studied for superficial gas velocities in the range from 1.5×10^{-2} m/s to 5×10^{-2} m/s, giving gas residence times in the range from 20 to 55 seconds. A cationic and nonionic surface-active agent and three different wire mesh sizes, giving bubble size distributions in the range from 2.2 to 5.4 mm Sauter mean diameters, were employed.

It is observed that gas holdup is insensitive to the type of surface-active agent; it is however, dependent on wire mesh size and gas velocity. The bubble diameter and, hence, the interfacial area are found to be insensitive to gas velocity in the range studied; they are, however, strong functions of wire mesh size. The liquid-phase mass transfer coefficient increases with increase in gas velocity. The surface-active agent introduces additional resistance to mass transfer in both reaction cases, this being the controlling one in the case of the fast reaction. A comparison with conventional packed bed contactors indicates the mass transfer rates to be about 8 times lower for the foam reactor, for the fast reaction case; for slow reactions, the foam reactor has mass transfer rates approximately 2-4 times higher than those for conventional packed bed contactors.

SCOPE

The foam bed reactor is rather new. It offers a very high gas-liquid interfacial area per unit volume of the bed, a large contact time, a fairly low-pressure drop—all these characteristics are very desirable for treatment of large quantities of lean gases, as in the case of contaminant separation from toxic gas streams.

Gas holdup, interfacial area, and mass transfer coefficients are the main parameters determining the mass transfer rates in gas-liquid absorbers and reactors. In foam reactors, usually surface-active materials are present. The presence of surface-

active materials, however, could reduce mass transfer rates significantly, by introducing an interfacial resistance and by altering the hydrodynamics.

The literature available on the application of foam as a medium for gas absorption and reactions is scarce. In the studies reported, no attempts were made to evaluate the mass transfer parameters encountered under various operating conditions in foam bed reactors. The objective of the present work is to evaluate gas holdup, interfacial area, liquid-phase mass transfer coefficient and the interfacial resistance due to surfactant in the form of a modified interfacial mass transfer coefficient, in foam reactors under various operating conditions.

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CONCLUSIONS AND SIGNIFICANCE

The foam reactor offers a very high gas-liquid interfacial area per unit volume of reactor. The interfacial area is about 8 to 10 times higher than that for conventional packed towers, plate towers, and bubble columns. The advantage of high interfacial area is, however, partly or completely lost by introduction of an interfacial resistance in the form of a surfactant film. For fast chemical reactions, say between carbon dioxide and sodium hydroxide, the interfacial resistance appears to become the controlling factor, and hence the advantage of higher interfacial area in a foam reactor is completely lost. Thus, for fast chemical reactions, foam reactors do not appear suitable. For slow chemical reactions, say between carbon dioxide and sodium carbonate-bicarbonate solution, the advantage of increased interfacial area appears to be lost only partially and foam re-

actors could thus become useful. In this case, a mass transfer rate about 2 to 4 times higher than that in conventional absorbers is obtained. Also, due to the large contact time available in foam reactors, slow reactions can conveniently be carried out in such units. Higher operating costs due to the use of surfactants, however, may outweigh the advantage of high mass transfer rates. When the cost of surfactant is reduced and the effect of surfactant on mass transfer rate is less significant, the foam reactor provides an extremely promising alternative for applications involving slow reactions. The mass transfer parameters determined in the present work could become applicable in the design of foam reactors and in comparing foam reactors with other gas-liquid contacting units.

INTRODUCTION

Present-day gas-liquid contactors have probably reached very nearly their best performance. It would seem that any major advance should come through a new design rather than through improvements on existing design. The foam bed contactor is rather new for gas-liquid operations. It offers a very high gas-liquid interfacial area per unit volume of the bed, a large contact time, and a fairly low-pressure drop all these characteristics being very desirable for treatment of large quantities of lean gases, as in the case of contaminant removal from toxic gas streams (Metzner and Brown, 1956; Biswas and Kumar, 1981; and Johansson and Brander, 1980). It is reported in the literature that a foam bed can be effective for the separation of fine particulates from industrial and toxic gas emissions (Damle, 1981; Damle and Mahalingam, 1982; Kaldor and Phillips, 1976). Thus, a foam reactor/scrubber could become attractive for the combined separation of both gaseous and particulate pollutants.

For producing a stable foam phase, the presence of surface-active materials is essential. The presence of surface-active materials, however, could reduce mass transfer rates significantly, by introducing an interfacial resistance and by altering the hydrodynamics.

EFFECT OF SURFACTANT ON MASS TRANSFER

The presence of surfactants at a gas-liquid interface significantly affects the rate of transfer of a solute from one phase to another, by introducing both hydrodynamic and physico-chemical effects. The important hydrodynamic effect is that the surfactants increase the rigidity of the interface and reduce interfacial turbulence. It is known that interfacial turbulence considerably enhances mass transfer rates (Sherwood et al., 1975). The physico-chemical effect is that the surfactants contribute their own intrinsic resistance to mass transfer, by changing the nature of the properties at the interface.

Springer and Pigford (1970), using a frequency response analysis, established that surfactants reduce interfacial turbulence. Similar conclusions about the suppression or elimination of surface turbulence have been reached by others (Goodridge and Robb, 1965; Davies and Rideal, 1961). Thompson (1970) was able to restrict surface motion by applying fine wire mesh screens to the liquid surface; in spite of this, he observed a considerable decrease in mass transfer rates in the presence of surfactants. Similar conclusions about the nonhydrodynamic interfacial mass transfer resistances caused by surfactants have been reached by Caskey and Barlage (1972), Harvey and Smith (1959), and Goodridge and Bricknell (1962).

Of the several models proposed to account for the interfacial resistance due to surfactant, the model proposed by Nguyen Ly et al. (1979) seems preferable to the other models. This is because their model is relatively simple, similar to those for simple diffusion in biological or polymer membranes and consistent with the experimental data reported in the literature (Emmert and Pigford, 1954; Plevan and Quinn, 1966; and Whitaker and Pigford, 1966). They assumed that the interface and the bulk liquid are separate phases. The interfacial region of surfactant film is assumed to be of finite thickness which obeys local equilibrium conditions, has a capacity for the dissolved gases greater than the solubility in water, and contributes to a diffusion coefficient three to four orders of magnitude lower than the bulk aqueous solution. In gas-liquid operations, the assumption of local equilibrium between the interfacial region and the bulk phase seems preferable to assumptions of local nonequilibrium (Sherwood et al., 1975). The equilibrium solubility of gas in the surfactant film has a precedent in models of mass transfer across biological membranes.

Interfacial resistance and hydrodynamic effects due to surfactants can be an important factor in the design of mass transfer equipment. This is particularly true in instances where the gas-side and liquid-side diffusional resistances are small as in the case of absorption accompanied by a fast chemical reaction.

ACTUAL MASS TRANSFER RATES IN THE FOAM BED REACTOR

The use of liquid foam as medium for gas absorption is of recent origin. Since foam has traditionally been considered a nuisance in chemical process industries, not much study has been reported in the literature for this application, except for a couple of articles.

Metzner and Brown (1956), and Johansson and Brander (1980) studied mass transfer rates in the foam bed reactor. Biswas and Kumar (1981) studied the mass transfer rates in an unstable foam bed reactor. They developed a model based on the idealized regular dodecahedral structure of the foam bed in order to predict the extent of chemical reaction. In all these studies, however, no attempts were made to evaluate the important mass transfer parameters. The objective in the present work is to evaluate the relevant mass transfer parameters.

THE PROCESS

The gas-liquid mass transfer process in liquid foam reactors may be divided into three steps.

1. *Foam Formation Stage.* The gas-phase containing the solute is bubbled through a pool of liquid containing the surfactant in

sufficient concentration as to produce a stable and persistent foam.

2. *Stable Foam Stage.* After foam formation, the foam flows up the column under a pressure gradient.

3. *Foam Breakup Stage.* After a predetermined height of travel, the foam is broken up by chemical or mechanical means. Here, the gas phase is separated from the foamate liquid phase. The foamate liquid may then be recycled.

Mass transfer takes place during the foam formation stage and during the stable foam stage. The foam breakup is so rapid that the gas and liquid phases separate without any further transfer of mass. The main focus of the present work is to evaluate the phenomenon of mass transfer with chemical reaction during the stable foam stage.

The following parameters are evaluated for the stable foam stage: gas holdup, gas-liquid interfacial area, interfacial resistance in the form of modified interfacial mass transfer coefficient, and liquid phase mass transfer coefficient.

To evaluate modified interfacial and liquid-phase mass transfer coefficients, the following two gas-liquid systems are considered:

Absorption of Carbon Dioxide in Sodium Hydroxide Solution

As established by Danckwerts and Sharma (1966), the reaction of carbon dioxide with hydroxide follows a pseudofirst-order reaction, the condition to be satisfied is:

$$\sqrt{D_A k_2 C_B} \leq \frac{1}{2} k_L (1 + C_B / b C_A^*) \quad (1)$$

Danckwerts and Sharma (1966) have shown that when condition 1 is satisfied, the rate of absorption per unit interfacial area for the liquid phase using Danckwerts surface renewal theory is given approximately by

$$R = C_A^* \sqrt{k_L^2 + D_A k_2 C_B} \quad (2)$$

This relationship also holds for any other type of chemical system in which the reaction is first order or pseudofirst order. When condition 2 is satisfied, and also when

$$\sqrt{D_A k_2 C_B} \geq 5 k_L \quad (3)$$

then the rate of absorption is given approximately by

$$R = C_A^* \sqrt{D_A k_2 C_B} \quad (4)$$

and the rate of absorption per unit interfacial area, thus, is independent of k_L .

Absorption of Carbon Dioxide in Sodium Carbonate-Bicarbonate Solution

Danckwerts and Sharma (1966) have shown that, under certain conditions, carbon dioxide undergoes a pseudofirst-order chemical reaction with the carbonate-bicarbonate buffer. The condition to be satisfied is

$$C_A^* \left(\frac{1}{C_C} + \frac{2}{C_D} \right) \left[\sqrt{1 + \frac{D_A k_2 C_B}{k_L^2}} - 1 \right] \ll 1 \quad (5)$$

When this reaction is slow and reversible, the rate of absorption per unit area for the liquid phase is given by

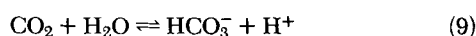
$$R = (C_A^* - C_{Ab}) \sqrt{k_L^2 + D_A k_2 C_B} \quad (6)$$

where

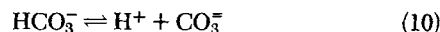
$$C_{Ab} = (K_2 C_D^2) / (K_1 C_C) \quad (7)$$

$$C_B = (K_W C_C) / (K_2 C_D) \quad (8)$$

K_W is the ion product for water; K_1 is the equilibrium constant for the reaction.



and K_2 is the second ionization constant for the reaction



The above gas-liquid system forms the basis for the determination of the liquid-phase mass transfer coefficient; as in this case, the first and second term inside the square root sign of Eq. 6 are of comparable magnitude.

GAS HOLDUP

The gas holdup ϵ_g in the stable foam stage can be determined as

$$\epsilon_g = (E - 1) / E \quad (11)$$

where E is the foam expansion ratio and is defined as the ratio of the total volume of the foam to the total volume of the liquid contained within the foam.

GAS-LIQUID INTERFACIAL AREA

The gas-liquid interfacial area using a physical method can be determined as

$$a = 6 \epsilon_g / d_s \quad (12)$$

where d_s is the Sauter mean bubble diameter and is given as

$$d_s = \sum N_j d_{b,j}^3 / \sum N_j d_{b,j}^2 \quad (13)$$

DETERMINATION OF MODIFIED INTERFACIAL AND LIQUID-PHASE MASS TRANSFER COEFFICIENTS

The modified interfacial mass transfer coefficient stated above is the product of the interfacial mass transfer coefficient and the Henry's constant for solute in the surfactant film. The assumptions made in the determination of both mass transfer coefficients are:

(i) The interface is treated as a separate phase of finite thickness in which gas can both dissolve in and diffuse through. Thus, it is assumed that the system consists of three phases: gas, liquid, and a surfactant interface.

(ii) Local equilibrium solubility conditions exist at either side of the surfactant film: a Henry's constant H_s for the gas-surfactant interface, and a distribution coefficient K at the plane between the interfacial region and the bulk phase.

(iii) Danckwerts surface renewal model holds for the liquid phase.

Modified Interfacial Mass Transfer Coefficient

Figure 1 illustrates a three phase—gas, interfacial and liquid—equilibrium model. Figure 2 shows the overall gas-liquid absorption process in a foam reactor. For a fast pseudofirst-order reaction between carbon dioxide and hydroxide ions, at steady state, the absorption process taking place in a differential volume $A \cdot dZ$ of stable foam stage can be represented by following set of equations.

$$-\frac{G_i}{A} \frac{dm}{dZ} = k_g a (P_A - P_{Ai}) \quad (14)$$

$$= k_{Ia} (C_{Ai} - C_A^*) \quad (15)$$

$$= a C_A^* \sqrt{D_A k_2 C_B} \quad (16)$$

where G_i is the molar flow rate of inerts, m is the mole of solute per mole of inerts, $k_g a$, k_{Ia} are the volumetric gas-side and interfacial mass transfer coefficients, respectively. P_A is the partial pressure of solute gas. From the assumption of local equilibrium solubility at either side of the film, the following relations are obtained

$$C_{Ai} = H_s P_{Ai} \quad (17)$$

$$C_A^* = K C_{Ai} = (H C_{Ai}^*) / H_s \quad (18)$$

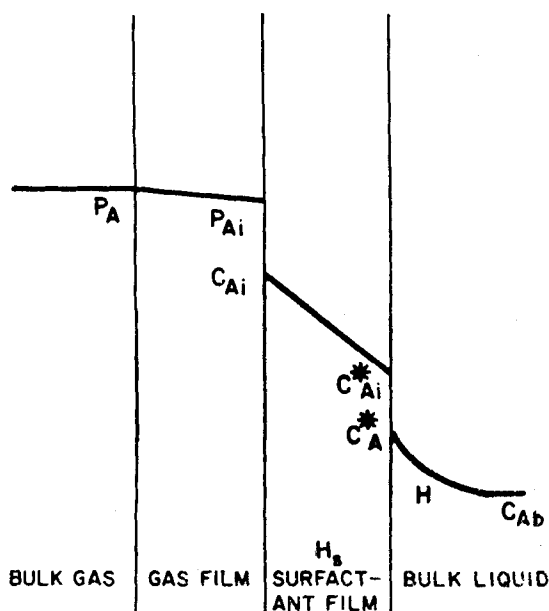


Figure 1. Diagram of the three-phase model.

where H is Henry's constant for direct contact of the gas with the liquid.

From Eqs. 14 through 18, expressing C_A^* in terms of measurable quantity P_A , substituting in Eq. 16, and rearranging, we get

$$-\frac{G_i}{A} \frac{dm}{dZ} = \frac{P_A}{\frac{1}{k_{ga}} + \frac{1}{k_{fa}H_s} + \frac{1}{aH\sqrt{D_A k_2 C_B}}} \quad (19)$$

The partial pressure of solute A is related to m and the total pressure P by the relation

$$P_A = mP/(m + 1) \quad (20)$$

The concentration of hydroxide at any position Z can be expressed in terms of m by taking an overall material balance up to the differential element as

$$G_i(m_1 - m) = L_o(C_{B1} - C_B)/b \quad (21)$$

or

$$C_B = C_{B1} - G_i b(m_1 - m)/L_o \quad (22)$$

where subscript 1 refers to the condition at the beginning of stable foam stage, n is a stoichiometric factor (for reaction between carbon dioxide and hydroxide it is 2), and L_o is the liquid flow rate through the stable foam stage.

Substituting Eqs. 20 and 22 into Eq. 19, and integrating between the limits $Z = Z_i$, $m = m_1$ and ZH_T , $m = m_o$, after some rearrangement, we get

$$\frac{1}{k_{ga}} + \frac{1}{k_{fa}H_s} = \{APA_4(H_T - Z_i)/G_i - 2(\sqrt{C_{B1}} - \sqrt{C_{Bo}})/A_1 - \ln A_3/(\sqrt{A_2})\}/\{A_4[m_1 - m_o + \ln(m_1/m_o)]\} \quad (23)$$

When gas-side resistance is negligible, i.e., $k_g \gg k_{fa}H_s$, $k_{fa}H_s$ can be obtained as

$$k_{fa}H_s = \frac{A_4\{m_1 - m_o + \ln(m_1/m_o)\}}{\frac{APA_4(H_T - Z_i)}{G_i} - \frac{2(\sqrt{C_{B1}} - \sqrt{C_{Bo}})}{A_1} - \frac{\ln A_3}{\sqrt{A_2}}} \quad (24)$$

where

$$A_1 = G_i b/L_o \quad (25)$$

$$A_2 = C_{B1} - A_1 m_1 \quad (26)$$

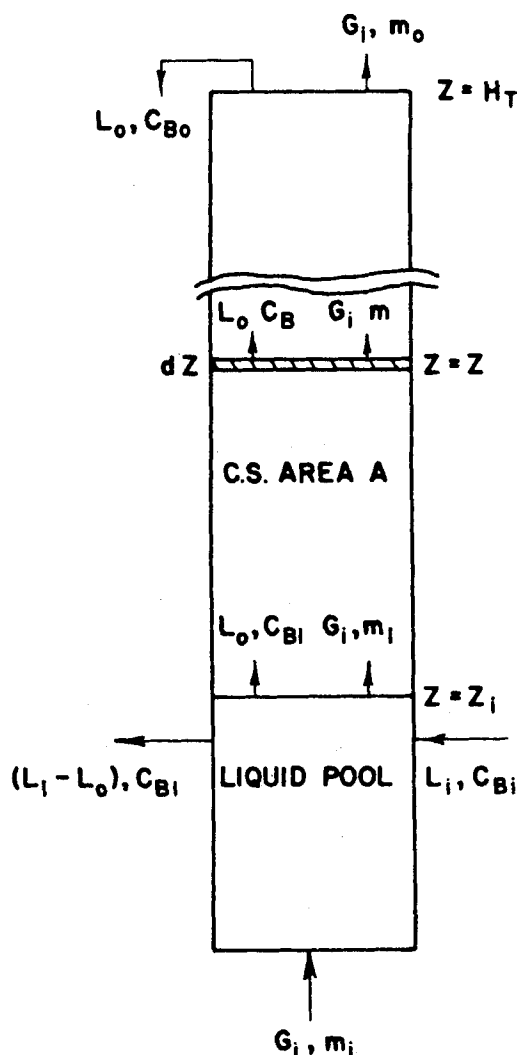


Figure 2. Gas-liquid absorption process in a foam reactor.

$$A_3 = \frac{(\sqrt{C_{B1}} - \sqrt{A_2})(\sqrt{C_{Bo}} + \sqrt{A_2})}{(\sqrt{C_{B1}} + \sqrt{A_2})(\sqrt{C_{Bo}} - \sqrt{A_2})} \quad (27)$$

$$A_4 = aH\sqrt{D_A k_2}, \text{ and} \quad (28)$$

subscript o refers to the condition at the outlet. The modified interfacial mass transfer coefficient $k_f H_s$, is then written as

$$k_f H_s = k_{fa} H_s / a \quad (29)$$

The overall material balance gives C_{B1} in terms of known conditions at inlet and at outlet of the reactor as

$$C_{B1} = \{L_i C_{Bi} - L_o C_{Bo} - b G_i (m_1 - m_o)\} / (L_i - L_o) \quad (30)$$

A material balance across foam formation stage then gives m_1 as

$$m_1 = m_i - L_i (C_{Bi} - C_{B1}) / (b G_i) \quad (31)$$

The diffusivity of carbon dioxide in mixed solutions of sodium hydroxide and sodium carbonate is given by the equation proposed by Hikita et al. (1976) as

$$D_A = D_{Aw} [1 - (a_1 C_B + a_2 C_C)] \quad (32)$$

where a_1 and a_2 are constants for the sodium hydroxide and sodium carbonate solutions; they were obtained from our experimental viscosity data as 0.129 and 0.261 m³/kmol, respectively.

The diffusivity of carbon dioxide in water, D_{Aw} , is given by Danckwerts and Sharma (1966), as

$$\log D_{Aw} = -8.1764 + 712.5/T - 2.59 \times 10^5/T^2 \quad (33)$$

where D_{Aw} is in m^2/s and T in K. Base of logarithm is 10 and is to be assumed as such in all cases unless specified otherwise.

Henry's constant H for carbon dioxide in aqueous solution of mixed electrolyte is given as (Danckwerts and Sharma, 1966)

$$\log (H/H_w) = -(h_1 I_1 + h_2 I_2) \quad (34)$$

where h_1, h_2 are salting out parameters with ionic strengths I_1 and I_2 . For sodium hydroxide and sodium carbonate with carbon dioxide as solute gas, the salting out parameters are 0.138 and 0.093 m^3/kg ion at 298 K, respectively.

Henry's constant, H_w , for carbon dioxide in water is given as (Haq, 1982)

$$\log H_w = 4.117 - 0.059T + 7.885 \times 10^{-5} T^2 \quad (35)$$

where H_w is in $\text{kmol}/(\text{m}^3 \cdot \text{Pa})$ and T in K.

The rate constant k_2 as a function of temperature and ionic strength of the solution is given by the relation (Astarita, 1967)

$$\log k_2 = 13.635 - 2,895.0/T + 0.13I \quad (36)$$

The physico-chemical constants are evaluated using average values of the concentrations of sodium hydroxide and sodium carbonate, in the stable foam regime.

Liquid-Phase Mass Transfer Coefficient

With the assumptions (i) through (iii) and also when the condition given by Eq. 5 is satisfied, at steady state, the absorption of carbon dioxide in a solution of carbonate-bicarbonate buffer containing a surfactant is given by the following equations when the gas phase resistance is assumed to be negligible.

$$-\frac{G_i}{A} \frac{dm}{dz} = k_1 a (C_{Ai} - C_{Ai}^*) \quad (37)$$

$$= a (C_A^* - C_{Ab}) \sqrt{D_a k_2 C_B + k_L^2} \quad (38)$$

and

$$C_{Ai} = H_s P_A = H_s m P / (m + 1) \quad (39)$$

$$C_A^* = H C_{Ai} / H_s \quad (40)$$

$$C_B = (K_w C_C) / (K_2 C_D) \quad (41)$$

where H_s is Henry's constant for solute in the interfacial region and H is Henry's constant for solute in direct contact with the bulk liquid.

From Eqs. 37 to 41, expressing C_A^* in terms of m and substituting in Eq. 38, after some rearrangement, we get

$$-\frac{dm}{dz} = \frac{aA[Pm/(m+1) - C_{Ab}/H]}{G_i \left(\frac{1}{k_1 H_s} + \frac{1}{B_1 H} \right)} \quad (42)$$

where

$$B_1 = \sqrt{D_a k_2 C_C / C_D + k_L^2} \quad (43)$$

$$k = (k_2 K_w) / K_2 \quad (44)$$

$$C_{Ab} = (K_2 C_D^2) / (K_1 C_C) \quad (45)$$

The first and second terms in the denominator of Eq. 42 represent the interfacial and liquid-phase resistances, respectively.

By taking material balances from the beginning of stable foam stage up to the differential element, concentration of carbonate and bicarbonate can be expressed in terms of m as

$$C_C = C_{C1} - G_i(m_1 - m)/L_o \quad (46)$$

$$C_D = C_{D1} + 2G_i(m_1 - m)/L_o \quad (47)$$

The concentrations of carbonate C_{C1} , bicarbonate C_{D1} and the moles of carbon dioxide per mole of inerts m_1 , at the beginning of stable foam stage can be obtained by performing overall material balances for each of them as:

$$C_{C1} = [L_i C_{Ci} - L_o C_{Co} - G_i(m_i - m_o)] / (L_i - L_o) \quad (48)$$

$$C_{D1} = C_{D1} + 2(C_{Ci} - C_{C1}) \quad (49)$$

and

$$m_1 = m_i - L_i(C_{Ci} - C_{C1})/G_i \quad (50)$$

where subscript i refers to conditions at inlet and o refers to those at outlet.

The equations for relevant physico-chemical data are:

The solubility H is given by Eqs. 34 and 35, with the salting out parameters for carbonate and bicarbonate being 0.093 m^3/kg ion and 0.093 m^3/kg ion, respectively.

The diffusivity D_A is given as

$$D_A = D_{Aw}[1 - (a_2 C_C + a_3 C_D)] \quad (51)$$

where $a_2 = 0.261 \text{ m}^3/\text{kmol}$ and $a_3 = 0.140 \text{ m}^3/\text{kmol}$ are found from viscosity data (Hikita et al., 1976). D_{Aw} is given by Eq. 33.

For sodium carbonate-bicarbonate solution having ionic strength between 1 and 2 kmol/m^3 , $k (= k_2 K_w / K_2)$ in the temperature range 293–303 K is given as (Sherwood et al., 1975)

$$\log k = 12.047 - 3,607.01/T \quad (52)$$

where k is in s^{-1} and T in K. K_2/K_1 at infinite dilution is given by Danckwerts and Sharma (1966) as

$$\log (K_2/K_1) = 505.3/T - 8.345 + 0.009 T \quad (53)$$

where T is in K.

The boundary conditions for the differential equation (Eq. 42) are

$$z = Z_i, \quad m = m_1 \quad (54)$$

and

$$z = H_T, \quad m = m_o \quad (55)$$

The determination of k_L from Eqs. 42 through 47 and 51 through 55 involves a numerical integration and an iterative procedure. The following stepwise procedure is employed to determine k_L :

(1) From a knowledge of the range of k_L values, an initial guess for k_L is made.

(2) Using the value of k_L , numerically integrate the differential equation (Eq. 42) with boundary condition (Eq. 54) using, for example, a fourth order Runge-Kutta method. At each increment in Z , calculate the concentration of carbonate, bicarbonate and relevant physico-chemical data using the equations given above. Repeat the procedure until $Z = H_T$ is reached.

(3) Compare the value of m obtained at $Z = H_T$ using guess value of k_L , say m_{cal} , with the experimental value m_o .

(4) If the absolute difference between m_{cal} and m_o is acceptable, the assumed k_L is the solution. If not, then perform Step 5.

(5) Using a convergence method, such as the method of bisection, make a new and better guess for k_L . Repeat Steps 2 to 5 until convergence is achieved.

Once k_L is known, check the condition for pseudofirst order given by Eq. 5. Knowing k_L , mass transfer rates and the relative magnitude of interfacial and liquid-phase resistances can also be calculated at each point along the reactor length.

EXPERIMENTAL

The flow diagram of the experimental setup is given in Figure 3. The foam reactor was a vertical glass column 25 mm ID and 0.91 m tall. The bottom of the column was a glass cone, with a wire-mesh gas distributor placed at the flange connection. The top of the column was connected to a foam disengagement chamber. Two pressure taps at a distance of about 0.6 m were provided on the column to measure the pressure drop in the flowing foam. The details on the foam reactor are given in Figure 4.

Compressed air from mains was filtered and allowed to flow through rotameter R_1 . Compressed carbon dioxide from a cylinder flowed through rotameter R_2 . The flow rate of air and carbon dioxide and the pressure in the gas line were controlled by valves V_1, V_2 and V_3 . Normally a pressure

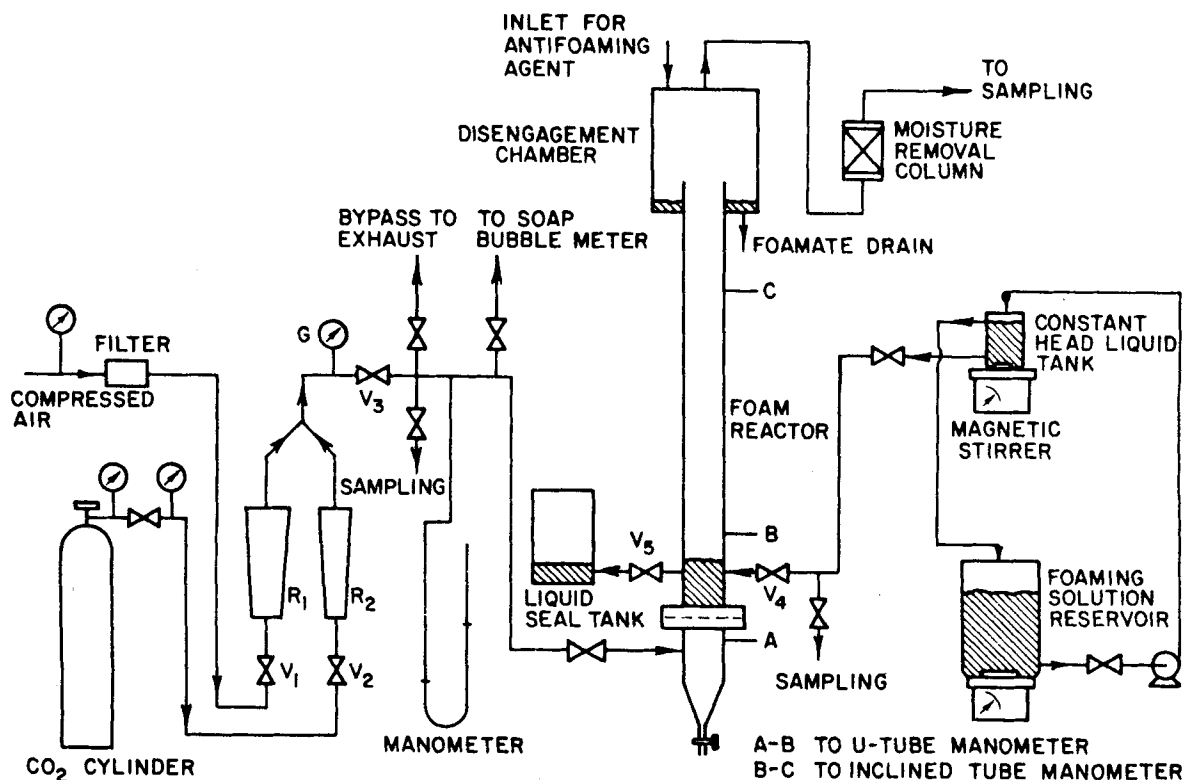


Figure 3. Schematic diagram of experimental setup.

of 10–15 psig (69–103 kPa) was maintained in the gas line. Valve V_3 was used to reduce this pressure to almost atmospheric. This throttling ensured good mixing between air and carbon dioxide. The gas flow rate was measured precisely by a soap bubble meter. U-tube manometers were used to measure the pressure of gas entering the reactor, and the pressure drop across the wire mesh and the liquid pool. A liquid pool of about 0.06 m height was maintained in all cases so as to minimize the absorption of carbon dioxide in foam formation stage. An inclined tube manometer was used to measure the small pressure drop encountered in the flowing foam. A

column containing drierite was provided to remove moisture from the outlet gas before being sampled. The recycled liquid was collected in a tank under a liquid seal.

SAMPLING AND ANALYSES

The liquid streams entering and leaving the reactor were analyzed, either for sodium hydroxide and sodium carbonate for absorption of carbon dioxide in sodium hydroxide solution or for sodium carbonate and sodium bicarbonate for absorption in sodium buffer solution, using standard volumetric procedure (Kolthoff and Sandler, 1952).

The gas streams entering and leaving the reactor were analyzed for carbon dioxide, either with a Carle analytical gas chromatograph using a Carbosieve B column and a thermal conductivity detector (TCD) for absorption of carbon dioxide in sodium buffer solution or with a Beckman model 864-A nondispersive infrared (NDIR) analyzer for absorption of carbon dioxide in sodium hydroxide solution.

Carbon dioxide concentration in air at inlet was kept around 10 mol % in all cases. For absorption of carbon dioxide into hydroxide solution, the concentration of hydroxide at the inlet was kept around 1 kmol/m³ so as to satisfy the condition for pseudo first order reaction. For absorption into buffer solutions, concentrations of approximately 0.6 kmol/m³ carbonate and 0.15 kmol/m³ of bicarbonate at the inlet were chosen so as to satisfy the condition for pseudofirst-order reaction and to keep the total ionic strength between 1 and 2 kmol/m³.

The variables studied were gas flow rate, wire mesh size, and type of surfactant. Gas velocities in the range from 0.015 to 0.05 m/s were used. Three different wire mesh sizes, 20, 60, and 100 mesh, were used to provide different bubble sizes. The surfactants used were: a) nonionic surfactant, Triton X-100 as a 1% v/v solution in water containing the reactant; b) cationic surfactant, hexadecyl trimethyl ammonium bromide (HDTMAB) as a 0.5% w/w solution in water containing the reactant.

The respective concentrations were selected to obtain stable foams. All experiments were carried out at around 298 K and near atmospheric pressure. Figure 5 gives the pressure drop per unit

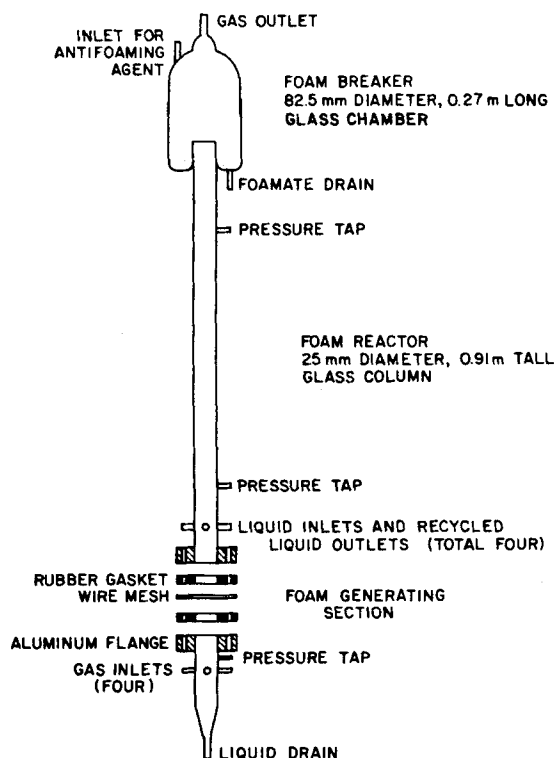


Figure 4. Schematic diagram of foam reactor.

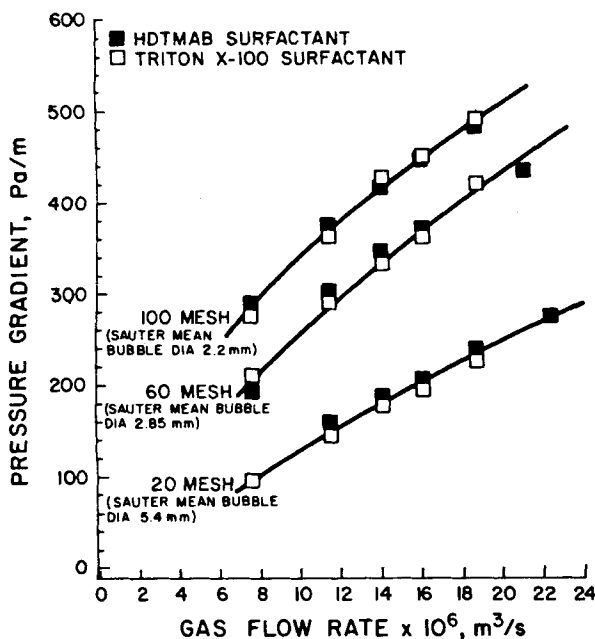


Figure 5. Pressure gradient for flowing foam, under various operating conditions.

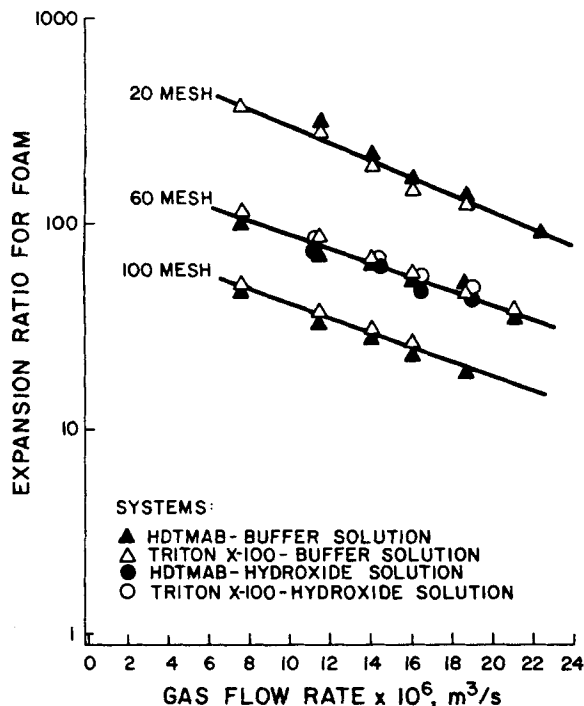


Figure 6. Expansion ratio for foam, under various operating conditions.

length for flowing foam; Figure 6 gives the expansion ratio of foam under various operating conditions.

FOAM BUBBLE SIZE MEASUREMENTS

A photographic technique (Damle, 1981) was used for measuring the foam bubble sizes. Consistency of foam in terms of bubble sizes, absence of channelling, etc., was observed to be good in all cases studied. The average bubble size was found to depend strongly on the wire mesh size used. Not much variation in bubble size was observed with either the foam flow rate or with the surfactant type. It was observed that the average bubble size distribution tended to become polydisperse with a coarser wire mesh.

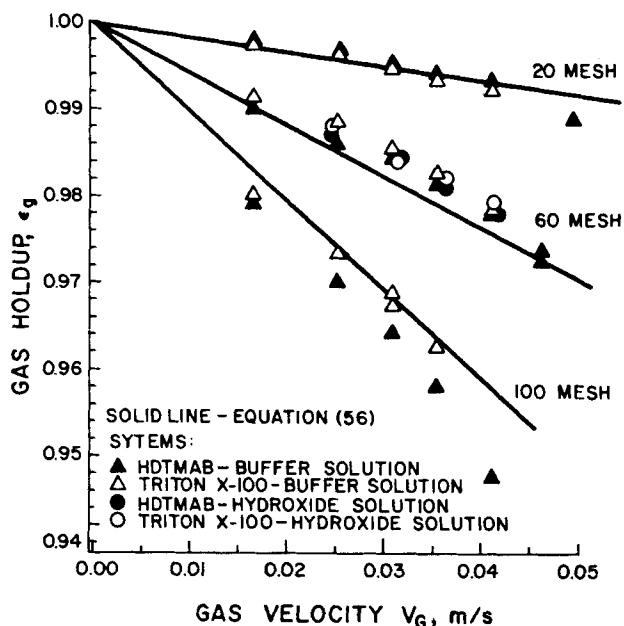


Figure 7. Gas holdup under various operating conditions.

RESULTS AND DISCUSSION

Gas Holdup

Figure 7 shows the gas holdup as a function of gas velocity for the wire mesh sizes used. The gas holdup obtained experimentally is correlated as a function of gas velocity and the bubble size using statistical analyses. The relation obtained is

$$\epsilon_g = 1 - 4.376 \times 10^{-6} V_G^{1.0125} d_s^{-2.02} \quad (56)$$

where V_G in m/s and d_s in m.

This agrees with the empirical correlation proposed by Hoffer and Rubin (1969) under identical condition, i.e., for stable foams moving vertically upward in plug flow regime.

The solid line in Figure 7 represents the gas holdup using Eq. 56.

Gas-Liquid Interfacial Area

The gas holdup decreases only slightly with an increase in gas velocity. The bubble size also decreases only slightly with an increase in gas velocity. Therefore, the variation in interfacial area with gas velocity is negligible. Hence an average value of interfacial area for each wire mesh size is used, Table 1. The interfacial area obtained in foam reactors appears to be one magnitude higher than that in conventional gas-liquid contactors, e.g., packed column, plate column, bubble column, etc. (Table 2). Higher interfacial areas could be obtained by using finer wire mesh sizes.

Modified Interfacial Mass Transfer Coefficient

The term, modified interfacial mass transfer coefficient used in this study, is the product of interfacial mass transfer coefficient and Henry's constant for solute in the interfacial region. Inserting experimental data for absorption of carbon dioxide in sodium

TABLE 1. GAS-LIQUID INTERFACIAL AREA

Wire Mesh Size	Pore Size mm	Sauter Mean Bubble Dia., mm	Interfacial Area, m ² /m ³ of Reactor
20	0.841	5.4	1,105
60	0.250	2.85	2,070
100	0.149	2.2	2,645

TABLE 2. COMPARISON OF GAS-LIQUID INTERFACIAL AREA

	Area m^2/m^3 of Reactor
Foam Reactor	
Wire Mesh Size 20	1,105
Wire Mesh Size 60	2,070
Wire Mesh Size 100	2,645
Packed Column	
Countercurrent	10-350
Cocurrent	10-1,700
Plate Columns	
Bubble Cap	100-400
Sieve Plates	100-200
Bubble Columns	50-600
Packed Bubble Columns	50-300
Spray Columns	10-100

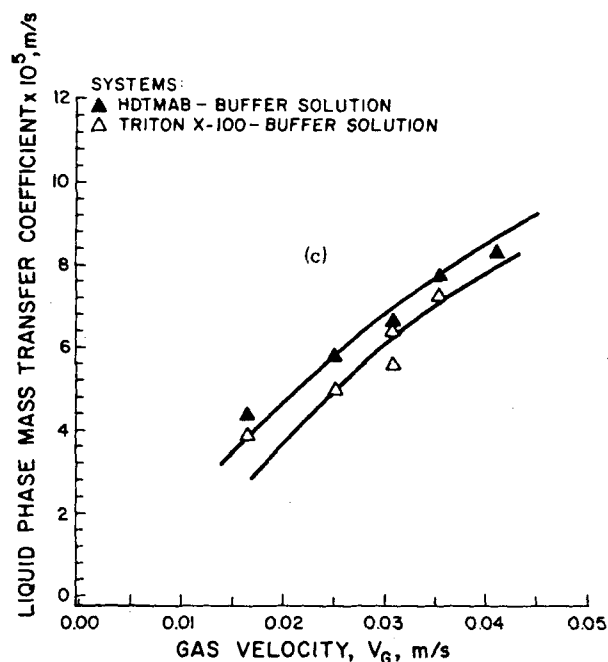
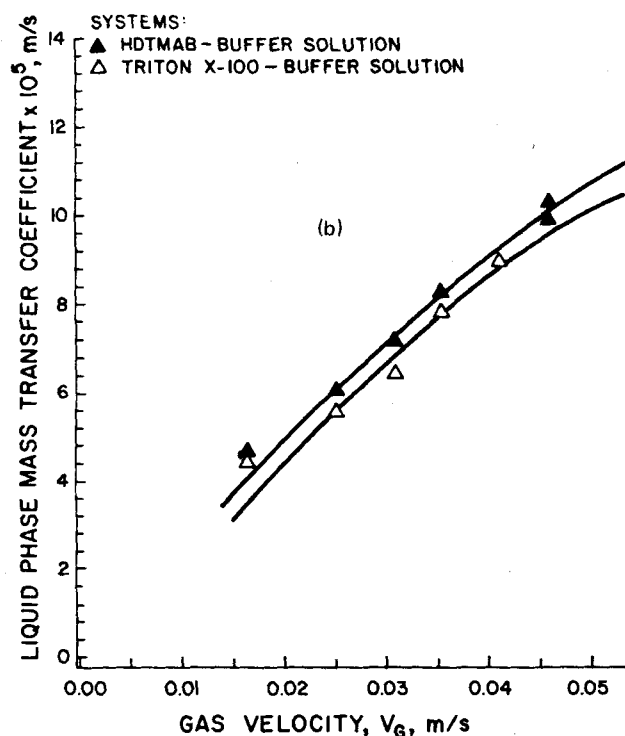
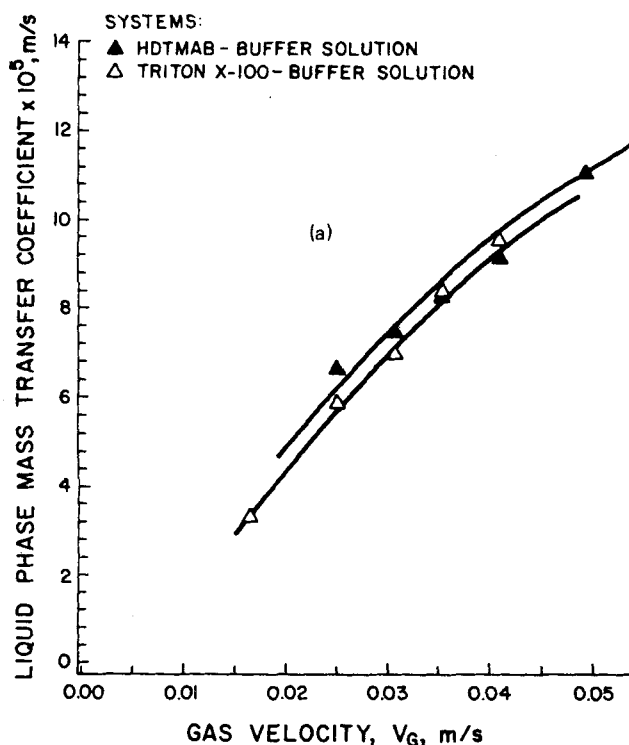


Figure 8. Liquid-phase mass transfer coefficient as a function of gas velocity (a) wire-mesh size 20; (b) wire-mesh size 60; (c) wire-mesh size 100.

hydroxide solution into Eqs. 24 through 29, $k_l H_s$ can be obtained. Since $k_l H_s$ is independent of gas flow rate and wire mesh size, the value of $k_l H_s$ for a particular surfactant is determined by taking a mean value for all experimental runs for that surfactant. The $k_l H_s$ for nonionic Triton X-100 surfactant is $1.27 \times 10^{-11} \text{ kmol/m}^2 \cdot \text{Pa} \cdot \text{s}$ and for cationic hexadecyl trimethyl ammonium bromide (HDTMAB) surfactant is $1.44 \times 10^{-11} \text{ kmol/m}^2 \cdot \text{Pa} \cdot \text{s}$, at 298 K. These results indicate that Triton X-100 offers about 10% higher resistance to mass transfer than that by HDTMAB. This is due to the higher viscosity of Triton X-100 at the interface compared to that of HDTMAB.

The lowest value of gas-side mass transfer coefficient obtained in a bubble column, a cocurrent packed column, or a plate column is about $1 \times 10^{-9} \text{ kmol/m}^2 \cdot \text{Pa} \cdot \text{s}$. Comparing this with the value of $k_l H_s$ obtained, it can be seen that the gas-side resistance is negligible. Therefore, the assumption of negligible gas-phase resistance is valid.

Because of the difficulty associated with independent measurements on k_l and H_s , the validity of $k_l H_s$ can be judged only in terms of order of magnitude evaluations. Data reported in the literature are not in the form of interfacial mass transfer coefficient k_l , but in the form of diffusivity of solute in the interfacial region D_1 . The interfacial mass transfer coefficient is related to diffusivity and the thickness of the interfacial region, according to the film model, as

$$k_l = D_1 / \delta \quad (57)$$

The interfacial thickness may be somewhat larger than the thickness of the surfactant monolayer. This is so because of the rather high degree of orientation of water and electrolyte molecules in the vicinity of charged ends of the surfactant molecules (Davies and Rideal, 1961). One likewise expects that diffusivity of solute in the interfacial region would reflect both the nature of the surfactant and of the oriented molecules in the bulk liquid. The interfacial region has a finite thickness in which the properties differ quite radically from those of the bulk liquid. Thus, the large resistance of the interfacial region may extend beyond the surfactant film, so that D_1 and H_s are essentially average values within the region of high molecular orientation (Nguyen Ly et al., 1979).

Nguyen Ly et al. (1979) concluded that the interfacial region has a higher solubility for the dissolved gases than the solubility in water and that the interfacial region has a thickness less than 2,000

Å. They reported a value of 6.9×10^{-6} kmol/m³·Pa at 298 K for Henry's constant for carbon dioxide in Petrowet-R surfactant. To make an order of magnitude comparison, the H_s value reported by Nguyen Ly et al. (1979) is used and an interfacial thickness of 1,500 Å is assumed. The diffusivities D_1 then obtained are 3.1×10^{-13} m²/s and 2.75×10^{-13} m²/s for HDTMAB and Triton X-100 surfactants, respectively. The diffusivity for solute in the interfacial region is, thus, four orders of magnitude lower than that in the bulk liquid. A similar conclusion was arrived at by Nguyen Ly et al. (1979). The value of modified interfacial mass transfer coefficient obtained in this study is, therefore, correct based on order of magnitude comparison.

Liquid-Phase Mass Transfer Coefficient

For determining the liquid-phase mass transfer coefficient, a modified interfacial mass transfer coefficient is needed. For the absorption of carbon dioxide in buffer solution, the value of $k_I H_s$ used is that obtained for the carbon dioxide in sodium hydroxide solution for the same surfactant. The modified interfacial mass transfer coefficient for sodium buffer solution may be slightly different from that of sodium hydroxide. In both cases, however, electrolyte nature and concentration are almost the same and hence the modified interfacial mass transfer coefficient for sodium buffer will not be much different from that for sodium hydroxide. In addition, in the absence of any knowledge about the effect of above electrolytes on $k_I H_s$ for the surfactants employed, the assumption of same value of $k_I H_s$ for sodium hydroxide and sodium carbonate-carbonate solution seems to be reasonable.

Figures 8a, b and c show the liquid-phase mass transfer coefficient as a function of gas velocity, for the wire mesh sizes employed. The liquid-phase mass transfer coefficient increases with gas velocity as V_G (0.75 ± 0.05) in all cases. It is observed that for a particular surfactant, k_L increases with increase in Sauter mean bubble diameter. Increase in k_L with bubble size, however, is less than 20%. A similar conclusion about the increase in k_L with bubble size was arrived at by Deckwer et al (1980) in bubble columns.

Mass Transfer Rate

For the case of absorption of carbon dioxide in an aqueous solution of sodium hydroxide in the presence of surfactant, the mass transfer rate is reduced significantly. Substituting relevant data in Eq. 19, it can be seen that the liquid-phase resistance is just 1% of the total resistance (Appendix I). Hence for this fast chemical reaction case, the interfacial resistance is the controlling factor.

For the case of slow reaction between carbon dioxide and sodium carbonate-bicarbonate in presence of surfactant, the interfacial resistance is 40–70% of the total resistance (Appendix II). The lower value is for the lowest gas flow rate and at the inlet, and the higher one for the highest gas flow rate at the outlet. This means that the interfacial resistance reduces the mass transfer rate per unit volume of stable foam by 70–200% of that which could be obtained in the absence of interfacial resistance. The magnitude of hydrodynamic effect due to surfactant, however, is not accounted for in this reduced mass transfer rate as there is no study reported in the literature under similar hydrodynamic conditions in the absence of surfactant. For this slow reaction case, however, the mass transfer rate is about 2 to 4 times higher than that of conventional absorbers. Also due to the large contact time available in foam reactors, slow reactions can conveniently be carried out in such reactors.

APPENDIX I: FAST REACTION CASE (CO₂/NaOH); RUN 4.4–6

$$P = 9.23 \times 10^4 \text{ Pa}; T = 298 \text{ K}$$

$$Li = 8.33 \times 10^{-7} \text{ m}^3/\text{s}; Lo = 2.5 \times 10^{-7} \text{ m}^3/\text{s}$$

$$\left. \begin{array}{l} \text{Volumetric flow rate of gas entering} \\ \text{reactor at 1.0133 kPa and 298 K} \end{array} \right\} = 1.423 \times 10^{-5} \text{ m}^3/\text{s}$$

$$\text{Mol fraction of CO}_2 \text{ entering the reactor (gas phase)} = 0.0965$$

$$\text{Mol fraction of CO}_2 \text{ leaving the reactor (gas phase)} = 0.00525$$

$$C_{Bi} = 1.0701 \text{ kmol/m}^3; C_{Bo} = 0.8134 \text{ kmol/m}^3;$$

$$C_{Ci} = 0.0035 \text{ kmol/m}^3; C_{B1} = 0.9935 \text{ kmol/m}^3;$$

$$y_1 = m_1/(m_1 + 1) = 0.04727$$

$$a = 2,070 \text{ m}^2/\text{m}^3 \text{ (HDTMAB surfactant);}$$

$$k_I H_s = 1.45 \times 10^{-11} \text{ kmol}/(\text{m}^2 \cdot \text{Pa} \cdot \text{s})$$

$$C_B = (C_{B1} + C_{Bo})/2 = 0.9035 \text{ kmol/m}^3;$$

$$C_{C1} = \{(C_{B1} - C_{Bo})/2\} + C_{Ci} = 0.0856;$$

$$C_{Co} = 0.1757 \text{ kmol/m}^3 \quad C_c = (C_{C1} + C_{Co})/2 = 0.1306 \text{ kmol/m}^3$$

$$I_1 = I_{\text{NaOH}} = C_B = 0.9035 \text{ kmol/m}^3;$$

$$I_2 = I_{\text{Na}_2\text{CO}_3} = 3C_c = 0.3918 \text{ kmol/m}^3;$$

$$H_w = 3.44 \times 10^{-7} \text{ kmol}/(\text{m}^3 \cdot \text{Pa});$$

$$H = 2.38 \times 10^{-7} \text{ kmol}/(\text{m}^3 \cdot \text{Pa})$$

$$D_{Aw} = 1.99 \times 10^{-9} \text{ m}^2/\text{s}; D_A = 1.69 \times 10^{-9} \text{ m}^2/\text{s};$$

$$I = 1.2953 \text{ kmol/m}^3; k_2 = 1.23 \times 10^4 \text{ m}^3/(\text{kmol} \cdot \text{s})$$

$$H\sqrt{D_A k_2 C_B} = 1.03 \times 10^{-9} \text{ kmol}/(\text{m}^2 \cdot \text{Pa} \cdot \text{s})$$

$$(1/K_I H_s)/\{(1/K_I H_s) + (1/H\sqrt{D_A k_2 C_B})\} = 98.5\%$$

Foam reactors: mass transfer rate per unit volume of reactor

$$-(G_i/A) dm/dZ = (aP_A)/\{(1/k_I H_s) + (1/H\sqrt{D_A k_2 C_B})\}$$

$$\approx 3.08 \times 10^{-8} P_A \text{ kmol}/(\text{m}^3 \cdot \text{s})$$

Conventional packed bed reactors:

$$a = 100 \text{ to } 400 \text{ m}^2/\text{m}^3, \text{ say } 250 \text{ m}^2/\text{m}^3$$

$$-(G_i/A) dm/dZ = aP_A H\sqrt{D_A k_2 C_B}$$

$$= 2.5 \times 10^{-7} P_A \text{ kmol}/(\text{m}^3 \cdot \text{s})$$

i.e., mass transfer rates in foam reactors about 8 times lower.

APPENDIX II: SLOW REACTION CASE (CO₂/BUFFER); RUN 4.3–13

$$P = 9.283 \times 10^4 \text{ Pa}; T = 298.5 \text{ K}$$

$$Li = 8.33 \times 10^{-7} \text{ m}^3/\text{s}; Lo = 3.5 \times 10^{-7} \text{ m}^3/\text{s}$$

$$\left. \begin{array}{l} \text{Volumetric flow rate of gas entering} \\ \text{reactor at 1.0133 kPa and 298 K} \end{array} \right\} = 1.142 \times 10^{-5} \text{ m}^3/\text{s}$$

$$\text{Mol fraction of CO}_2 \text{ entering the reactor (gas phase)} = 0.1081$$

$$\text{Mol fraction of CO}_2 \text{ leaving the reactor (gas phase)} = 0.01835$$

$$C_{Ci} = 0.5348 \text{ kmol/m}^3; C_{Co} = 0.4271 \text{ kmol/m}^3;$$

$$C_{Di} = 0.1936 \text{ kmol/m}^3;$$

$$C_{Do} = 0.4089 \text{ kmol/m}^3; C_{C1} = 0.5229 \text{ kmol/m}^3;$$

$$C_{D1} = 0.2175 \text{ kmol/m}^3; y_1 = 0.08905$$

$$a = 2,650 \text{ m}^2/\text{m}^3 \text{ (Triton X-100 surfactant);}$$

$$k_I H_s = 1.27 \times 10^{-11} \text{ kmol}/(\text{m}^2 \cdot \text{Pa} \cdot \text{s});$$

$$y_o = 0.018409 \text{ (calcd.)};$$

$$k_L = 4.93 \times 10^{-5} \text{ m/s}$$

$$K_2/K_1 = 1.05 \times 10^{-4}; k_2 K_w/K_2 = 0.88;$$

$$D_A = 1.66 \times 10^{-9} \text{ m}^2/\text{s}; H = 2.35 \times 10^{-7}$$

$$\text{kmol}/(\text{m}^3\cdot\text{Pa}); B_1H = 1.8 \times 10^{-11} \text{ kmol}/(\text{m}^2\cdot\text{Pa}\cdot\text{s});$$

$$K_1H_s = 1.27 \times 10^{-11} \text{ kmol}/(\text{m}^2\cdot\text{Pa}\cdot\text{s})$$

Foam reactors: mass transfer rate per unit volume of reactor

$$-(G_i/A) dm/dZ = a\{Py - (C_{Ab}/H)\} / \{(1/k_1H_s) + (1/B_1H)\}$$

$$= 7.5 \times 10^{-12} a\{Py - (C_{Ab}/H)\} \text{ kmol}/\text{m}^3\cdot\text{s}$$

$$a = 1,500 - 3,000$$

therefore,

$$-(G_i/A) dm/dZ = 11.25 \times 10^{-9} \text{ (to } 22.5 \times 10^{-9})$$

$$\times \{Py - C_{Ab}/H\} \text{ kmol}/\text{m}^3\cdot\text{s}$$

Conventional packed bed reactors

$$k_L \approx 1 \times 10^{-4} \text{ m/s}$$

$$a \approx 200 \text{ m}^2/\text{m}^3$$

$$-(G_i/A) dm/dZ = aH\sqrt{D_a(k_2K_w/k_2)(C_c/C_D) + k_L^2}$$

$$\{Py - (C_{Ab}/H)\} = 5.5 \times 10^{-9} \{Py - (C_{Ab}/H)\} \text{ kmol}/\text{m}^3\cdot\text{s},$$

i.e., mass transfer rate in foam reactors 2-4 times higher.

NOTATION

A	= cross-sectional area of reactor, m^2
a	= gas-liquid interfacial area, m^2/m^3
a_1, a_2, a_3	= constants in diffusivity relation for sodium hydroxide, sodium carbonate, and sodium bicarbonate, respectively, m^3/kmol
b	= stoichiometric factor for reaction between carbon dioxide and sodium hydroxide
C_A^*	= concentration of free carbon dioxide at liquid interface, kmol/m^3
C_{Ab}	= concentration of free carbon dioxide in bulk liquid, kmol/m^3
C_{Ai}	= concentration of carbon dioxide at surfactant-liquid interface in equilibrium with C_A^* , kmol/m^3
C_{Ai}^*	= concentration of carbon dioxide in equilibrium with the solute having partial pressure P_{Ai} at gas-surfactant interface, kmol/m^3
C_C	= concentration of carbonate, kmol/m^3
C_D	= concentration of bicarbonate, kmol/m^3
C_{Bi}, C_{Bo}, C_{B1}	= concentration of hydroxide in liquid stream entering the reactor, leaving the reactor and at the beginning of stable foam stage, respectively, kmol/m^3
C_{Ci}, C_{Co}, C_{C1}	= concentration of carbonate in liquid stream entering the reactor, leaving the reactor and at the beginning of stable foam stage, respectively, kmol/m^3
C_{Di}, C_{Do}, C_{D1}	= concentration of bicarbonate in liquid stream entering the reactor, leaving the reactor and at the beginning of stable foam stage, respectively, kmol/m^3
D_1	= diffusivity of solute in the interfacial region, m^2/s
D_A	= diffusivity of carbon dioxide in liquid, m^2/s
D_{Aw}	= diffusivity of carbon dioxide in water, m^2/s
d_{bj}	= individual bubble diameter, m
d_{ds}	= Sauter mean bubble diameter, m
G_i	= molar flow rate of inerts, kmol/s
H	= Henry's constant for carbon dioxide in direct contact with liquid, $\text{kmol}/(\text{m}^3\cdot\text{Pa})$
H_s	= Henry's constant for carbon dioxide in the interfacial region, $\text{kmol}/\text{m}^3\cdot\text{Pa}$

H_T	= total length of the reactor, m
H_w	= Henry's constant for carbon dioxide in water, $\text{kmol}/\text{cm}^3\cdot\text{Pa}$
h	= salting out parameter for electrolyte having ionic strength I , m^3/kg ion
I	= ionic strength of electrolyte, kmol/m^3
K	= distribution coefficient between interfacial and bulk liquid phase
K_1	= equilibrium constant for reaction $\text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{H}^+ + \text{HCO}_3^-$, kmol/m^3
K_2	= second ionization constant for $\text{HCO}_3^- \rightleftharpoons \text{H}^+ + \text{CO}_3^{2-}$, kmol/m^3
K_w	= ion product for water, $(\text{kmol}/\text{m}^3)^2$
k	= k_2K_w/K_2 , s^{-1}
k_2	= second-order reaction rate constant for reaction between carbon dioxide and hydroxide, $\text{m}^3/\text{kmol}\cdot\text{s}$
k_I	= interfacial mass transfer coefficient, m/s
k_{Ia}	= volumetric interfacial mass transfer coefficient, s^{-1}
k_{ga}	= volumetric gas-phase mass transfer coefficient, $\text{kmol}/\text{m}^3\cdot\text{Pa}\cdot\text{s}$
k_L	= liquid-phase mass transfer coefficient, m/s
k_1H_s	= modified interfacial mass transfer coefficient, $\text{kmol}/\text{m}^2\cdot\text{Pa}\cdot\text{s}$
L_i, L_o	= flow rate of liquid entering the reactor, and leaving the reactor, respectively, m^3/s
m	= moles of carbon dioxide per mole of inerts
m_i, m_o, m_1	= moles of carbon dioxide per unit mole of inerts, at inlet, at outlet, and at the beginning of stable foam stage, respectively
N_j	= number of bubbles having diameter d_{bj}
P	= total pressure, Pa
P_A	= partial pressure of carbon dioxide in bulk gas phase, Pa
P_{Ai}	= partial pressure of carbon dioxide at gas-surfactant interface, Pa
R	= rate of absorption per unit interfacial area, $\text{kmol}/\text{m}^2\cdot\text{s}$
T	= temperature, K
Z	= distance from the wire mesh gas distributor, m
Z_i	= height of liquid pool, m
δ	= thickness of the interfacial region, \AA
ϵ_g	= gas holdup
ϕ	= expansion ratio

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Analysis of Staged Liquid Surfactant Membrane Operations

Graphical and numerical procedures are presented for the design and analysis of cocurrent and countercurrent multistaged mixer-settler units for liquid membrane extraction operations—these are based on the recently developed Advancing Front Model of Ho et al. (1982) and Hatton et al. (1983). The numerical results indicate that the extraction efficiency in a countercurrent cascade is only slightly improved over that for cocurrent operation. However, for any given stage, marked improvements in extraction efficiency are indicated if an external emulsion recycle over the mixer settler unit is used to ensure adequate holdup ratios in the mixer. Premixing of the emulsion feed and recycle streams will impair the extraction performance, however,

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SCOPE

Despite a continuing strong interest in liquid surfactant membrane operations, very little attention has been devoted to discussion of scale-up and design procedures for these processes. In the one paper addressing this problem directly, it was suggested that the model of Cahn and Li (1974) be used for the sizing of continuous, multistage processing equipment. However, their implicit assumption of an unchanging resistance to mass transfer during extraction could not be substantiated by their experimental findings, nor by those of subsequent workers (Hochhauser and Cussler, 1975; Kondo et al., 1979; Hatton et al., 1983). In an attempt to improve the predictive value of these process models, Ho et al. (1982) and Hatton et al. (1983) developed the so-called Advancing Front Model for batch and con-

tinuous operations, respectively, in which cognizance was taken of the increase in mass transfer resistance with increasing contact time. They found that the model provided a very good description of the mass transfer processes occurring during their solute extraction experiments. Their results bode well for the effective implementation of this model in the design and analysis of multistage, liquid membrane extraction operations, and the purpose of this paper is to extend the earlier developments for single-stage, continuous extraction processes (Hatton et al., 1983) to allow also for the analysis and design of cocurrent and countercurrent multistage operations in terms of the Advancing Front Model.

CONCLUSIONS AND SIGNIFICANCE

Liquid surfactant membrane extraction operations are well described by the Advancing Front Model; the model assumes that an instantaneous and irreversible reaction between the diffusing solute and the immobile internal reagent occurs at a reaction front which moves into the emulsion globule as the

reaction proceeds. Based on this model, a simple graphical procedure has been developed for the design and analysis of cocurrent and countercurrent multistaged liquid membrane operations. The results indicate an increase in extraction efficiency when the countercurrent rather than the cocurrent arrangement is used, although the differences in overall fractional utilization of internal reagent for the two cases are slight, being

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