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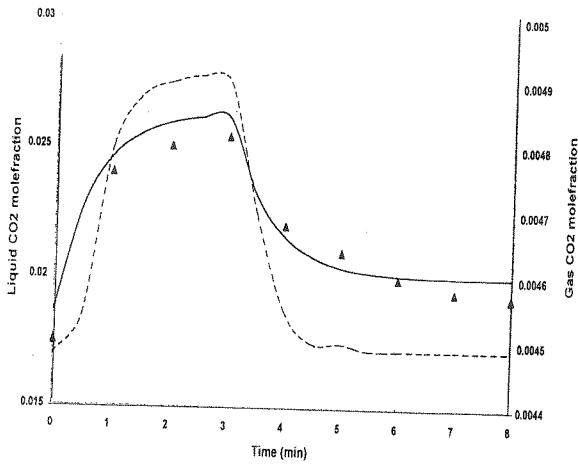
Figure (3) Outlet composition response to a positive pulse forcing of feed gas concentrtion. (▲) Experimental, (__) predicted for liquid and (---) gas phase.
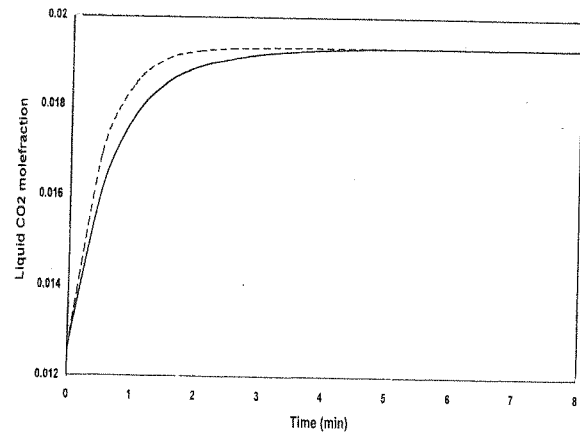

Figure (5) Outlet composition response to a positive step forcing of gas feed flow (▲) Experimental, (__) predicted for liquid and (---) gas phase
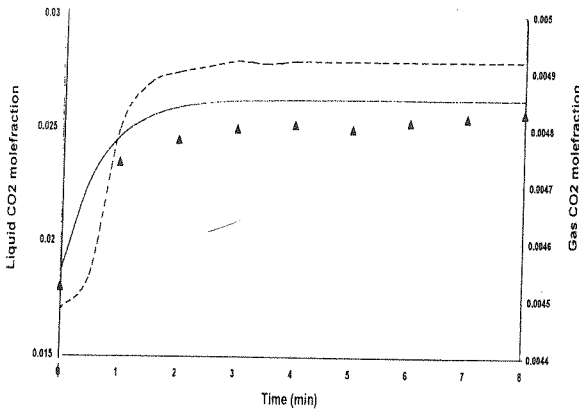

Figure (4) Outlet composition response to a positive step forcing of feed concentration. (▲) Experimental, (__) predicted for liquid and (---) gas phase.
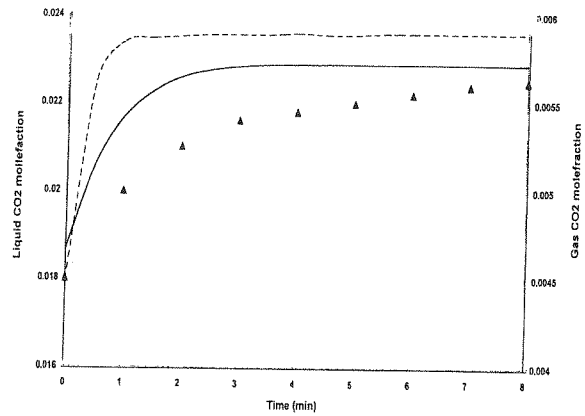

Figure (6) Outlet composition response to a positive step forcing of liquid feed flow rate. (▲) Experimental , (__) predicted for liquid and (---) gas phase
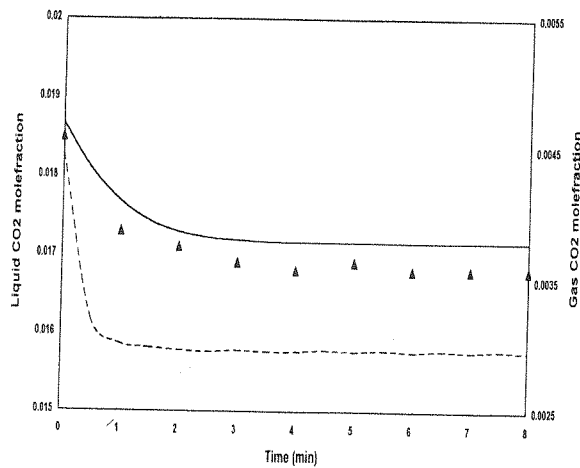

Figure (7) Outlet composition response to positive step forcing of feed gas concentration .(__) with gas holdup , (---) without holdup.

H = enthalpy, kJ/kmole

Ha = Hatta number

He = Henry law constant, $kmole/m^3.kpa$

ΔH_{si} = heat of solution of component i, kJ/ kmole in solution

ΔH_{ri} = heat of ith reation kJ/kmole of A_i that reacts

$k_{G,Ai}$ = gas mass transfer coefficient for absorbed component i, $kmole/m^2.s.kPa$

$k_{l,Ai}$ = liquid mass transfer coefficient for absorbed component i, m^2/s

L = liquid flow rate, $kmole/m^2.s$

L' = solute free liquid flow rate, $kmole/m^2.s$

M = molecular weight, kg/kmole

$n_{f,G}$ = number of free component in gas phase

$n_{f,L}$ = number of free component in liquid phase

N_{Ai} = mass flux of absorbed component i, $kmole/m^2.s$

r = reaction rate $kmole/m^3.s$

R = ideal gas constant, kJ/kmole.K

t = time, s

T = temperature, K

x_i = mole fraction of Component i in the liquid phase

y_i = mole fraction of component i in the gas phase

y = distance from gas-liquid interface, m

z = axial distance in the column, m

Greek symbols

δ = liquid film thickness, m

φ = fugacity coefficient

ϕ = total volumetric holdup

λ = latent heat of vaporization, kJ/ kmole

ρ = density, kg/m^3

Subscripts

A_i = respect to absorbed component i

b = in the bulk of the gas or liquid phase

G = gas

L = liquid

p_j = respect to product j

R_j = respect to reactant j

ref = reference

Superscripts
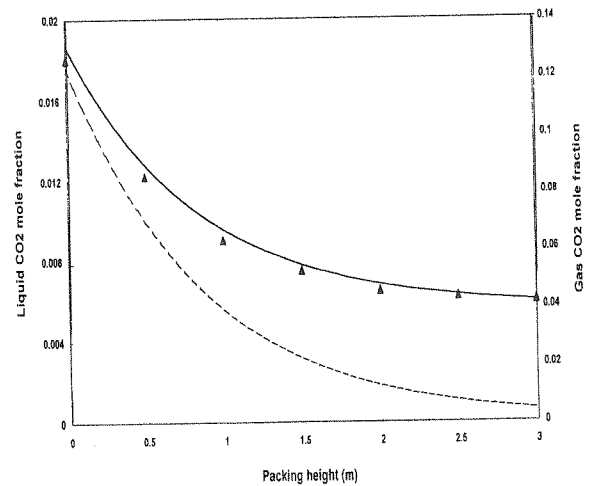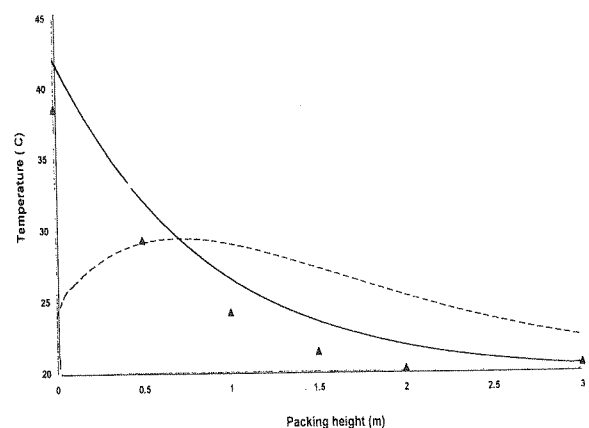
G = gas

L = liquid

Figure (1) Liquid and gas phase of co_2 concentration profile at steady - state. (▲) Experimental, (__) predicted for liquid and (---) gas phase.

Figure(2) liquid and gas temperature profile at steady- state (▲) Experimental, (__) predicted for liquid and (---) gas phase

Results And Discussion

Typical plots of experimental results and predicted ones for liquid and gas concentrations and temperatures along the column under steady-state are shown in figures 1, 2 corresponding to operating conditions of run S2. As can be seen from figure 1 the effective irrigated interfacial area correlation and the channelling of liquid that occures along the column and the assumption of plug flow for both phases reduce the accuracy of results.

Figure 2 shows the predicted temperature with experimental data for both phases. It can be seen that the predicted temperatures are higher that the measured values. This, can be attributed to the fact that the column was assumed adiabatic and energy losses through the wall was neglected . The gas temperature at the entrance of the column raises rapidly because of contact with liquid that is at a higher temperature due to the heat of solution and chemical reaction . After reaching its maximum temperature, the gas starts cooling down by transfering its energy to the liquid that is trickling down the column .

The transient response of the gas and liquid phases at outlet compositions based on various step changes are illustrated in figures 3-6. As shown in these figures, there is no linear relation between step forcing and responses. Among the factors affecting the transient outlet concentration of both outlet phases are solute solubilities, rate of mass transfer , flow rate of the liquid and gas phases and holdups within the column. De Maria and White (1960) showed that the axial dispersion for both phases makes only a small contribution to the dynamics of the system.

In figure 7 predictions of model based on change step forcing of CO_2 feed concentration at 1200 Kpa pressure with and without gas holdup are compared. Clearly gas holdup plays a singificant role in the dynamics of relativly high- pressure systems. The total molar gas phase holdup is directly proportional to the column operating pressure and this term could be omitted at low pressure without loss of accuracy.

Conclusions

This study presents a method for steady and unsteady-state simulation of nonisothermal absorption with chemical reaction in packed columns. Assumptions such as considering adiabatic condition for column , or negligible axial dispersion in both phases might decrease accuracy of results. Otherwise the gas-liquid equilibrium relationship and mass transfer coefficient could affect the accuracy of results. Likely the effect of gas phase holdup on system dynamics cannot be ignored at high pressures. In addition there is a good agreement between steady-state and transient experimental data and model predictions for CO_2 -MEA system.

Nomenclature

a   = interfacial are per unit volume of packing, m^2/m^3

C_i = concentration of component i in the liquid, $kmole/m^3$

$C_{p,L}$ = average heat capacity of liquid mixture, $kJ/kmole.°C$

$C_{p,G}$ = average heat capacity of gas mixture, $kJ/kmole.°C$

D_i = molecular diffusivity of component i in solution, m^2/s

E   = enhancement factor

G   =total gas flow rate, $kmole/m^2.s$

G'   = solute free gas flow rate, $kmole/m^2.s$

h_G = total molar gas holdup, $kmole/m^3$

h_L = molar liquid holdup, $kmole/m^3$

substituted back into the model equations 26-28, and the process is repeated until the difference between consecutive solutions has met the convergence criterion.

Similarly to achieve each time step; constant values are determined based on previous time step and calculations continue to an approximation to the actual individual component concentration and bulk temperature profiles over the entire column at that point in time.

Experimental equipment and procedure

The absorption process were performed in a 10 cm i. d. stainless steel column3. 2 m long packed with 12. 7mm (1/2") ceramic Rashig rings. The column consisted of three sections with about 1 m of packing height and six liquid sampling ports and thermocouple probes. Between each section redistributors were inserted to prevent the liquid channelling down the wall.

Dry CO_2 from cylinder and dry pressurized air from the compressor, after mixing were fed to the bottom and MEA solution from 150 l tank pumped to the top of the colmn. Air, CO_2 and MEA solution flow rates were measured by rotameters.

Steady-state data obtained from six different runs (S1-S6) were used to adjust the mathematical moldel. The range of operating conditions for these runs is shown in table 1. The operating pressure for all runs was 150 KPa and amine concentrations of 15 weight percent were used throughout.

Several runs were done to provide transient data to use in model for comparison purposes. Initial coditions of all dynamic runs is similar to run S2 that is : total gas flow rate. 48 Kmol/ hr; liquid flow rate .09 m^3 / hr; feed CO_2 concentration 12. 3%; inlet gas temperature

24.5°C; inlet liquid temperature 20.5° C. Step changes were consisted of : 40% change as a positive rectangular pulse forcing of the feed CO_2 concentration (run T1); 40% change as a positive step forcing of the feed CO_2 concentration (run T2); 25% change as a positive step forcing of the feed solution flow rate (run T3); and 15% change as a positive step forcing of the total gas feed flow rate (run T4).

For each dynamic run before imposing step change, steady-state conditions must be established.

These conditions were usually reached in about half an hour and could be determined from the constancy of temperature and pH outlet solution

Liquid samples were analyzed to determine the total amine concentration and CO_2 content.

The total amine concentration was found by titration with standard 1 N HCL to the methyl orange end point and the CO_2 content in the liquid sample was determined by acidification with 2 N HCL and volumetric measurment of the CO_2 evolved .

Due to results that presented by

Tontiwachwuthikul et al. (1992), negligible massbalance error, similarity of equipments were used and accuracy of liquid analysing method, outlet gas analysis was omitted

Table (1) Range of Operating Variable in the Packed Absorption Column

Variable	Range
Gas flow rate (kmol/hr)	.46-.57
Liquid flow rate (m³/hr)	.085-.115
Gas CO_2 concentration (%)	12-17.5
Inlet gas temperature (° C)	23.5-26
Inlet liquid temperature (° C)	18-24.5

$$E_{Ai\infty} = 1 + \frac{D_{Ri}C_{Ri}}{2D_{Ai}C_{Ai,L}} \qquad (23)$$

Hikita and Asai (1990) correlated the reaction rate constant (k_1) by the following equation:

$$\log k_1 = 10.99 - \frac{2152}{T} \qquad (24)$$

Numerical Solution

The finite difference technique is a suitable numerical solution for the unsteady-state simulation. This method was employed to convert the differential equations into a coupled system of algebraic equations.

To simplify the differential equations, equation 2 can be written as:

$$h_G \frac{\partial y_{Ai}}{\partial t} + G \frac{\partial y_{Ai}}{\partial z} + y_{Ai}(\frac{\partial h_G}{\partial t} + \frac{\partial G}{\partial z}) = -N_{Ai}\Big|_{y=0} a \qquad (25)$$

The term in the bracket is in fact equation 11. Therfore substitution yields:

$$h_G \frac{\partial y_{Ai}}{\partial t} + G \frac{\partial y_{Ai}}{\partial z} + y_{Ai}(-\sum_{i=1}^{nA} N_{Ai}\Big|_{y=0}\alpha) = -N_{Ai}\Big|_{y=0} a \qquad (26)$$

Similarly equation 3 reduces to:

$$h_L \frac{\partial x_{Ai}}{\partial t} - L \frac{\partial x_{Ai}}{\partial z} = R_{Ai} \qquad (27)$$

To linearize the equations, the dependent variables functionality must be eliminated . This was accomplished by treating the values of dependent variables such as physical and transport properties as constants based on pervious values for the independent variables, which was proposed by Hitch and Rousseau (1986). Likewise, values of L, G, h_L, h_G, N_{Ai} are treated as constants determined from the solution of equations at the previous time step.

Employing the aforementioned method and combining equations 6 and 12, equation 6 can be written as:

$$h_L \frac{\partial x_{R_{j,i}}}{\partial t} - L \frac{\partial x_{R_{j,i}}}{\partial z} = \frac{a_{R_{j,i}}}{a_{A_i}}(N_{Ai}\Big|_{y=0}\alpha - R_{Ai}) \qquad (28)$$

Likely equations 5 and 8 after combination with eqations 6 and 7 become;

$$C_{Lm}h_L \frac{\partial T_L}{\partial t} - C_{Lm}L \frac{\partial T_L}{\partial z} + C_{Gm}G \frac{\partial T_G}{\partial z} =$$

$$\sum_{i=1}^{n_A} (N_{Ai}\Big|_{y=0}\alpha(-\Delta H_{sAi}) + (N_{Ai}\Big|_{y=0}\alpha - R_{Ai})(-\Delta H_{ri})) \qquad (29)$$

$$C_{Gm}G \frac{\partial T_G}{\partial z} = Ua(T_L - T_G) \qquad (30)$$

Approximating the solution of equations 26- 30 is generally referred to as an implicit integration method. By using this method, the differential equatios will reduce to linear algebraic equations. Each of quations 26-28 may be solved independently to yield the new values of the mole fractions of each component in both liquid and gas phases throughout the finite-difference grid at arbitrary time steps. The new values are then used to determine the molar liquid and gas flow rates L and G from equations 16 and 17 and interfacial mass flux N_{Ai} from equation 13. This new values are then used in conjunction with equations 29 and 30 to determine the liquid and gas temperatures throughout the column.

The numerical procedure used to solve above equations can be found in Hitch and Rousseau (1987). Consequently the values of physical and transport properties are corrected by the calculated variables and then these are

the time derivative terms to zero.

In this study, the physical properties of amine solution such as density, specific heat, viscosity, and thermal conductivity were obtained using predictive methods suggested by Cheng et al. (1996) and the Henry's constant for CO_2-MEA system proposed by Hikita et al. (1979). The diffusivity of CO_2 in amine solutions was estimated with the N_2O analogy by Versteand Swaaij (1988) at 298 K in water and corrected for viscosity and temperature using the modified Stoke-Einstein relationship. The total heat of absorption, which is the sum of the heat of solution and the heat of reaction, were taken from kohl and Riesenfeld (1979).

The mass transfer coefficients and specific interfacial area were estimated by using correlations developed by Onda et al. (1968a, b). Heat transfer coefficients were determined by using the Chilton Colburn analogy between heat and mass transfer.

The total volumetric Liquid holdup (ϕ_L) is the summation of static liquid holdup and dynamic liquid holdup which where correlated as a function of packing size and the flow rate and physical properties of the solvent are given by Treybal (1980). The total volumetric gas holdup (ϕ_G) may be assumed to be the difference between the dry packing void fraction and the total volumetric liquid holdup.

The total molar liquid and gas holdup are defined as:

$$h_L = \Phi_L \frac{\rho_L}{M_L} \qquad (14)$$

$$h_G = \Phi_G \frac{\rho\phi}{RT_G} \qquad (15)$$

The total molar liquid and gas flow rates based on solute free flow rate can be written as:

$$L = \frac{L'}{\sum_{i=1}^{nf,L} x_L} \qquad (16)$$

$$G = \frac{G'}{\sum_{i=1}^{nf,G} y_i} \qquad (17)$$

The absorption of CO_2 in MEA solution is accompanied by the following overall rate:

$$CO_2 + 2RNH_2 \Leftrightarrow RNHCOO^- + RNH_3^+ \qquad (18)$$

This reaction is stablished to be very fast, irreversible and second order with the kinetic equation

$$r = k_1 C_{CO2} C_{RNH2} \qquad (19)$$

Van Kervelen and Hoftijzer (1948) were the first who presented an approximation method for the description of the absorption with a second order reaction. Likewise porter (1966), Denckwerts (1970) and Decoursey (1974) derived an approximate solution for this process. Wellek et al. (1978) derived a new expression that could be used for the estimation of the enhancement factor as:

$$\frac{1}{(E_{Ai}-1)^{1.35}} = \frac{1}{(E_{A_{L,\infty}}-1)^{1.35}} + \frac{1}{(E_{A_{i,I}}-1)^{1.35}} \qquad (20)$$

where

$$E_{A_i,I} = \frac{(Ha)_{A_i}}{\tanh(Ha)_{A_i}} \qquad (21)$$

$$(Ha)_{Ai} = \frac{1}{k_{L,Ai}} \sqrt{D_{Ai} K_1 C_{Ri}} \qquad (22)$$

the bulk of liquid that has not yet reacted. Since the reactions of instantaneous and fast reactions are completed in the film, R_{Ai} becomes zero. Otherwise for slow reactions consumption in the film is negligible and it is carried out in the bulk so:

$$R_{Ai} = N_{Ai}\big|_{y=0} a - a_{Ai} r_i (\phi_L - a\delta) \tag{4}$$

Other cases, reaction occurs in both film and bulk, therfore:

$$R_{Ai} = N_{Ai}\big|_{y=\delta} a - a_{Ai} r_i (\phi_L - a\delta) \tag{5}$$

3) unsteady-state mass balance for each component in liquid phase which reacts with A_i

$$\frac{\partial(h_L x_R{}_{J,i})}{\partial t} - \frac{\partial(Lx_R{}_{J,i})}{\partial Z} = -\frac{a_R{}_{J,i}}{a_{A_i}}(N_{Ai}\big|_{y=0} a - R_{A_i}) \tag{6}$$

Analogous equations can be written for the product component like $p_{J,i}$

4) unsteady-state overall adiabatic energy balance

$$\frac{\partial(h_L H_L)}{\partial t} - \frac{\partial(LH_L)}{\partial Z} + \frac{\partial(GH_G)}{\partial Z} =$$

$$\sum_{i=1}^{nA} ((N_{Ai}\big|_{y=0}\alpha(-\Delta H_{sAi}) + (N_{Ai}\big|_{y=0}\alpha - R_{Ai})(-\Delta H_{ri})) \tag{7}$$

were

$$H_L = C_{pL}(T_L - T_{ref}) \tag{8}$$

$$H_G = C_{pG}(T_G - T_{ref}) + \lambda_m \tag{9}$$

5) gas phase energy balance

$$\frac{\partial(GH_G)}{\partial Z} = Ua(T_L - T_G) \tag{10}$$

where U is the overall heat transfer coefficient $(1/U = 1/u_L + 1/u_G)$.

In equations 7 and 10 accumulation term for gas phase was omitted due to very low heat capacity of gas phase relative to the liquid phase.

6) unsteady-\state overall mass balance in gas phase

$$\frac{\partial h_G}{\partial t} + \frac{\partial G}{\partial z} = -\sum_{i=1}^{nA} N_{Ai}\big|_{y=0} a \tag{11}$$

7) unsteady-state overall mass balance in liquid phase after neglecting the contribution of mass trasfer to the bulk liquid flow

$$\frac{\partial h_L}{\delta t} - \frac{\partial L}{\partial z} = 0 \tag{12}$$

N_{Ai} in equations 2,4 - 7 and 11 is the mass flux of component A_i and can be written based on two film theory in each time step as:

$$N_{Ai}\big|_{y=0} = \frac{(P_{Ai})_b - He_{Ai}(C_{Ai})_b}{\dfrac{1}{k_{G,Ai}} + \dfrac{He_{Ai}}{E_{Ai}k_{L,Ai}}} \tag{13}$$

E_{Ai} is the enhancement factor of the transfer phenomena due to the chemical reaction. The analytical expressions for numerical values of E_i for various reactions have been determined by different authors. Swaaij and Versteeg (1992) presented an overview of the absorption models that are available for the calculation of the mass transfer rates in gas-liquid systems with complex reactions.

Equations 2, 3, 6, 7 and 10 must be solved simultaneously to determine values of the component concentration and temperature in both phases as function of time and position along the column.

The final steady- state results are given by the solution of these equations after setting

analysis of packed absorber of carbon dioxide into an aqueous MEA solution by using the method of characteristcs for solving equations. They assumed isothermal condition in gas phase correlation.

Hitch et al. (1986) proposed a rigorous algorithm for the unsteady-state simulation of multicomponent adiabatic physical absorption in packed columns. They used an implicit integration method to solve the partial differential equations. Their simulation results(dynamic and final steady state) agreed wellwith experimental results. Lakshmanan and Potter (1989) used a numerical model namely the "cinematic" model to solve the dynamic absorption in packed and tray columns. They assumed isothermal condition, physical absorption, linear and nonlinear equilibrium and plug flow in packed bed model. Wajg et al. (1997) have used orthogonal collocation method to solve the dynamic equations describing multicomponent nonreactive and reactive distillation in packed columns. They showed that orthogonal collocation is more accurate and effcient than the finite difference method.

In this study a mathematical model is presented to describe the unsteady-state behaviour of packed absorption columns in adiabatic as well as nonisothermal conditions with chemical reaction.

The results obtained from the model have been tested and compared by experimental data obtained from the unsteady-state absorption of carbon dioxide from air into an aqueous monoethanolamine (MEA) solution in a pilot scale packed absorption column.
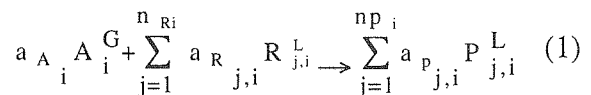
Mathematical model

The mathematical model of gas absorption process in packed columns is based upon the two film theory in each time step and simultaneous solution of differential mass and enthalpy balances of two phases along the column.

In this work the following assumptions were made:

1) The column is adiabatic and heat capacities of column and packing are negligible;
2) The pressure drop through the column is negligible compared to the overall column pressure;
3) The partial molal gas enthalpy at the interface is negligible as compared to the heat of reaction and solution;
4) Gas and liquid streams are in plug flow so that there are no radial temperature and concentration gradients;
5) Gas-liquid equilibrium is described by Henry's law.

Let the i-th chemical reaction which occures in the liquid phase be:

$$a_{A_i} A_i^G + \sum_{j=1}^{n_{Ri}} a_{R_{j,i}} R_{j,i}^L \longrightarrow \sum_{j=1}^{np_i} a_{P_{j,i}} P_{j,i}^L \quad (1)$$

A_i is the component of the gas phase which absorbs and reacts with the liquid phase component $R_{j,i}$ to yiell the products $P_{j,i}$

With the above assumptions, the basic equations of the model may be written in the following manner:

1) unsteady-state mass balance in gas phase for each component that transfers to liquid phase

$$\frac{\partial(h_G y_{Ai})}{\partial t} + \frac{\partial(G y_{Ai})}{\partial Z} = -N_{Ai}|_{y=0} a \quad (2)$$

2) unsteady-state mass balance in liquid phase for each component which transfers from gas phase

$$\frac{\partial(h_L x_{Ai})}{\partial t} - \frac{\partial(L x_{Ai})}{\partial Z} = R_{Ai} \quad (3)$$

Where R_{Ai} is the amount of A_i diffused to

Dynamic Simulation of Packed Absorption columns

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Abstract

A model has been developed for unsteady-state simulation of nonisothermal, gas absorption with chemical reaction in packed columns. The model is based upon the two film theory in each time step and finite difference technique is used to solve the governing system of partial differential equations. The constructed model is capable to predict the concentration of all components in both gas and liquid phases as well as the temperature of each phase along the bed in dynamic and the final steadystate.

Results of the model, subjected to various forcing functions were compared with the experimental data obtained from a pilot-plant gas absorption unit, where CO_2 was removed from an AIR-- CO_2 mixture into an aqueous monoethanolamine (MEA) solution.

Keywords

Dynamic simulation, gas absorption with chemical reaction, packed columns

Introduction

The basic studies on dynamic behaviour of absorption columns began half a century ago.

A theoretical investigation of the unsteady-state behaviour of continuous counter current heat and mass transfer operations including packed gas absorption columns was made by Jaswan and Smith (1954). They derived an analytical solution for the time response of the outlet gas and liquid concentrations in a packed absorption column. Gray and Pados (1963) carried out a theoretical and experimental investigation on the response of outlet gas concentration to the inlet gas concentration for the absorption of carbon dioxide from air into water in a packed column using a frequency response technique. Their equations were analytical solutions of the frequency response of gas concentration in a packed bed with counter current constant gas and liquid flows using plug flow, mixed stages and axial diffusion model.

Mc Daniel and Holland (1970) performed a dynamic modeling of packed absorber by defining the heat and mass transfer section. For each section mass and energy balances were derived by integral difference equations over the time period and transformed to algebraic forms by use of the implicit method. Bradly and Andre (1972) presented a dynamic