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Research Article

Modeling and Simulation of CO_2 Absorption into Promoted Aqueous Potassium Carbonate Solution in Industrial Scale Packed Column

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Abstract

Carbon dioxide gas is a harmful impurity which is corrosive and it can damage the utilities and the piping system in industries. Chemical absorption is the most economical separation method which is widely applied in chemical industries for CO_2 removal process. Hot potassium carbonate (K_2CO_3) is the most effective solvent that has been used extensively, especially for the CO_2 separation process from gas synthesis and natural gas. This paper aims to develop mathematical model for investigating the CO_2 absorption into promoted hot K_2CO_3 solution in industrial scale packed column in an ammonia plant. The CO_2 was removed from the gas stream by counter-current absorption in two stages column. To represent the gas-liquid system, a rigorous mathematical model based on the two-film theory was considered. The model consists of differential mass and heat balance and considers the interactions between mass-transfer and chemical kinetics using enhancement factor concept. Gas solubility, mass and heat transfer coefficients, reaction kinetics and equilibrium were estimated using correlations from literatures. The model was validated using plant data and was used to compute temperature and concentration profiles in the absorber. The variation of CO_2 recovery with respect to changes in some operating variables was evaluated. The effect of various kinds of promoters added into K_2CO_3 solution on the CO_2 recovery was also investigated. The simulation results agree well with the plant data. The results of the simulation prediction, for the absorber pressure of 33 atm with a lean flow rate of 32,0867 kg/h, temperature of 343 K, and semi lean flow rate of 2,514,122 kg/h, temperature of 385 K, showed $\%CO_2$ removal of 95.55%, while that of plant data is 96.8%. © 2015 BCREC UNDIP. All rights reserved

Keywords: Reactive Absorption; packed column; industrial scale; carbon dioxide; promoter; multicomponents

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1. Introduction

Carbon dioxide (CO_2) needs to be separated from gas stream, especially in Petrochemical, oil, and natural gas industries, because CO_2 is an acid gas and corrosive which can damage the plant utility and the piping system. CO_2 can decrease the heating value of natural gas. In liquefied natural gas (LNG) refinery, CO_2 gas has to be removed because CO_2 will freeze at very low temperature so that can block the piping system and also can damage the tube in

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the main heat exchanger. Instead of that, CO_2 is a byproduct of the ammonia industry. CO_2 is a poison to the ammonia synthesis catalyst [1], therefore CO_2 must be separated from the gas stream before heading to the ammonia synthesis unit.

A number of separation technologies could be employed for CO_2 capture. These include adsorption, physical absorption, chemical absorption, cryogenic separation and membranes [2,3]. Chemical absorption is the most practical and effective technique and is widely used in chemical and petrochemical industry [4,5]. Chemical absorption involves the reaction of CO_2 with a chemical solvent to form a weakly bonded intermediate compound which may be regenerate with the application of heat producing the original solvent and a CO_2 stream [6]. The selectivity of this form of separation is relatively high. In addition, a relatively pure CO_2 stream could be produced. These factors make chemical absorption well suited for CO_2 capture for industrial applications.

The biggest cost factor for CO_2 capture is the energy consumption for solvent regeneration, which can attain 49% of the total cost of CO_2 capture [7]. Another factor is the capacity of the solvent that can determine the amount of CO_2 absorbed per unit of solvent. Many solvents are used in the gas processing, but alkanolamine and "hot" K_2CO_3 solvents are the most effective solvents that can be used [8]. The advantage of alkanolamine is the rapid absorption rate and high solvent capacity. While the disadvantage of alkanolamine, mainly in primary and secondary groups, is the formation of a stable carbamate compounds and it requires high energy for regeneration. Moreover, it cannot separate the mercaptan compounds, the loss of a large vapor causes a high vapor pressure, degraded at overheating (>100 °C), and occur further reaction which produce byproducts that cannot be degraded [9].

Potassium carbonate (K_2CO_3) aqueous solution has been used extensively, especially for the separation of CO_2 from gas synthesis and natural gas. Because it has a high stability, low cost, and energy consumption for solvent regeneration is low, but the reaction rate is slow compared to alkanolamine [10-12]. The addition of promoter in the K_2CO_3 aqueous solution can increase the CO_2 absorption process. Potassium carbonate solution with alkanolamine as a promoter show an effective way to improve the overall performance of solvents, especially at primary and secondary groups of alkanolamine [1, 13-17].

In industries, the process of CO_2 absorption occurs at high pressure. Several components beside CO_2 can be absorbed and interaction between the components can be occurred in the gas film and liquid film that affect the mass transfer process and chemical reactions. Reactive absorption involves various components of the gas mixtures and liquid mixtures that represent multicomponent system. The properties of mass transfer in the multicomponent system are more complex than the binary mass transfer. Furthermore, it is an industrial practice to use two stages column each with different diameter to meet certain hydrodynamic characteristic requirement. Most of previous models assume only one component which moves through the interface or single component absorption and use one stage column [1, 18-20].

The objective of this study is to develop a mathematical model of CO_2 absorption in industrial scale packed column, consisting of two stages, with different diameter of each, using promoted K_2CO_3 aqueous solution by considering the absorption of carrying gases and the multicomponent diffusion model was devised. To simplify the model, the concentration of species was defined based on the influent molal flow rate.

2. Theoretical Background

2.1. Reactions Kinetic and Absorption Rate

The following elementary reactions take place when CO_2 is absorbed in potassium carbonate and bicarbonate solution [11, 21]:

$$CO_2 + OH^- \leftrightarrow HCO_3^- \tag{1}$$

$$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+ \tag{2}$$

$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+ \tag{3}$$

$$H_2O \leftrightarrow OH^- + H^+ \tag{4}$$

The overall stoichoimetric reaction can be represented in the following reaction,

$$CO_2 + CO_3^{2-} + H_2O \leftrightarrow 2HCO_3^- \tag{5}$$

Reactions (2)-(4) are assumed always in equilibrium condition. The equilibrium constant of these reactions are available in the literature [22] and are expressed as follows:

$$K_i = \exp\left(\frac{a_i}{T} + b_i \ln T + c_i T + d_i\right) \qquad (6)$$

Then,

$$K_2 = \frac{C_{HCO_3^-} C_{H^+}}{C_{CO_2,e}} = \exp\left(-\frac{12092.1}{T} - 36.7816 \ln T + 231.4\right) \qquad (7)$$

$$K_3 = \frac{C_{CO_3^{2-}} C_{H^+}}{C_{HCO_3^-}} = \exp\left(-\frac{12431.7}{T} - 35.4819 \ln T + 216.067\right) \qquad (8)$$

$$K_4 = K_W = C_{H^+} C_{OH^-} = \exp\left(-\frac{13445.9}{T} - 22.4773 \ln T + 132.932\right) \qquad (9)$$

From equations (7)-(9), the concentration of OH- ion and equilibrium concentration of CO_2 in liquid phase can be obtained as follow:

$$C_{CO_2,e} = \frac{K_3 C_{HCO_3^-}^2}{K_2 C_{CO_3^{2-}}} \qquad (10)$$

$$C_{OH^-} = \frac{K_W C_{CO_3^{2-}}}{K_3 C_{HCO_3^-}} \qquad (11)$$

Reaction (1) is the rate controlling step for CO_2 absorption in hot K_2CO_3 solution. When the forward reaction is pseudo first order and reverse reaction rate is constant then the reaction rate can be expressed as follows [1, 11, 23]:

$$r_{OH} = k_{1,OH}\left(C_{CO_2} - C_{CO_2,e}\right), \qquad k_{1,OH} = k_{OH} C_{OH^-} \qquad (12)$$

In Equation (12), is second order reaction rate constant of the forward reaction and is obtained from the following correlation [24]:

$$k_{OH} = 2.53 \times 10^{11} e^{-4311/T} \qquad (13)$$

It is industrial practice to add promoter to the carbonate-bicarbonate solution to increase the reaction rate. The reaction between several types of promoters with CO_2 is given as follows. Reaction of CO_2 with primary and secondary amine follows zwitterion mechanism with the formation of zwitterion intermediate which react with any base, B to form carbamate and or bicarbonate. Reaction of CO_2 with monoethanolamine (MEA) [25, 26]:

$$CO_2 + RNH_2 \leftrightarrow RNH_2^+COO^- \qquad (14)$$

$$RNH_2^+COO^- + B \leftrightarrow RNHCOO^- + BH^+ \qquad (15)$$

$$RNHCOO^- + H_2O \leftrightarrow RNH_2 + HCO_3^- \qquad (16)$$

$$K = \frac{C_{RNH_2} C_{HCO_3^-}}{C_{RNHCOO^-}} = exp(-3.4 - 5851/T)$$

Reaction of CO_2 with diethanolamine (DEA) [14, 27, 28]:

$$CO_2 + R_2NH \leftrightarrow R_2N^+HCOO^- \qquad (17)$$

$$R_2NH_2^+ \leftrightarrow R_2NH + H^+$$

$$K = \frac{C_{R_2NH} C_{H^+}}{C_{R_2NH_2^+}} = exp\left(\frac{-3071.15}{T} + 6.776904 \ln T - 48.7594\right)$$

$$R_2N^+HCOO^- + OH^- \leftrightarrow R_2NH + HCO_3^- \qquad (18)$$

$$K = \frac{C_{R_2NH} C_{HCO_3^-}}{C_{R_2NHCOO^-} C_{OH^-}} = exp\left(-\frac{17067.2}{T} - 66.8007 \ln T + 439.709\right)$$

Reaction of CO_2 with Arginine [29]:

$$CO_2 + H_2N - CHR' - COO^-K^+ \rightleftarrows \ {}^-COO^+H_2N$$

$$-CHR' - COO^-K^+ \tag{19}$$

$$^-COO^+H_2N - CHR' - COO^-K^+ + B \rightarrow \; ^-COOHN - CHR' - COO^-K^+ + BH^+ \tag{20}$$

Reaction of CO_2 with boric acid can be described through the following mechanism [30]:

$$B(OH)_3.H_2O \leftrightarrow B(OH)_4^- + H^+ \tag{21}$$

$$B(OH)_4^- + CO_2 \leftrightarrow B(OH)_4CO_2^- \tag{22}$$

$$B(OH)_4CO_2^- + H_2O \leftrightarrow B(OH)_3.H_2O + HCO_3^- \tag{23}$$

Reactions (14), (17), (19) and (22) are the controlling step for the reaction between CO_2 with promoter MEA, DEA, Arginine and Boric acid respectively. When the forward reaction is pseudo first order and reverse reaction rate is constant then the reaction rate can be expressed as follows:

$$r_{cat} = k_{1,cat}\left(C_{CO_2} - C_{CO_2,e}\right), \quad k_{1,cat} = k_{cat}C_{cat} \tag{24}$$

where k_{cat} is second order reaction rate constant of the forward reaction and is obtained from Table 1.

If the operating condition of absorption process is in fast reaction regime, then, the absorption rate can be obtained from the following equation:

$$N_{CO_2}a = E\, k_{L,CO_2}a\left(C^*_{CO_2} - C_{CO_2,e}\right) \tag{25}$$

where the enhancement factor (E) is obtained from Equation (26).

$$E = H_a \tag{26}$$

In Equation (26), H_a is Hatta number which can be obtained from equation (27) and k_1 is overall pseudo first order reaction rate constant defined as Equation (28):

$$H_a^2 = \frac{D_A k_1}{k_{L,CO_2}^2} \tag{27}$$

$$k_1 = k_{OH}C_{OH} + k_{cat}C_{cat} \tag{28}$$

Fast pseudo first order reaction assumption is valid under the following condition:

$$3 < H_a < 0.5\, E_\infty \tag{29}$$

where E_∞ is enhancement factor for instantaneous reaction which can be obtained using film theory described in Equation (30):

$$E_\infty = 1 + \frac{D_{CO_3^{2-}}C_{CO_3^{2-}}}{D_{CO_2}C^*_{CO_2}} \tag{30}$$

Table 1. Second order reaction rate constant of various promoters

No	Catalyst	k_{cat} [m^3.kmole^{-1}.s^{-1}]	Reference
1	MEA	$k_{MEA} = 4.24\text{x}10^9\, e^{-3925/T}$	[24]
2	DEA	$k_{DEA} = 5.65\text{x}10^{11}\, e^{-5284.4/T}$	[19]
3	Boric Acid	$k_{boric} = 5.5\text{x}10^{11}\, e^{-6927/T}$	[30]
4	Arginine	$k_{Arg} = 2.58\text{x}10^{16}\, e^{-8645/T}$	[29]

2.2. Gas Solubilities

The solubility of gases in promoted carbonate solutions were estimated using modified Henry law with empirical model of Schumpe which describes the solubility of gases in mixed electrolytes solutions considering salting out effects:

$$log\left(\frac{H_{e,jw}}{H_{e,j}}\right) = \sum (h_i + h_G)c_{i,L} \tag{31}$$

where h_i is the ion-specific parameter (m^3/kmole), h_G is the gas-specific parameter (m^3/kmole) and $c_{i,L}$ is the concentration of ion i (kmole/m^3). The Henry constant of gas-water system (can be obtained from Equation (32):

$$H_{e,jw}(T) = H_{e,jw}(298\ K) * exp\left(\frac{-d\ ln\ k_H}{d(\frac{1}{T})} * \left(\frac{1}{T} - \frac{1}{298}\right)\right) \tag{32}$$

The value of $H_{e,jw}$ (298 K) and $-d$ ln k_H/d(1/T) are shown in Table 2 [31]. Gas-specific parameter was extended from Equation (31) to wider temperature range using Weissenberger and Schumpe method [30] and expressed in Equation (33).

$$h_G = h_{G,0} + h_T(T - 298.15) \tag{33}$$

In equation (33), h_T is the temperature correction (m^3/kmole.K). The values of h_{i^+}, h_{i^-}, $h_{G,0}$, and h_T can be seen in Table 3 [32] and Table 4 [32, 33]. Equations (32) and (33) are substituted into Equation (31) to obtain the value of $H_{e,j}$.

2.3. Mass and Heat Transfer Coefficient

Gas side mass transfer coefficient is obtained from the empirical correlation by Onda *et al.* [34] shown in Equation (34) where constant A equal to 2 for packing diameter less than 0.012 m and equal to 5.23 for packing diameter greater than 0.012 m.

$$k_G = \frac{A}{RT}(R_{eG})^{0.7}(S_{cG})^{1/3}(a\ d_p)^{-2.0}(a\ D_{kG}) \tag{34}$$

Reynold and Schmidt number are defined as, respectively:

$$R_{eL} = \frac{L}{a\ \mu_L} \qquad \text{and} \qquad S_{cL} = \frac{\mu_L}{\rho_L D_{kL}}$$

Liquid side mass transfer coefficient is obtained from the empirical correlation by Taylor and Krishna [35] shown in Equation (35):

$$k_{L,k} = 0.0051\ (R_{eL})^{2/3}(S_{cL})^{-0.5}(a_p d_p)^{0.4}\left(\frac{\mu_L g}{\rho_L}\right)^{1/3} \tag{35}$$

The liquid Reynold and Schmidt number are defined as, respectively:

$$R_{eL} = \frac{L}{a\ \mu_L} \qquad \text{and} \qquad S_{cL} = \frac{\mu_L}{\rho_L D_{kL}}$$

Gas-liquid interfacial area per unit volume of packed column, *a*, is obtained from packing specific area from the correlation provided by Onda *et al.* [34].

$$\frac{a}{a_p} = 1 - exp\left[-1.45\left(\frac{\sigma_c}{\sigma_L}\right)^{0.75}(R_{eL})^{0.1}(F_{rL})^{-0.05}(W_{eL})^{0.2}\right] \tag{36}$$

In Equation (36), Froude and Weber number are defined as, respectively:

$$F_{r,L} = \frac{a_p L^2}{\rho_L^2 g} \qquad \text{and} \qquad W_{e,L} = \frac{L^2}{\rho_L a_p \sigma}$$

Diffusion coefficient of species in gas phases was determined from binary diffusion coefficient using Maxwell-Stevan equation as follows:

$$\frac{1}{D_{im}} = \frac{\sum\left(\frac{1}{D_{ij}}\right)(x_j N_i - x_i N_j)}{N_i - x_i \sum N_j} \tag{37}$$

where the binary diffusion coefficient was obtained from correlation by Fuller *et al.* [36] recommended by Taylor and Krishna [35], Reid *et al.* [37], and also Daubert and Danner [38].

Table 2. The values of $H_{e,jw}$ (298 K) and $-d \ln k_H / d(1/T)$ for various gases [31]

Component	$1/He^{o}_{298\,K}$ (kmole/m^3.Pa)	$-d \ln k_H/d(1/T)$ (K)
CO_2	3600	2200
CO	99	1300
H_2	78	500
N_2	61	1300
CH_4	140	1600
Ar	140	1500

Table 3. The value of gas parameters [32]

Component	$h_{G,0}$ (m^3/kmole)	h_T (m^3/kmole.K)
CO_2	-1.72×10^{-5}	-3.38×10^{-7}
CO	-	-
H_2	-2.18×10^{-5}	-2.99×10^{-7}
N_2	-1×10^{-6}	-6.05×10^{-7}
CH_4	2.2×10^{-6}	-5.24×10^{-7}
Ar	5.7×10^{-6}	-4.85×10^{-7}

Table 4. The value of ion specific parameter [32, 33]

Cation	h_{i^+} (m^3/kmole)	Anion	h_{i^-} (m^3/kmole)
K^+	0.0922	HCO_3^-	0.0967
$MEAH^+$	0.0550	CO_3^{2-}	0.1423
$DEAH^+$	0.0470	Arg^-	0.1452
		OH^-	0.0610
		$MEACOO^-$	0.0540
		$DEACOO^-$	0.0430

$$D_{ij,G} = \frac{1\times 10^{-7}T^{1.75}\left(\frac{1}{M_i}-\frac{1}{M_j}\right)}{P\left(v_i^{1/3}+v_j^{1/3}\right)^2} \tag{38}$$

Due to dilute solution condition, diffusion coefficient of species in liquid phase was assumed binary with respect to water and determined using Wilke and Chang Equation [23]:

$$D_{iw,L} = \frac{7.4\times 10^{-8}T\,(\phi M_w)^{0.5}}{\mu_L v_i^{0.6}} \tag{39}$$

Heat transfer coefficient in gas phase was determined from mass transfer coefficient in gas phase using Chilton-Colburn analogy, while the heat transfer resistance in liquid phase was neglected.

3. Method

3.1 Mathematical Model

This study was conducted with the theoretical approach (simulation) by developing mathematical model for heat and mass transfer phenomena accompanied by chemical reaction in CO_2 removal process at non-isothermal condition using promoted K_2CO_3 aqueous solution. The model is based on the following assumptions: steady state and adiabatic operations, plug-flow pattern for gas and liquid, neglected amount of solvent evaporation and constant pressure throughout the column.Thereaction in the liquid phase is fast enough for a substantial amount of the gas absorbed to react in the liquid film, rather than to be transfered unreacted to the bulk. Mathematical model development was conducted by constructing differential mass balances in the packed column. Figure 1 shows schematic diagram of

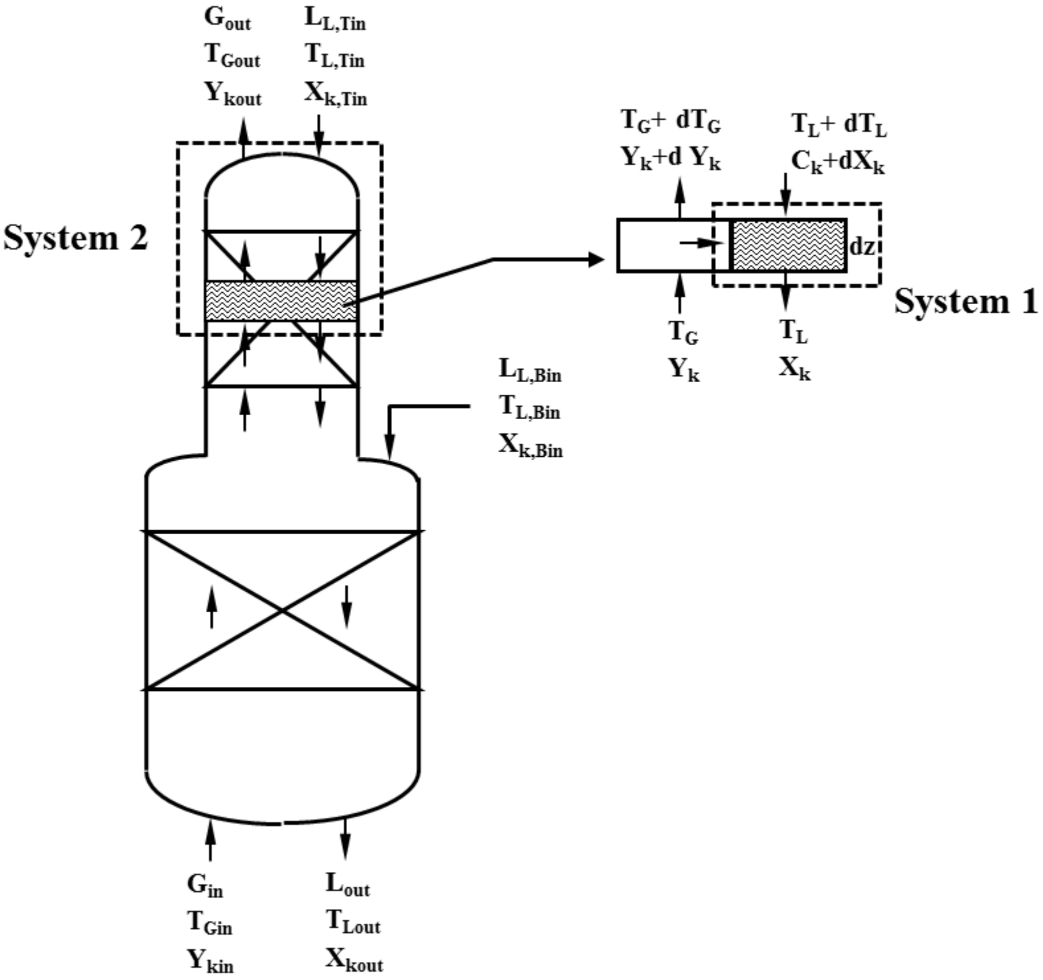


Figure 1. Schematic diagram of packed bed absorption column showing an infinitesimal element for mass and energy balances

packed bed absorption column showing an infinitesimal element for mass and energy balances. Microscopic or differential mass and energy balance was constructed based on System I, while macroscopic balance to correlate several process variables in packed column was constructed through System II (see Figure 1).

Differential Mass Balances :
System I

CO_2 :

$$N_{CO_2} a\, A\, dz = r_{CO_2} \emptyset_L A\, dz \quad (40)$$

K_2CO_3:

$$-L_{in} dX_{CO_3^{2-}} - \nu\, r_{CO_2} \phi_L A\, dz = 0 \quad (41)$$

Equation (31) was substituted to Equation (32):

$$-L_{in} dX_{CO_3^{2-}} - \nu\, N_{CO_2} a\, A\, dz = 0 \quad (42)$$

$$\frac{dX_{CO_3^{2-}}}{dz} = -\frac{\nu N_{CO_2} aA}{L_{in}} \quad (43)$$

When $N_{CO2}a$ was substituted from Equation (25), then the following equation was obtained after some rearrangements:

$$\frac{dX_{CO_3^{2-}}}{d\zeta} = -\frac{\nu E k_{L,CO_2} aC \left(x^*_{CO_3^{2-}} - x^e_{CO_3^{2-}}\right) Z_T A}{L_{in}} \quad (44)$$

where $\zeta = Z/Z_T$ and $x_{CO_3^{2-}} = C_{CO_3^{2-}}/C$

k (dissolved gas): $L_{in}\, dX_k = N_k a\, A\, dz$ (45)

$$\frac{dX_k}{dz} = \frac{N_k aA}{L_{in}} \quad (46)$$

where $N_k a$ was obtained from $N_k a = k_{L,K} a(C_k^* - C_k^o)$, and Equation (46) can be rearranged as follows:

$$\frac{dX_k}{d\zeta} = \frac{k_{L,k} aC\, (x_k^* - x_k^0) Z_T\, A}{L_{in}} \quad (47)$$

The concentration of bicarbonate ion was determined using stoichiometry as follows:

$$X_{HCO_3^-} - X_{HCO_3^-,in} = 2\left[X_{CO_3^{2-},in} - X_{CO_3^{2-}}\right] \quad (48)$$

The concentration of other species (expressed as mole ratio) in liquid phase was determined using equilibrium and electro neutrality constrain. Molar concentration of various species in liquid phase was determined from $C_i = x_i\, C$, where mole-fraction, x_i, was determined from mole ratio as follows:

$$x_i = \frac{X_i}{\sum X_k} y_i = \frac{Y_i}{\sum Y_k} \quad (49)$$

and molar density, C, was calculated from liquid mass density as $C = \rho_L/M$, where M is molecular weight of liquid mixture, $M = \Sigma M_i\, x_i$, and ρ is its density which was obtained by regression fitting of data from literature [39] as follows:

$$\rho_L = \rho_{L,293} + 17816.45\left(\frac{1}{T} - \frac{1}{293}\right) \quad (50)$$

$$\rho_{L,293} = \frac{49196.07}{49.693 - 39.1902\, w_{K_2CO_3} - 29.4723\, w_{KHCO_3}} \quad (51)$$

The presence of catalyst in the liquid phase does not affect significantly liquid density.

The concentration of CO_2 and carrying gases in the gas phase can be obtained by performing a mass balance over System II:
CO_2:

$$G_{m,in}\left[Y_{CO_2} - Y_{CO_2,out}\right] = \frac{L_{m,in}}{\nu}\left[X_{CO_3^{2-},in} - X_{CO_3^{2-}}\right] \quad (52)$$

k (carrying gases):

$$G_{m,in}\left[Y_k - Y_{k,out}\right] = L_{m,in}\left[X_{k,in} - X_k\right] \quad (53)$$

CO_2 concentration on the interface (C^*_{CO2}):

$$C^*_{CO_2} = \frac{k_{G,CO_2} y_{CO_2} P + E\, k_{L,CO_2} C_{CO_2,e}}{E\, k_{L,CO_2} + k_{G,CO_2} H_{CO_2}} \quad (54)$$

Concentration of other gases on the interface (C^*_k):

$$C^*_k = \frac{k_{G,k} y_{CO_2} P + k_{L,k} C_k}{k_{L,k} + k_{G,k} H_k} \quad (55)$$

Differential heat balance on the gas side is given in Equation (56):

$$\frac{dT_G}{dz} = -\frac{h_G a}{C_{pG} G}(T_G - T_L) \quad (56)$$

while liquid temperature was calculated from energy balance for System II:

$$T_L = T_{L,in} + \frac{G\, C_{pG}}{C_{pL}}\left[c_{pG}(T_G - T_{G,in})\right] -$$

$$(-\Delta H_{rx})\frac{G_{in}}{L\,C_{pL}}\left[Y_{CO_2,in}-Y_{CO_2}\right] \tag{57}$$

3.2. Numerical Solution

Equations (44) and (47) were solved numerically using orthogonal collocation method with 6 internal collocation points, thus:

$$X_{CO_3^{2-},j} = X_{CO_3^{2-},in} - \frac{v\,Z_T}{H_{T,CO_3^{2-}}} \sum_{m=1}^{NC+2} H_{jm} E_m \left(x^*_{CO_3^{2-},m} - x^e_{CO_3^{2-},m}\right) \tag{58}$$

$$X_{k,j} = X_{k,in} + \frac{Z_T}{H_{T,k}} \sum_{m=1}^{NC+2} H_{jm}\left(x^*_{k,m} - x^0_{k,m}\right) \tag{59}$$

where:

$$H_{T,CO_3^{2-}} = \frac{L_{in}}{A\,k_{L,CO_3^{2-}}\,a\,C}\;,\;H_{T,k} = \frac{L_{in}}{A\,k_{L,k}\,a\,C} \tag{60}$$

With collocation orthogonal method on Equation (56), the result for gas temperature distribution in the column was shown by Equation (61):

$$T_{G,j} = T_{G,out} - N_G \sum_{m=1}^{NC+2} H_{jm}\left(T_{G,m} - T_{L,m}\right) \tag{61}$$

where N_G was defined as:

$$N_G = \frac{h_G a\,Z_T}{C_{pG}\,G} \tag{62}$$

The solution of nonlinear algebraic equation obtained from orthogonal collocation method was conducted by successive approximation method. Thus, % CO_2 removal can be calculated by following equation:

$$\%\,Removal = 1 - \frac{Y_{A,out}}{Y_{A,in}} \tag{63}$$

4. Results and Discussion

This study was carried out by constructing a simulation program for CO_2 gas absorption process with reversible reaction at non-isothermal condition. The system studied was packed column in a large-scale ammonia plant to remove CO_2 from the process gas stream containing 18.47% CO_2, 59.88% H_2, 20.82% N_2 , 0.32% CH_4 , 0.25% CO and 0.25% mole Argon with flow rate of 204020 kg/h. The CO_2 is removed from the gas stream by counter-current absorption in two stages column, lower part with diameter of 3.89 m and upper part with diameter of 2.365 m. The lower part column is filled with 7 cm IMPT packing to the height of 18.29 m, while the upper part is filled with 5 cm IMPT packing to the height of 15.85 m. Lean solution, containing 21.22 % K_2CO_3 and 5.60 % $KHCO_3$, was fed into the top of upper part column while semi lean solution containing 18.12 % K_2CO_3 and 9.76 % $KHCO_3$ was fed into the top of lower part column. To enhance the absorption rate, an amine promoter, DEA, was added into the carbonate-bicarbonate solution. It can be seen from Table 5 that for the same operating condition the predicted percent CO_2 removal is 95.5465 compared to 96.8 % in the ammonia plant.

Table 5. Comparison between simulation result and ammonia plant data

Variable comparison	Simulation	Ammonia plant data
Flow rate of gas (kg/h)	204020	204020
Flow rate of lean solution (kg/h)	320867	320867
Flow rate of semi lean solution (kg/h)	2514122	2514122
Temperature of gas (K)	362	362
Temperature of lean solution (K)	343	343
Temperature of semi lean solution (K)	385	385
Pressure (atm)	33	33
Promotor	DEA	DEA
CO_2 removal (%)	95.3465	96.8

4.1. Concentration Distribution in Packed Column

Predicted concentration distribution of dissolved gases, K_2CO_3 and $KHCO_3$ in liquid phase in packed column can be seen in Figures 2-4. Figure 2 shows that the concentration of CO_2 gas in the liquid increases because it is absorbed by K_2CO_3 aqueous solution from 0 (no CO_2 in K_2CO_3 solution) to 1.98x10^{-1} kmole/m^3, beside that the composition of other gas such as CH_4, CO, H_2, N_2 and Ar increase. Axial position 0 is the position at the bottom of top packing while the axial position 1 is the position at the top. The liquid effluent from the top packing mixed with the semi lean solution is the influent of the bottom packing. In this packing the concentration of dissolved gases in liquid phase increases down the column. This phenomenon is shown in Figure 3. The liquid effluent from the bottom packing contains 5.9588×10^{-5} kmole/m^3 CO, 4.1834×10^{-2} kmole/m^3 H_2, 6.9029×10^{-3} kmole/m^3 N_2, 1.7063×10^{-4} kmole/m^3 CH_4, and 1.2599×10^{-4} kmole/m^3 argon. It shows that not only CO_2 gas are dissolved in K_2CO_3 solution, but also other gases, such as: CO, H_2, N_2, CH_4 and Ar, are also dissolved in K_2CO_3 solution in significant amount to affect the purity of recovered CO_2 in stripping column. Increasing pressure will increase the concentration of these gases in liquid solution.

The predicted concentration distribution of K_2CO_3 and $KHCO_3$ are shown in Figure 4 both for top packing and bottom packing. From Figure 4, it can be seen that the K_2CO_3 concentration decreases as liquid moves down the col-

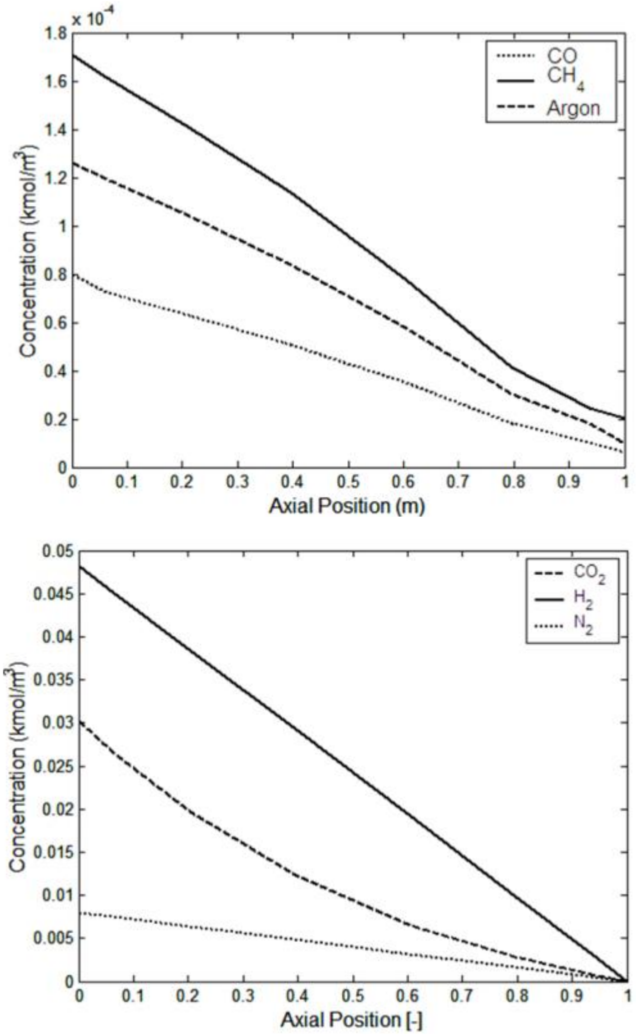


Figure 2. Concentration distribution of dissolved gases in Top Packing

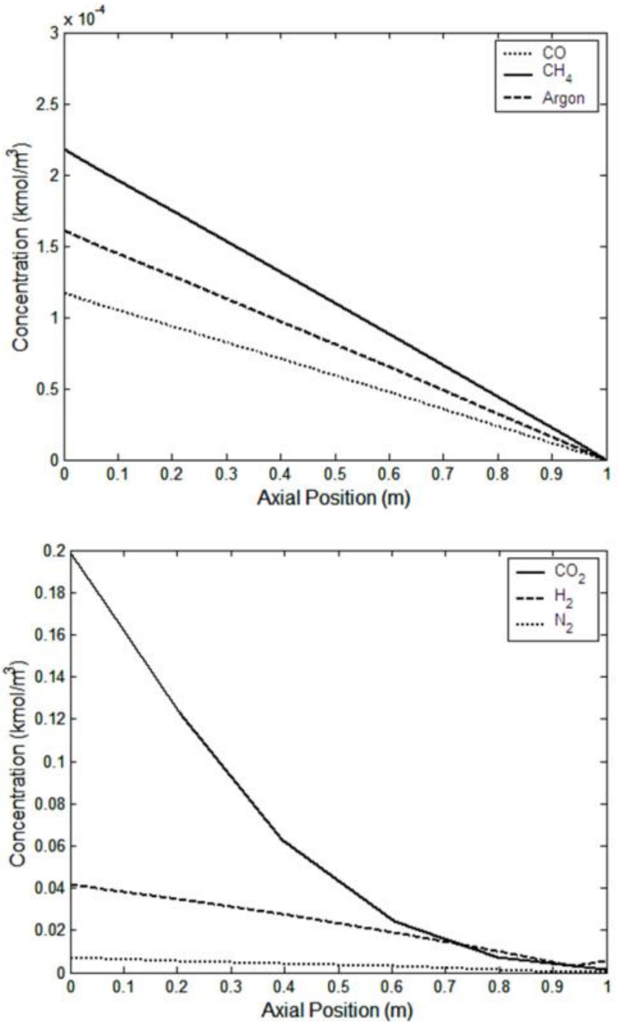
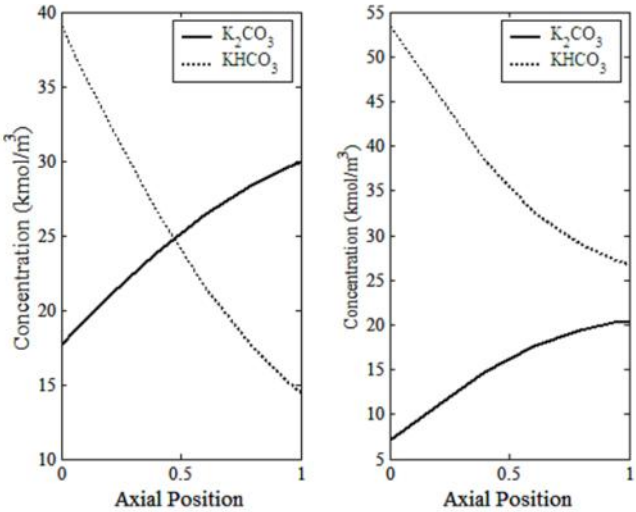


Figure 3. Concentration distribution of dissolved gases in lower section of the column



(a) (b)

Figure 4. Concentration distribution of K_2CO_3 and $KHCO_3$ in: a) column upper section, and b) column lower section

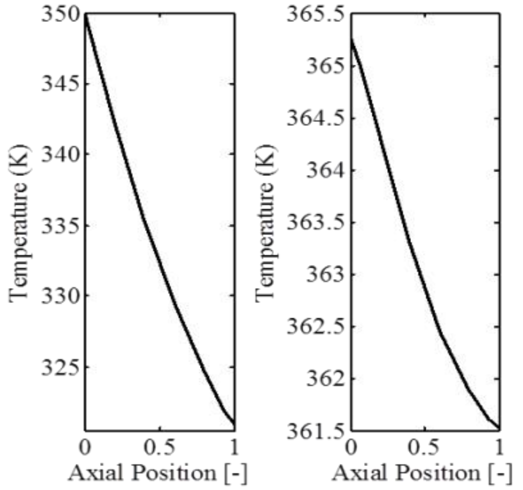
umn, where the initial concentration of K_2CO_3 is 29.986 and the outlet K_2CO_3 concentration is 6.959 kmole/m^3. It is because the K_2CO_3 solution reacts with CO_2 gas in a packed column forming $KHCO_3$. Thus, $KHCO_3$ concentration increases from 14.393 to 53.853 kmole/m^3 as the liquid stream flows from the top to the bottom of the column.

4.2. Temperature Distribution

The predicted liquid temperature distribution in packed column is shown in Figure 5. At the upper section of the column, the liquid temperature increase around 29 K due to the exothermic nature of the reactive absorption process, while at the lower part section the liquid temperature increase around 4 K.

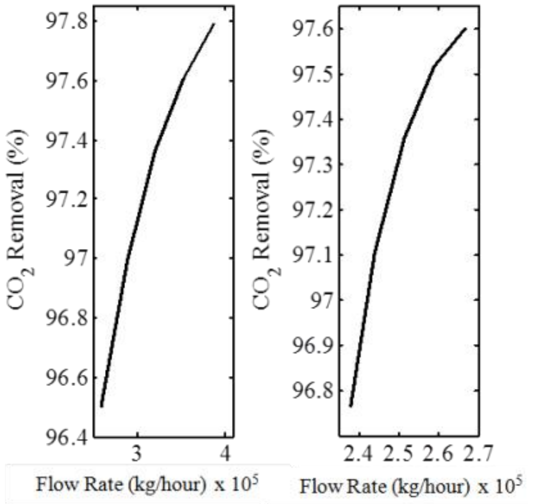
4.3. The Effect of Absorbent Flow Rate on %CO_2 Removal

The effect of absorbent flow rate on % CO_2 removal from simulation result is shown in Figure 6 under the operation conditions of 30% K_2CO_3 solution and DEA concentration of 3%. From Figure 6, it can be seen that the increase of the absorbent flow rate (lean solution and semi lean solution) give significant effect on the increase of %CO_2 removal. It indicates that the liquid side resistance has a considerable effect on the process of CO_2 absorption in K_2CO_3 aqueous solution. Although in this simulation the mass transfer resistance of gas side is also counted. The increase of absorbent flow rate will increase the turbulence and driving force and shorten life time of liquid film [40], consequently the mass transfer coefficient increases so that CO_2 absorption increases. The liquid side mass transfer coefficient is proportional to liquid flow rate to the power of 2/3 [35].



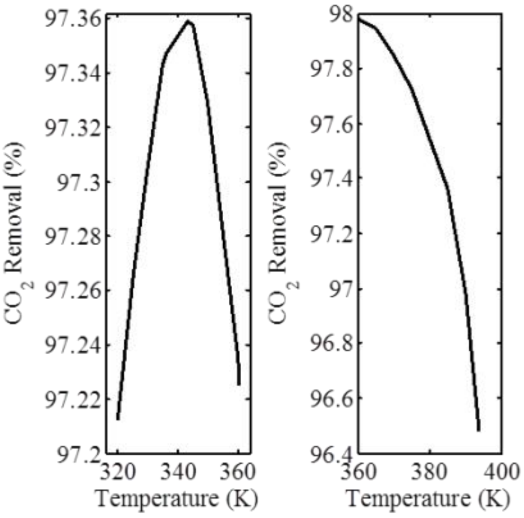
(a) (b)

Figure 5. Liquid temperature distribution: (a) column upper section, and (b) column lower section



(a) (b)

Figure 6. (a) The effect of lean solution flow rate on %CO_2 removal for semi lean solution flow rate = 2514122 kg/h; (b) The effect of semi lean solution flow rate on %CO_2 removal for lean solution flow rate = 320867 kg/h. Gas flow rate = 204020 kg/h, pressure = 33 atm, temperature of inlet lean solution= 343 K, temperature of inlet semi lean solution = 385 K, and temperature of inlet gas = 362 K



(a) (b)

Figure 7. (a) The effect of lean solution temperature on %CO_2 removal for semi lean solution temperature of 385 K; (b) The effect of semi lean solution temperature on %CO_2 removal for lean solution temperature of 343 K

4.4. The Effect of Temperature on %CO_2 Removal

The effect of temperature (lean solution and semi lean solution) on %CO_2 removal from simulation result can be seen in Figure 7. Figure 7 represents that the temperature gives a considerable effect in %CO_2 removal, indicating that the process of absorption of CO_2 into promoted K_2CO_3 solution is sensitive to temperature change. A higher temperature resulting in a higher reaction rate constant according to the Arrhenius equation, a higher diffusivity and a lower gas solubility. From Figure 7a, it indicates that at temperature less than 343 K reaction rate and diffusivity factor are more important than the gas solubility factor, therefore CO_2 removal efficiency increases with increasing lean solution temperature. Cullinane and Rochelle [41] also reported the same results for this temperature range. However, at temperature more than 343 K the solubility factor is more important than reaction rate and diffusivity factors that the CO_2 removal efficiency decrease with further increasing lean solution temperature. The highest CO_2 removal efficiency is 97.3599 % at lean solution temperature of 343 K. Thus, the increase of absorption rate depends on the relative effect of temperature on the reaction rate constants, diffusivity and solubility of gas absorbed. Hence, it is beneficial for a reactive absorption, to increase temperature to some extent for higher CO_2 removal efficiency. Figure 7b indicates that CO_2 removal efficiency decreases with increasing semi lean solution above 360 K.

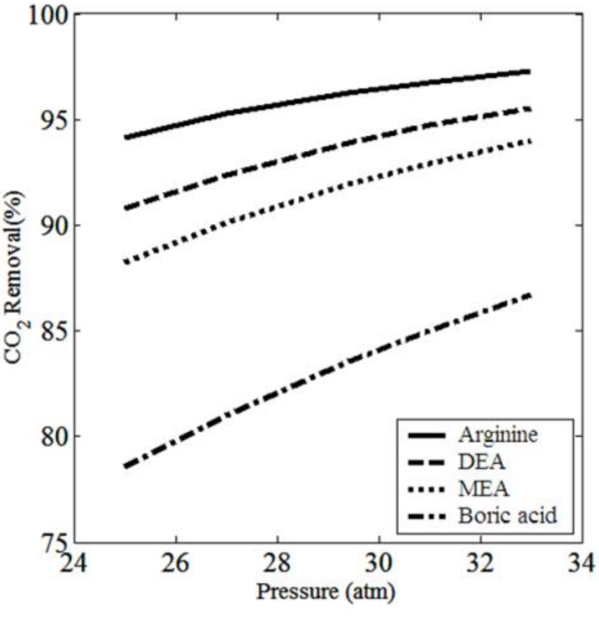


Figure 8. The effect of absorber pressure on %CO_2 removal

4.5. The Effect of Absorber Pressure and Promoter Types on % CO_2 Removal

The effect of absorber pressure on %CO_2 removal from simulation result can be seen in Figure 8. Figure 8 shows that CO_2 removal efficiency increases with increasing of operation pressure of the absorption column. This is due to the higher pressure will increase gas solubility and therefore the absorption rate. Figure 8 show that increasing pressure from 25 atm to 33 atm will increase the CO_2 removal efficiency for various promoters. At pressure of 25 atm, the CO_2 removal efficiency for promoter arginine, DEA, MEA, and boric acid are 94.1554%; 90.7901%; 88.2275%; and 78.5847%, respectively. Meanwhile, at pressure of 33 atm the CO_2 removal efficiency are 97.2671%; 95.5465%; 94.0046%; and 86.6837%, respectively. These show that Arginine gives the highest CO_2 removal efficiency. The reason for this is that Arginine contains a primary amine group which have fast reaction rate with carbon dioxide [29].

6. Conclusions

In this study, a mathematical model has been developed for investigating the CO_2 absorption into promoted hot potassium carbonate (K_2CO_3) solution in industrial scale packed column consisting of two sections, lower section with diameter larger than upper section. The model was based on two-film theory incorporating reaction-diffusion interaction through enhancement factor concept. Heat effect due to exothermic chemical reaction and the absorption of carrying gases such as H_2, N_2, CO, CH_4 and Argon were taken into account in the model. In this case the multi-component diffusion model was devised. The model was used to simulate CO_2 removal from synthesis gas at packed absorber column in an ammonia plant using hot K_2CO_3 solution promoted by DEA. The result of the simulation prediction for the absorber pressure of 33 atm with a lean flow rate of 320,867 kg/h, temperature of 343 K and semi lean flow rate of 2,514,122 kg/h, temperature of 385 K showed %CO_2 removal of 95.55%, while the plant data showed 96.8% CO_2 removal. This result shows that the accuracy of our prediction model is high with 1.29% error compared with plant data. The effect of various process variables such as flow rate of lean solution and semi lean solution, temperature, and pressure of the absorber on CO_2 removal efficiency was investigated. The effect of various kinds of promoters (such as Arginine, DEA, MEA and Boric Acid) added into K_2CO_3 solution on the CO_2 recovery was also investigated.

Nomenclatures

A : Sectional area of the column, m^2
a : Gas-liquid interfacial area per unit volume of packed column, m^2.m^{-3}
a_p : Specific area of packing, m^2.m^{-3}
C : Molar density, kmole.m^{-3}
C^*_{CO2} : CO_2 concentration at interface, kmole.m^{-3}
$C^*_{CO2,e}$: Equilibrium concentration of CO_2 in liquid phase, kmole.m^{-3}
C_i : Molar concentration of component *i*, kmole.m^{-3}
$C_{i,L}$: Concentration of ion *i*, kmole.m^{-3}
C^*_k : Molar concentration of component *k* at interface, kmole.m^{-3}
C^o_k : Molar concentration of component *k* in the bulk liquid phase, kmole.m^{-3}
C_{pG} : Heat capacity of gas, Joule.kg^{-1}.K^{-1}
C_{pL} : Heat capacity of liquid, Joule.kg^{-1}.K^{-1}
D : Diffusivity, m^2.s^{-1}
d_p : Packing diameter, m
E : Enhancement factor
E_∞ : Enhancement factor for instantaneous reaction
F_{rL} : Froude number
G : Mass velocity of gas, kg.m^{-2}.s^{-1}
g : Gravity acceleration, m.s^{-2}
H_a : Hatta number as defined by Equation (27)
H_{CO2} : Henry constant of CO_2 in aqueous electrolyte solution system, Pa.m^3.kmole^{-1}
$H_{e,j}$: Henry constant of gas-aqueous electrolyte solution system, Pa.m^3.kmole^{-1}
$H_{e,jw}$: Henry constant of gas-water system, Pa.m^3.kmole^{-1}
H_{jm} : Quadrature weight
$H_{T,k}$: Height of transfer unit for component *k*
h_G : Gas-spescific parameter, m^3.kmole^{-1}
$h_{G,0}$: Gas-spescific parameter at 298.15 K, m^3.kmole^{-1}
h_i : Ion-spescific parameter, m^3.kmole^{-1}
h_{i^+} : Cation-spescific parameter, m^3.kmole^{-1}
h_{i^-} : Anion-spescific parameter, m^3.kmole^{-1}
h_T : Temperature correction, m^3.kmole^{-1}.K^{-1}
K : Equilibrium constant
k_{cat} : Second order reaction rate constant of the forward reaction with catalyst, m^3.kmole^{-1}.s^{-1}
k_G : Mass transfer coefficient of gas side, kmole.m^{-2}.s^{-1}
k_H : The reverse of Henry Constant, kmole.m^{-3}.Pa^{-1}
k_L : Mass transfer coefficient of liquid side, kmole.m^{-2}.s^{-1}
k_{OH} : Second order reaction rate constant of the forward reaction with OH-, m^3.kmole^{-1}.s^{-1}
k_1 : Overall pseudo first order reaction rate constant, s^{-1}
L : Mass velocity of liquid, kg.m^{-2}.s^{-1}
L_{in} : Inlet molar flow rate of liquid, kmole.s^{-1}
M_i : Molecular weight of component i, kg.kmole^{-1}
N_{CO2} : Molar flux (absorption flux) of CO_2, kmole.m^{-2}.s^{-1}
N_G : Dimensionless quantity defined in Equation (62)
N_i :Molar flux of component i, kmole.m^{-2}.s^{-1}
R : Gas constant in Joule.kmole^{-1}.K^{-1}
R_{eG} : Reynold number of gas
R_{eL} : Reynold number of liquid
r_{cat} : Reaction rate of CO_2 with catalyst, kmole.m^{-3}.s^{-1}
r_{CO2} : Reaction rate of CO_2, kmole.m^{-3}.s^{-1}
r_{OH} : Reaction rate of CO_2 with OH^-, kmole.m^{-3}.s^{-1}
S_{cG} : Schmidt number of gas
S_{cL} : Schmidt number of liquid
T : Temperature, K
T_G : Temperature of gas stream, K
$T_{G,m}$: Temperature of gas in collocation points in packed column, K
T_L : Temperature of liquid stream, K
$T_{L,m}$: Temperature of liquid in collocation points m in packed column, K
v : Stoichiometric coefficient of K_2CO_3
v_i : Molar volume of component *i*, m^3.kmole^{-1}
W_{eL} : Weber number
w_i : Mass fraction of component *i*
$x^*_{CO_3^{2-}}$: Mole fraction of CO_3^{2-} at interface
$x^e_{CO_3^{2-}}$: Equilibrium mole fraction of CO_3^{2-} in liquid phase
x_k^* : Mole fraction of component *k* at interface
$x^*_{k,m}$: Mole fraction of component *k* at interface in collocation points *m* in packed column
x_k^o : Mole fraction of component *k* in the bulk liquid phase
$x^o_{k,m}$: Mole fraction of component *k* in the bulk liquid phase in collocation points *m* in packed column

x_i : Mole fraction of component *i* in liquid phase
X_i : Molar ratio of component *i* (mole component *i* per mole inlet liquid)
y_i : Mole fraction of component *i* in gas phase
Y_i : Molar ratio of component *i* (mole component *i* per mole inlet gas)
z : Axial position in packed column, m
Z_T : Height of packing, m

Greek letters

ΔH_{rx} : Heat reaction, Joule.kmole^{-1}
ζ : Dimensionless axial position in packed column
μ : Viscosity, Pa.s
ρ : Density, kg.m^{-3}
σ_C : Critical surface tension, N/m
σ_L : Surface tension of liquid, N/m
ϕ : Association factor for solvent ($\phi = 2.6$ for water)
ϕ_L : Liquid hold up in packed column

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