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Modelling Studies on Reactive Absorption of Carbon Dioxide in Monoethanolamine Solution from Flue Gas in Coal Based Thermal Power Plants

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ABSTRACT

In this paper the detailed theoretical investigation on absorption of carbon dioxide, from flue gas in coal based thermal power plants has been presented. For absorption studies, monoethanolamine solution is considered as a solvent. The mathematical model for the absorption column has been developed by considering thin film model approach. Unified method is used for an overall estimation of carbon dioxide absorption. The carbon dioxide concentration profile at a given stage, using the thin film layer model, has been predicted at 298 K and 318 K. From the results it is noted that carbon dioxide concentration decreases from interface concentration at equilibrium to a minimum of 0 kmol/m^3 up to a distance of ±2 micrometers. Overall estimation of carbon dioxide absorption has been carried out using the unified model approach. The total amount of carbon dioxide absorbed in absorption column is estimated to be 95.60% of the inlet carbon dioxide with 30 trays, *L/G* ratio of 8.5 and carbon dioxide flow rate of 95.74 kmol/m^3. The results revealed that reactive absorption is very effective in absorbing carbon dioxide into monoethanolamine solvent.

KEYWORDS

Carbon dioxide absorption, Mass transfer, Henry's law, Film theory, Monoethanolamine.

INTRODUCTION

India is amongst the top four Carbon dioxide (CO_2) producing countries with China, US, and countries under European Union, it produced about 2,341,000 kt of CO_2 in 2014 [1]. In India, 38% of total CO_2 emission is due to electricity production [2]. 71% of the total electricity supplied in India is generated using thermal power plant of which 62% is fulfilled using coal based thermal power plants [3]. This number is huge because India requires a large amount of electricity which is produced by power plants present across the country.

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The CO_2 emission from thermal power plants in India was estimated to be 4.98×10^8 metric tons in the year 2009-2010 [4]. India has about 116 coal based thermal power plants with near about 429 units.

The total power capacity from all these plants comes around 167,707.88 MW per year [5]. The impact of this huge CO_2 emission is rising of global temperature, i.e. global warming, which is posing great threat not only to the environment but to human life as well [6].

Therefore, there is a need to study post combustion treatment technologies for CO_2 removal. Among the existing technologies for CO_2 removal, absorption is one of the most suitable technology [7]. In this paper the authors have carried out detailed theoretical investigation on reactive absorption of CO_2 using Monoethanolamine (MEA) solution as the solvent. The method involved in this process is reactive absorption, i.e. the absorption of CO_2 from the gaseous stream into MEA accompanied by the reaction where CO_2 and MEA combine to form ester (R_1HCOO^-) and primary amine ($R_1NH_3^+$) which enhances the mass transfer between the two. In order to understand the state of art in CO_2 absorption, the literature survey has been carried out in presented in following section.

Literature studies

Several studies have been carried out over the years on absorption and reactive absorption of CO_2 using various solvents such as Methyldiethanolamine (MDEA), MEA, ammonia, Diethanolamine (DEA), blend of MDEA and ethanolamine. Zhang and Chau-Chyun [8] used electrolyte Nonrandom Two-Liquid (NRTL) activity coefficient model to develop a rigorous and thermodynamically consistent representation for the MDEA-water-CO_2 system. The model has been validated for predictions of Vapor-Liquid Equilibrium (VLE), heat capacity, and CO_2 heat of absorption of the MDEA-water-CO_2 system temperature range of 313 K to 393 K. Liu *et al.* [9] studied absorption of CO_2 absorption in ammonia. They concluded that ammonia could be better solvent for CO_2 absorption. However, they have mentioned further experimental investigations are necessary. Lawal *et al.* [10] presented a study on chemical absorption of CO_2 in MEA solution based dynamic modeling of the absorber and regenerated columns linked together. They concluded that the model predicts the absorber and regenerator temperature profiles and CO_2 profiles very well. Molina and Bouallou [11] investigated kinetics of CO_2 absorption into mixed solutions of MDEA and DEA. Their study primarily focused on to optimize the blend composition to capture CO_2.

In most of these studies, a single stage CO_2 absorption using thin film mass and heat transfer laws have been considered. These studied predicted the distance, from gas-liquid interface to the point at which CO_2 get absorbed in the solvent, using either equilibrium stage or rate based model.

A study done using gPROMS showed rate based model gives better prediction than the equilibrium model for CO_2 absorption using MEA. It also states that effect of *L/G* ratio is more sensitive than effect of flow rate change [12] which was verified in this study as well. Another study rate based model was validated using 0.1 MW pilot plant in South Korea which showed good agreement with data except when the *L/G* ratios taken were low [13]. Study of CO_2 absorption using aq. MEA using a packed tower has been done which were in agreement with industry scale pilot plant data [14].

Dynamic modeling of CO_2 absorption from coal-fired power plants into an aqueous monoethanolamine solution has been carried out using Aspen Tech software [15]. The authors have studied the transient simulation of absorption column and the results are validated with experimental data. A detailed a state-of-the-art review on post-combustion CO_2 capture with chemical absorption is presented by Wang *et al.* [16]. The authors have concluded that more efforts in future should be directed to reduce energy combustion in post combustion CO_2 capture with chemical absorption. Dynamic behavior of coal-fired power plants with post combustion CO_2 capture is studied by Wellner *et al.* [17]. The authors used a model-based control strategy based on existing directives for power plants with post combustion CO_2 capture. Aboudheir *et al.* [18], investigated the kinetics of the reaction between CO_2 and high CO_2-loaded, concentrated aqueous solutions of MEA for the temperature range from 293 to 333 K. They proposed a new termolecular-kinetics model, for CO_2-MEA solution, which proved to be better than previously published kinetic models. Zhang and Chen [19] have performed simulation studies with both the rigorous rate-based model and the traditional equilibrium stage model for CO_2 absorption with MEA. The model results were validated with the pilot plant data from recently published literature. They concluded that the rate-based model yields better predictions compared to equilibrium stage model. Plaza *et al.* [20], presented results of CO_2 absorption in MEA with a new model that uses a rigorous thermodynamic model from the published literatures.

From above mentioned literature survey it is noted that researchers across the globe are striving to find the unique model for prediction of the performance of absorption column for the CO_2 absorption. Authors have used rate based and equilibrium NRTL models and simulated them using various software like gPROMS and ASPEN plus. Most of papers don't provide the data for overall estimations of concentration profile of CO_2 (performance of the column) and its variation with *L/G* ratio and number of stages.

In view of above, this study tried to provide the theoretical investigation of the absorption tower to predict the performance of the single stage as well as the absorption column for CO_2 absorption. Further, it provides the stage concentration profiles of CO_2 at various temperatures, inlet flow rates and liquid to gas ratios. The liquid phase CO_2 concentration on a particular stage is estimated using interfacial mass transfer concepts and by applying Henry's law at the gas-liquid interface. Further the prediction of overall estimate of the CO_2 absorption has been done using unified method. The overall objective of this study is to predict and understand the dynamics of absorption column for the removal of CO_2. The outcome of the study can be used for improvements in the operations of the commercial absorption columns for CO_2 absorption as well as for the designing of new column for the same.

MODEL DEVELOPMENT

CO_2 containing outlet stream of desulphurization unit is the input to the absorption column. This stream consists of CO_2 mixed with compounds like Sulphur dioxide (SO_2) and Nitrogen oxides (NO_x). SO_2 also has a high tendency to react with aqueous MEA solution and interfere in the CO_2 absorption and therefore a desulphurization unit has to be installed before the absorption column to bring the concentration of SO_2 down enough so that it wouldn't have much effect on CO_2 absorption. A maximum of 10 ppmv of SO_2 concentration and less than 20 ppmv for NO_x concentration [18] is acceptable for the absorption column to work efficiently.

A column with multiple trays is considered in which the treated flue gas is moved counter currently with MEA solution. The CO_2 present in the flue gas reacts with the solvent in a pseudo first order kinetics where carbamate formation takes place [19]. This reaction enhances the mass transfer from gas phase to the liquid phase, thus the absorbed CO_2 is separated from the flue gas as it moves upwards in the absorption column. To find the amount of CO_2 which gets absorbed horizontally (on a stage) undergoing mass transfer forming a thin film between gas and liquid phase is considered under the following assumptions:

- The gas phase offers no resistance to mass transfer, the resistance offered is by the liquid film only;

- No vaporization of solvent, since the vapor pressure of MEA is very less at the temperature rage considered for this work;
- Henry's law is applicable at the interface of the gas and liquid phase;
- Temperature is assumed to be constant throughout the complete process;
- Gas liquid equilibrium is assumed at each stage.

N-stage equilibrium model is developed using material balance equation, equilibrium relations and summation equations as follows:

Material balance:

$$V_{i+1} + L_{i-1} - V_i - L_i + F_i = 0 \tag{1}$$

Equilibrium relation:

$$Y_i = K_i \times X_i \tag{2}$$

Summation equation:

$$\Sigma Y_i = 1 \text{ and } \Sigma X_i = 1 \tag{3}$$

Reaction involved

The reaction of CO_2 with MEA involves the following reactions [19] into ester and amine:

$$R_1NH_2 + CO_2 <\text{--}> H^+ + R_1HCOO^-$$

$$R_1NH_2 + H^+ <\text{--}> R_1NH^+{}_3$$

Overall reaction:

$$CO_2 + 2R_1NH_2 <\text{--}> R_1HCOO^- + R_1NH_3^+$$

Parameters used

Parameters involved in this model are diffusivity of CO_2 and Nitrogen dioxide (N_2O) in MEA and water, Henry's constant at gas-liquid interface, density of aqueous MEA, rate and equilibrium constant of the reactions. The relations for above said parameters are provided below.

Diffusivity equation. The diffusivity coefficient of CO_2 in MEA was estimated using the N_2O analogy developed by Ko [20]:

$$D_{\mathrm{CO_2.MEA}} = D_{\mathrm{N_2O.MEA}} \times (D_{\mathrm{CO_2}}/\, D_{\mathrm{N_2O}})_{\mathrm{water}} \tag{4}$$

The equations of the diffusivity used were given by Versteeg and Vanswaaij [21]:

$$D_{\mathrm{N_2O.MEA}} = b_0 e^{[-2,371/r]} \tag{5}$$

$$D_{\mathrm{CO_2.MEA}} = b_s e^{[-2,119/r]} \tag{6}$$

$$D_{\mathrm{N_2O.MEA}} = b_0 + b_1 C_{\mathrm{amine}} + b_2 C_{\mathrm{amine}} \times \ e^{[b_3 + b_4 C_{\mathrm{amine}/T}]} \tag{7}$$

The values of constants in diffusivityrelations are taken from Ko *et al.* [20], and are given in Table 1.

Table 1. Values of constants in diffusivity relations

$b_0 = 5.07 \times 10^{-6}$	$b_1 = 8.65 \times 10^{-7}$	$b_2 = 2.78 \times 10^{-7}$	$b_3 = -2{,}371$	$b_4 = -93.4$	$b_5 = 2.35 \times 10^{-6}$	$b_6 = -2{,}119$

Density relations. The density of aq. MEA solution was taken as 1,013 kg/m^3 using the relations from Weiland *et al.* [22].

Solubility relation (Henry's law). The Henry's law constant is estimated using N_2O analogy by Clarke [23]:

$$H_{\mathrm{CO_2.MEA}} = H_{\mathrm{N_2O.MEA}} \times (H_{\mathrm{CO_2}} \,/\, H_{\mathrm{N_2O}})_{\mathrm{water}}$$

$$H_{\mathrm{gas.H_2O}} = e^{\left(a_0 + \frac{a_1}{T} + a_2 \mathrm{In} T + a_3 T\right)} \tag{8}$$

These constants are function of temperature Penttila *et al.* [24], and are provided in Table 2.

Table 2. Values of constants for Henry's constant relations

Henry's constants	a_0	a_1	a_2	a_3
$H_{\mathrm{N_2O,H_2O}}$	158.24	−9,048.59	−20.86	−0.00252
$H_{\mathrm{CO_2,H_2O}}$	145.36	−8,172.35	−19.30	0
$H_{\mathrm{N_2O,MEA}}$	−9,172.5	39.59	-	-

Rate of reaction. There have been many discussions regarding the order of rate of reaction with respect to both CO_2 and MEA. In most of literature studies, the order of reaction with respect to CO_2 is assumed to be 1 but the order of reaction with respect to MEA was found to be in the range 1 to 2. However, in most of the studies reaction rate constant has been taken as 1st order with respect to MEA. Thus, the overall order was found to be 2 [25]. But in this study the authors have considered pure MEA with no vaporization in gas phase due to very less vapor pressure of MEA (40 pascal at 20 °C), hence in this study a pseudo first order reaction is considered. The rate constant was found using following relation given by Aboudheir *et al.* [26]:

$$k = 4.61 \times 10^9 \, \mathrm{e}^{(-4{,}412/r)} \tag{9}$$

Equilibrium constant was calculated with the help of following relation [27, 28]:

$$\mathrm{In}\, K = \mathrm{A} + \mathrm{B} \,/\, T + \mathrm{C} \times \mathrm{In}\, T + \mathrm{D} \times T \tag{10}$$

The values of these constants are given in Table 3.

Table 3. Values of constants for equilibrium constant

A	B	C	D
231.46	−12,092.1	−36.78	0

Operational details. A pure MEA solution is supplied from top of the absorption column counter currently to the gaseous stream with CO_2 as a major component. Mathematical model has been developed for this system. Simulation studies are carried out at two temperatures 298 K and 313 K, with CO_2 compositions of 0.6 and 0.8 to find

CO_2 concentration profile across the gas-liquid interface at a given stage. Simulation studies are further carried out to estimate overall absorption of CO_2 in absorption column, at different inlet flow rates of CO_2: 65.74, 75.74, 85.74 and 95.74 kmol/m^3, at different *L/G* ratios: 2, 6.5, 8.5 and at different number of stages 15, 25 and 30.

Development of model using unified method

In this method, *N* numbers of stages are considered for absorption of CO_2 using MEA as the solvent. Flue gas and MEA are flowing counter currently in the tower where the flue gas is fed from the bottom most tray (V_n), where as pure MEA is added from the top tray. At each tray, fractional amount of CO_2 gets absorbed into MEA and flue gas which is now CO_2 lean moves to the next upper plate. The process continues up to the N^{th} stage, post which the lean flue gas moves into the stack for removal. The schematic of i^{th} stage is shown in Figure 1.

The overall mass balance equation gives:

$$V_{i+1} + L_{i-1} - V_i - L_i + F_i = 0 \tag{11}$$

The component balance gives:

$$(Y_{I+1}V_{i+1}) + (X_{i-1}L_{i-1}) - (Y_iV_i + X_iL_i) + F_i = 0 \tag{12}$$

where V_i decreases by N_aAV_m amount at each stage because of CO_2 absorption and its reaction with monoethanolamine.

Equilibrium relation:

$$Y_i = K_i \times X_i \tag{13}$$

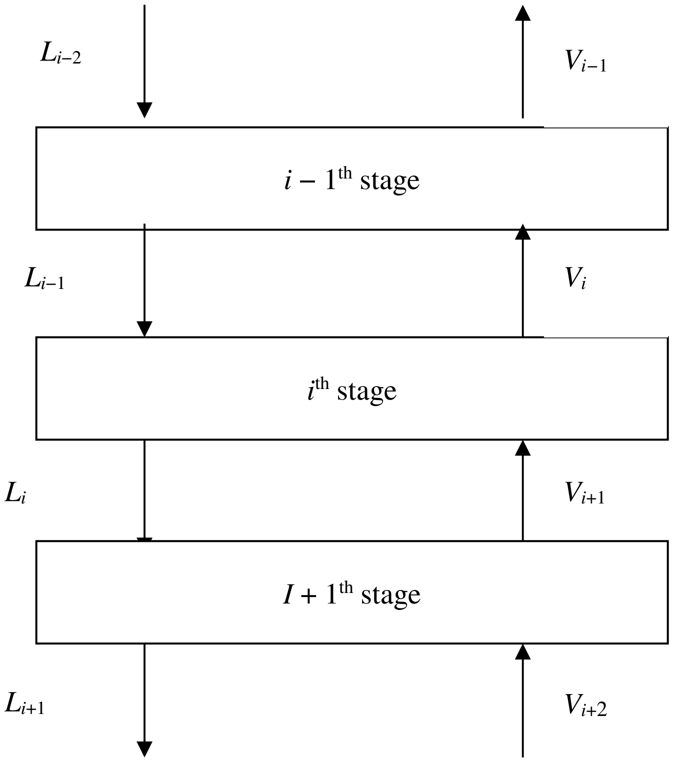

Figure 1. *N*-stage equilibrium model

The process depends on the stripping factor (S_i), (V_i K_i)/L_i, which makes the equation:

$$Y_{i+1,j}V_{i+1,j} + y_{i,j}V_{i,j}\,[(L_{i,j} / G_{i,j}K_{i,j}) - 1] + y_{i-1,j}V_{i-1,j}\,[(L_{i,j} / G_{i,j}K_{i,j})] + F_i = 0 \tag{14}$$

or:

$$Y_{i+1,j}V_{i+1,j} + y_{i,j}V_{i,j}\,[(1 / S_{i,j}K_{i,j}) - 1] + y_{i-1,j}V_{i-1,j}\,[(1 / S_{i,j}K_{i,j})] + F_i = 0 \tag{15}$$

where $S_{i,j}$ is the stripping factor, i.e:

$$S_{i,j} = (G_{i,j} \times K_{i,j}) / L_{i,j} \tag{16}$$

Solution procedure. The model equations are solved using the equilibrium matrix method (Thomas algorithm) to estimate overall absorption CO_2 in the tower. It is simplification of Gaussian elimination method which is used to solve system of equations, using forward and backward substitutions. For this purpose the MATLAB software has been used. The K value in the equilibrium relation is calculated using the Henry's constant at the interface of the two medium.

RESULTS AND DISCUSSION

The simulation studies for the absorption column were carried outto find the mole fraction of CO_2 absorbed at each stage. The overall absorption of CO_2 in absorption column is also estimated.

Parameters values

The parameters values used, at two temperatures 298 K and 318 K, in simulation studies are mentioned in Table 4. The parameters viz., rate constant, Henry's constant, diffusion coefficient of CO_2 and MEA are used for this study.

Table 4. Effect of variation of temperature on different parameters

Parameters	T = 298 K	T = 318 K
Rate constant [s^{-1}]	1.713211×10^3	4.34716×10^3
Henry's constant [kPa m^3/kmol]	2.162313×10^8	2.927188×10^8
Diffusion coefficient of CO_2 [m^2/s]	1.293194×10^{-9}	2.2309×10^{-9}
Diffusion coefficient of MEA [m^2/s]	7.280607×10^{-10}	1.15787×10^{-9}

Variation of rate constant with temperature. As the temperature increases the rate constant also increases which in turn enhances the reaction rate of CO_2 with MEA solution and thus increases absorption.

Variation of Henry's constant with temperature. The value of Henry's constant increases at the interface of gas and liquid with increase in temperature.

Variation of diffusion coefficient with temperature. Diffusion coefficients also increase with temperature and hence increase the diffusion of the acidic gas into the liquid solution.

Stage-wise variation in concentration profile

The effect of change in the inlet gas temperature and inlet CO_2 composition, on the concentration profile of CO_2 from the gas-liquid interphase to bulk MEA at a given stage is presented in Figure 2 and Figure 3, respectively. These studies were carried out at constant inlet CO_2 flow rate of 75.74 kmol/m^3 and L/G ratio of 8.5.

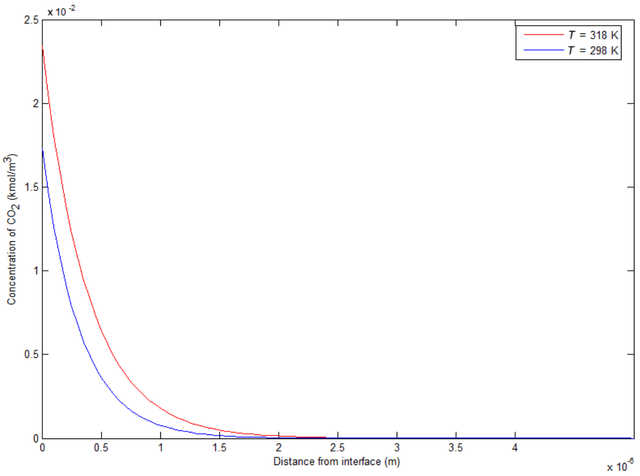

Figure 2. Effect of change in temperature on CO_2 concentration profile

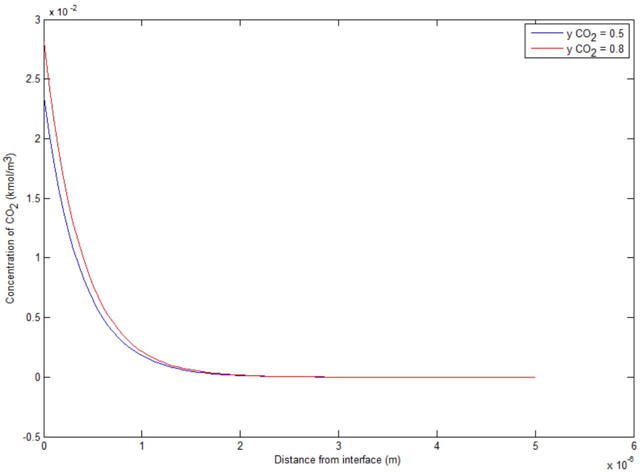

Figure 3. Effect of change in inlet CO_2 composition on CO_2 concentration profile

Effect of temperature on concentration profile ($y_{CO_2} = 0.8$). Figure 2 presents the effect of temperature on CO_2 profile from gas-liquid interphase to bulk liquid phase. From Figure 2 it is noted that 2.35×10^{-2} kmol/m^3 of CO_2 gets absorbed completely into the aqueous MEA solution at a distance of 2.1×10^{-6} m from gas-liquid interface at temperature 318 K. However, at 298 K, 1.76×10^{-2} kmol/m^3 of CO_2 gets absorbed completely into the aq. MEA solution at a distance of 1.5×10^{-6} m from the gas-liquid interface.

Thus, from above results it is noted that with increase in temperature the concentration of CO_2 increases at the interface which gets absorbed within film thickness. This is due to the fact that with increase in temperature, the solubility of the acidic gas increases in the solvent. Hence, using the thin film layer model, CO_2 concentration was found to decrease from interface concentration at equilibrium to a minimum of 0 kmol/m^3 up to a distance within ±2 micrometers.

Effect of inlet CO_2 composition on concentration profile ($T = 318$ K). The Figure 3 shows effect of change in inlet CO_2 composition on CO_2 profile from gas-liquid interphase to bulk liquid phase. From Figure 3 it is observed that as the inlet CO_2 composition in the flue gas is increased from 0.5 to 0.8 the interface concentration of CO_2 also increased from 2.35×10^{-2} kmol/m^3 to 2.75×10^{-2} kmol/m^3 at that particular stage. However, there is no change in the distance from gas-liquid interface for CO_2 concentration to reduce to zero.

Overall estimate

The overall estimate of CO_2 absorption in absorption column has been carried out using unified method. Here at each stage, it is considered that a constant amount ($N_a \times A \times V_m$) of CO_2 is getting absorbed due to the reaction of CO_2 with MEA solvent. Where N_a is the molar flow rate, A is area and V_m is the molar volume at that temperature. The film area is taken as 2.5 m × 0.4 m and molar volume is taken as 24.86 m^3/kmol using gas law at 318 K. The major component of inlet flue gas is considered to be CO_2 as the outlet from the desulphurization unit is rich in CO_2 concentration because most of the SO_2 content is removed in the desulfurization unit.

The overall percentage of CO_2 absorption is calculated by subtracting outlet mole fraction at stage 1 at the top from inlet mole fraction at the bottom and dividing it by inlet mole fraction.

Effect of change in *L/G* on overall CO_2 absorption. Figure 4 present the effect of change in *L/G* on overall CO_2 absorption. The simulation studies are carried out at inlet CO_2 composition of varying in the range from 0.85 to 0.95, inlet CO_2 flow rate of 75.74 kmol/m^3, temperature of 318 K and with 30 numbers of theoretical stages. The details of input/output streams composition along with *L/G* ratio is provided in Table 5. From the figure it is observed that overall CO_2 absorption increases from 66.73% to 95.60% with increase in *L/G* ratio from 2 to 8.5, respectively. This must be attributed to fact that the increase in solvent rate enhances the absorption rate. It is also seen from the Figure 4 that for *L/G* ratio 8.5, CO_2 profile across the column is almost flat as compared to CO_2 profile at *L/G* ratio 2 and 4.5.

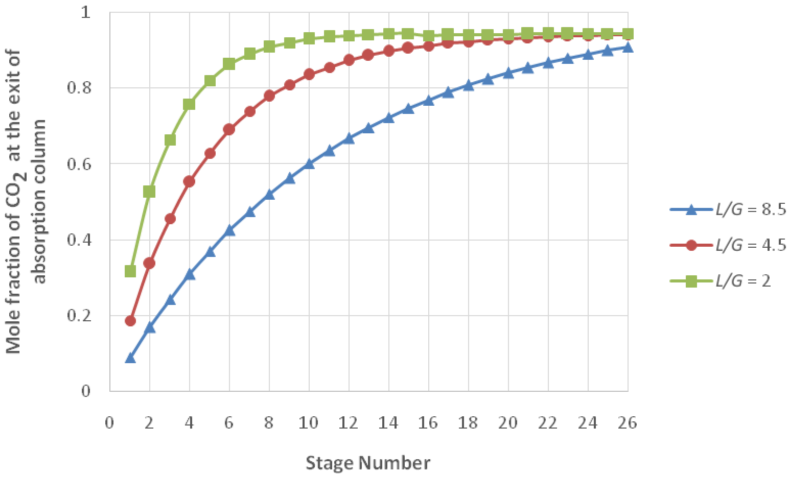

Figure 4. Effect of change in *L/G* on overall CO_2 absorption

Table 5. Input-output streams data

Sr No.	*L/G* ratio	Inlet CO_2 composition	Outlet CO_2 composition
1	2	0.95	0.31
2	4.5	0.95	0.185
3	8.5	0.92	0.039

Effect of change in no of stages on overall CO_2 absorption. The effect of change in no of stages on overall CO_2 absorption is presented in Figure 5. The simulation studies are carried out at constant inlet CO_2 composition of 0.8, inlet CO_2 flow rate of 75.74 kmol/m^3, temperature of 318 K and with *L/G* ratio of 8.5. It can be noted from Figure 5 that overall CO_2 absorption increases as the number of stages are increased from 15 to 30.

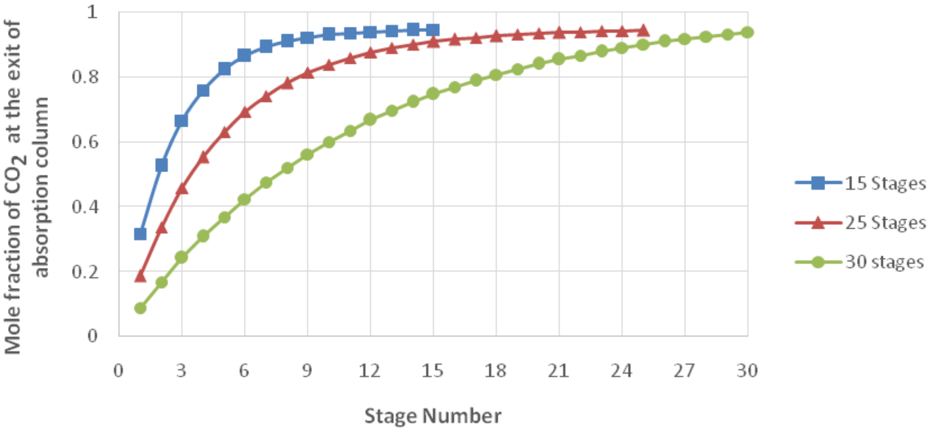

Figure 5. Effect of change in no of stages on overall CO_2 absorption

Effect of change in inlet CO_2 flow rates on overall CO_2 absorption. Figure 6 presents effect of change in inlet CO_2 flow rate on overall CO_2 absorption. The simulation studies are carried out at constant inlet CO_2 composition of 0.8, temperature of 318 K, number of stages as 30 and with *L/G* ratio of 8.5. It can be seen from the figure that as the flow rate increases the absorption rate also increases but the change is very gradual. The absorption is the minimum for 65.74 kmol/hr and maximum for 95.74 kmol/hr. This is because as the inlet CO_2 flow rate increases, the interface CO_2 concentration increases and hence absorption of CO_2 into MEA increases.

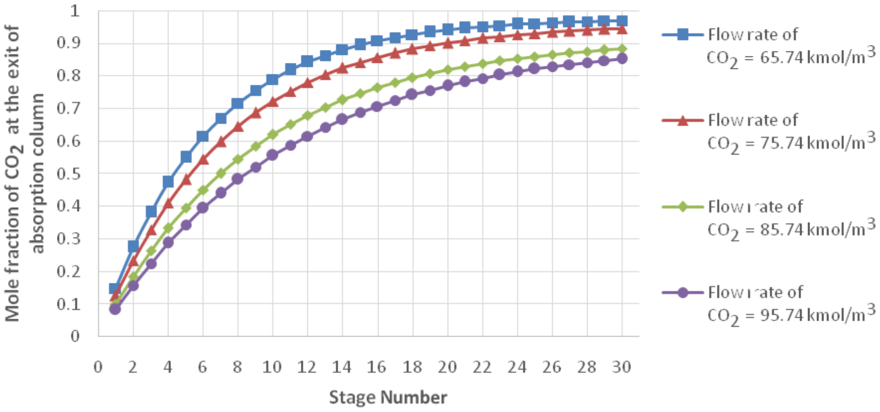

Figure 6. Effect of change in inlet CO_2 flow rates on overall CO_2 absorption

CONCLUSIONS

The expansion of economy and population with current technology leads to higher rates of urbanization, industrialization and deforestation, all factors contributing to an increase in CO_2 level in the surrounding. So, steps need to be taken in order to reduce this level wherein the first step that has to be taken would be to trap the industrial CO_2 emissions in the most efficient and economical way. This can be achieved by post combustion capture of CO_2 using absorption columns.

The film model gives an overview as to how the CO_2 absorption takes place for a particular tray and the thickness up to which entireamount of CO_2 gets absorbed. From the stage wise CO_2 concentration profile it is noted that the absorption of CO_2 into MEA is very instantaneous. This may be attributed to the fact that reactive absorption is very effective in CO_2 absorption. This study also gives us visualization as to how absorption takes place at each tray throughout the column. This study has revealed the important design variables which controls the overall absorption of CO_2.

The unified model approach is best suited for overall estimation of CO_2 absorption. From the results it is noted that, 95.60% of the inlet CO_2 can be absorbed in absorption column with 30 trays, *L/G* ratio of 8.5 and inlet CO_2 flow rate of 95.74 kmol/m^3. From the results it is also noted that CO_2 absorption in the tower is more sensitive to *L/G* ratio as compared to composition or flow rate changes which is in accordance with literature studies.

NOMENCLATURE

A	film area	[m^2]
D_{CO_2}	diffusion coefficient of CO_2	[m^2/s]
D_{MEA}	diffusion coefficient of monoethanolamine	[m^2/s]
F_i	feed stage	[-]
H_{CO_2}	Henry's constant	[kPa m^3/kmol]
k	rate constant	[sec^{-1}]
K	equilibrium constant	[-]
L	liquid flow rate	[m^3/sec]
N_a	mass flux	[kmol/m^2s]
$S_{i,j}$	stripping factor	[-]
V	vapor flow rate	[m^3/sec]
V_m	molar volume	[m^3/kmol]
X	liquid mole fraction	[-]
Y	vapor mole fraction	[-]

Subscripts

i	stage
j	component

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