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Investigation of overall mass transfer coefficient of CO_2 absorption in packed Column

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ABSTRACT

Mixtures of different types of amine solution Monoethanolamine, Diethanolamine, and Triethanolamine were experimentally used to investigate the overall mass transfer coefficient (K_{Ga}) at different operating parameters. The experiments were made in a packed bed reactor (PBR) with 75 cm in high and10 cm inside diameter as a gas-liquid contactor at 25℃ and atmospheric pressure, using a simulation gaseous mixture (air, carbon dioxide) with recycle stream (semi-continuous process). Experimental design process Taguchi was employed. Four factors and three levels were chosen and exploded using L9 (3 ^4) orthogonal array design. These parameters for semi-continuous process were namely: gas flow rate 5,10,and 15 L/min ,airflow rate80,90,and 100 L/h, liquid flow rate 400,450,and 500 mL/min and time absorption time 30,45,and 60min . A Shimadzu GC-8A Gas Chromatograph with a thermal conductivity detector was used to measure the CO_2 concentration absorbed in aqueous blended solution. The maximum value for CO_2 loading was 8.622 (mol CO_2/mol amine) at 15 L/min gas flow and 450 mL/min liquid flow and 100 L/h airflow for 60 min from absorption time. The results showed that the max value of K_{Ga} is 0.048 S^{-1}.

1. Introduction

Elevation in the emissions regarding atmosphere's anthropogenic CO_2 is considered to be challenging due to the fact that it is mainly contributing to the global warming. Fossil fueled plants are the main emission source, particularly plants using coal as the main fuel [1]. CO_2 is one of the gasses that are naturally-occurring, also it has main impact in reflection related to the solar radiation back to Earth, and that will keep the surface temperature of the planet at suitable levels for the life [2]. Throughout the past tens of years, there has been an increase in the emissions related to other greenhouse gases (GHGs), in addition to CO_2, such as CH_4, N_2O, HFC, perfluorocarbons as well asSF6 [2]. Worldwide, CO_2 which is emitted from the power plants is considered to be about 40% of the overall emissions of CO_2 and within the ongoing businesses and industries, it is anticipated to be elevating to about 60% prior to the end of this century [3, 4]. For the

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Nomenclature

CO_2	Carbon dioxide
K_{Ga}	Overall mass transfer Coefficient
K_g	Gas film physical mass transfer Coefficient
K_L	Liquid mass transfer coefficient
$n_{(CO2,abs)}$	Number of moles of CO2 absorption
PBR	Packed Bed Reactor

Greek symbols

α	CO_2 loading
Δ	Film thickness

purpose of preventing excessive CO_2release in atmosphere, carbon capture and storage (CSS) is of high importance in the industry of fossil fuel. Such an approach includes CO_2 capturing, compress it to transport, and after that store it permanently (in gas fields and depleted oil). The major application regarding CSS is in the industrial point sources as fossil-fuel power plants, production of fossil fuel, facilities for hydrogen production, industrial plants (steel and iron blast furnaces, chemical processes, and cement kilns) [5].

The absorption is the most important approach to remove CO_2 from the industrial waste gases as well as for synthesizing and for purification of natural gas. Such a process passes flue gas via liquid which have the ability to absorb CO_2 (in absorber vessel) and after that releasing CO_2 at increased temperatures in regenerator vessel (stripper) [6, 7]. Aqueous alkanolamines are the major chemical solvent for capturing acid gaseous. Alkanolamines have been generally applied as absorbents for the capturing of CO_2, alkanolamine's structures involve primary, secondary, ternary amines including no less than single (OH) as well as amine group. In the case of chemical solvents, primary amines such as MEA, secondary amines DEA, amino-ethoxyethanol (DGA), and diisopropanolamine (DPA), tertiary amines such as TEA and MDEA, as well as alternatives to amines such as hot potassium carbonate, are used. Such alkanolamines were applied as chemical absorbents for removing acidic gases like (H_2S, CO_2). A lot of the works on the chemical absorption with the tertiary alkanolamines like DEA, TEA, in addition to their associated apply of mixed amine absorbents, particularly blends the primary and tertiary amines (like TEA and MEA) or the secondary, as well as tertiary amines (like TEA and DEA), has increased or them together [8].

The amine blending process has many benefits, the most notable of which is:

• Dynamic efficiency improvement.

• Reducing problems related to the decomposition and operation of solvents resulting from corrosion.

• Flexibility in the range of amines available to tailor and optimize the composition of the solvent to achieve the highest absorption efficiency.

• Maintain a high absorption rate in single amino solvents in a mixture of individual ingredients.

• Reduced energy requirements for solvent regeneration [9].

The main benefits related to the technology of chemical absorption are as follows:

1. Chemical absorption can be considered as a major developed approach to capture CO_2, operating at normal pressure and temperature.

2. For tens of years, it was commercialized, though not for the capture of CO_2 from power plants. 3. It can be used efficiently to dilute CO_2 system (general flue gas from power plants) [10].

Also, there are certain disadvantages related to this approach, as follows:

1. Amine oxidative degradation through SO_2, NO_2, and O_2 in flue gases that induce a high absorbent makeup rate.

2. The energy consumption is high throughout high temperature absorbent regeneration.

3. The equipment corrosion rate is high.

4. The equipment size is large.

It was initiated primarily for:

1. Investigating the capacity of (DEA) as a blend for MEA and TEA solution in CO_2 scrubbing via a semi-continuous process.

2. Evaluating the effect of various parameters on the process using Taguchi analysis.

2. material and methods

MEA, DEA, and TEA were used as solvents. A CO_2 cylinder (pure 99.99 percent) was used as the inlet gas source. During every cycle of the experiment distilled water has been used. **Table 1.** Shows the chemical material used in this work.

Table 1. Chemical material (Amine) use in academic work

Chemical Name	Abbreviation	Chemical formula	Molecular weight [g/mol]	Density g/cm^3
Monoethanolamine	MEA	C_2H_7NO	61.084	1.0117
Diethanolamine	DEA	$C_4H_{11}NO_2$	105.137	1.097
Tiethanolamine	TEA	$C_6H_{15}NO_3$	149.190	1.124

2.1. Mass Transfer for Gas Absorption

Mass transfer can be considered as one of the common phenomena which happen in simple, daily life in addition to the engineering process. In the case of absorption, the absorption of the gas in the liquid happens in the case of the transfer of the gaseous components from gas- to liquid-phase. In the process of the chemical absorption, the gaseous component undergoes the absorption by liquid phase through combining the reaction and the diffusion mechanism. The processes of the chemical absorption are like systems that are aqueous amine-based, systems that are based on the ionic liquids, and ammonia manufacturing systems. As an explanation, the entire process rate can be viewed a layer of gas–liquid contact. The mass transfer must be enhanced with the increase of the turbulence in each of the liquid as well as the gas phases. The mass transfer happens through combining the diffusional and the mechanism of the chemical reaction in the boundary layer so the entire rate may be represented with each of the mass transfer and the chemical reaction. Numerous hypotheses may define the process of the gas absorption, like the penetration theory, film theory, boundary layer theory, and the surface renewal theory. The film model which is simpler mathematically, compared to the models of penetration is usually utilized, based on the film theory, a stagnant film of thickness δ_L is supposed to be existing at gas and liquid interface as has been illustrated in **Fig. 1.**The mass transfer through the molecular penetration takes place via a thin liquid gas layer which is δ thick, and there isn't any concentration gradient in liquid bulk. In the theory of the film, the coefficient of the mass transfer k_L and kg are proportionate to the coefficient of the diffusion DCO_2-amine and inversely proportionate to the thickness of the film. k_L stands for the mass transfer coefficient for the liquid side and is equal to: [11]

$$\delta_L = \frac{DCO_2 - amine}{k_L} \quad (1)$$

Similarly, the equation below may be resulted for the coefficient of the mass transfer in the gas phase:

$$\delta_G = \frac{DCO_2 - amine}{kg} \quad (2)$$

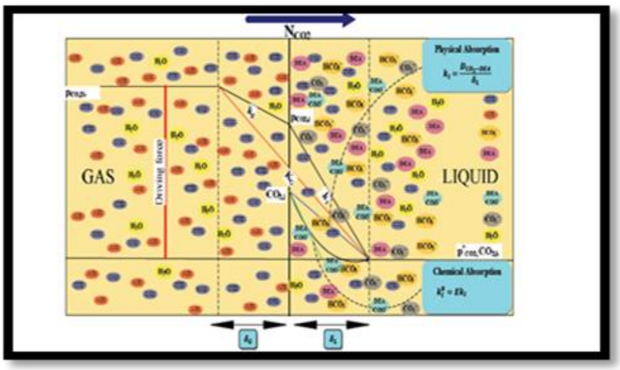

Figure 1. Carbon dioxide mass transfer into the liquid phase with a chemical reaction based on the film theory [11]

2.2. Determining the Overall Mass Transfer Coefficient (K_{Ga})

K_{Ga} is an important part of this research. Mass transfer occurs when a component. In the scrubber, a simulated mix of gas which contains A (CO_2) and B (air) which flow toa packed column from bottom continuously contacts with the solution of the amine which flows from the top in a column. At the same time, all current touches the opposite current column. In the present study, has been a reasonable theory. Taking under consideration Ideal gas regulations for inlet and outlet gases at different temperatures values, F_1/F_2 may be substituted by (P_1/P_2) $(T_2/T_1)(y_1/y_2)$. Which is why, the coefficients of the general volumetric mass transfer becomes :

$$K_{Ga}\,(s^{-1}) = \frac{Q_g\,(\frac{L}{s})}{V_L\,(L)}\, ln \frac{F_1(\frac{mol}{s})}{F_2(\frac{mol}{s})} \quad (3)$$

Where:

Qg=Gas flow rate, (L/S)

F_1= CO_2 molar flux at inlet, [mol/S]

F_2= CO_2 molar flux at outlet, [mol/S]

V_L = is the absorber volume of the liquid.

2.3. Taguchi method of analysis

A packed bed reactor was used to study the capture. Mixed amine concentration was 5V/v% MEA+10 V/v% DEA +5V/v% TEA. We used the Taguchi approach as an experimental design to reduce the multitude of experiments [12],[13]. This study utilized three levels for the variables.
When using the Taguchi method, it is important to determine the minimum number of experiments based on the following equation:

$$N = 1 + \sum_{i=1}^{NV} (Li - 1) \quad (4)$$

The main factors affecting the rate of absorption in the essential series are (Time of Absorption) > (Gas Flow rate) > (Air Flow Rate) > (Liquid Flow Rate), meaning the time of absorption has the most important influence on the absorption rate followed by the rate of gas flow. **Table 2.** shows the response of the removal efficiency factors to the S / N ratio, a maximum values from K_{Ga}, when gas flow was 15 L/min, and airflow 100 L/h and liquid flow 500 ml/min.

Table 2. S/N ratio for removal efficiency

Level	gas(CO_2) flow L/min	Air flow L/h	Liquid flow ml/min	Time min
1	-2.865	-2.870	-2.868	-2.812
2	-2.868	-2.868	-2.868	-2.880
3	-2.871	-2.867	-2.867	-2.912
Delta	0.006	0.003	0.001	0.100
Rank	2	3	4	1

3. Experimental set-up

A Plexiglas column 0.75 cm of height and internal diameter of10cm [14] Taken as scrubbing column as shown in **Fig.2**

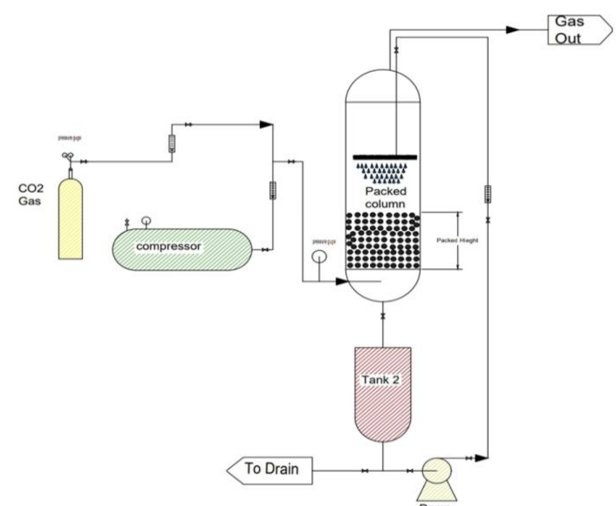

Figure 2. Experimental set-up for CO_2 capture using aqueous solution

A metal frame is provided to the base. The gas inlet is located at the bottom of the column through which the gas travels to the top of the stream. We used a perforated spray at the gas inlet to help distribute gas in the column, by positioning the liquid inlet at the top of the column. Where aluminum balls with a length of 10 mm and a diameter of 10 mm are used as random packing within the column, one of the most important advantages of packing is to enhance the surface area to transfer the mass and increase the contact time between gas and liquid. A drain valve is positioned at the bottom of the column to clear the column after the procedure and is inherently corrosive to collect the sample stream Monethanolamine (MEA), hence the use of a PVC container for feeding all tubes and fittings used are made of polyvinyl amine. The amino (MEA, DEA, TEA) solution was pumped from the top of the column. A side current is arranged from the pump to minimize the load on it, using the gas cylinder as the source of CO_2 The recycling stream is arranged at the bottom of the column leading to the feeding tank. For gas, the rotameter scale was 1–25 $Lmin^{-1}$, for liquid 100–500 $mLmin^{-1}$ and 10–100 Lh^{-1} for air. CO_2 samples were obtained in the flask pipe. During each operation, the setup was cleaned with water to eliminate any pollutants and carbamates that were created during the column reaction.

4. Results and discussion

The experiments for mass transfer in a packed bed were studied based on CO_2 absorption into blended solutions from MEA-DEA-TEA. Sample taken after absorption experiments were analyzed for $CO_2$and amine content. In order to determine the CO_2loading in terms of (moles of CO_2/ moles of amine). Gas chromatography (GC) was used to analyze the concentration of CO_2. The overall mole of CO_2 absorbed in the absorbent can be calculated by subtracting mol of CO_2 inlet from the outlet and the CO_2 loading can be calculated through

$$n_{co_2,abs} = n_{CO_2,IN} - n_{CO_2,out} \quad (5)$$

$$\alpha = \frac{mol\ of\ CO_2}{mol\ of\ amine} \quad (6)$$

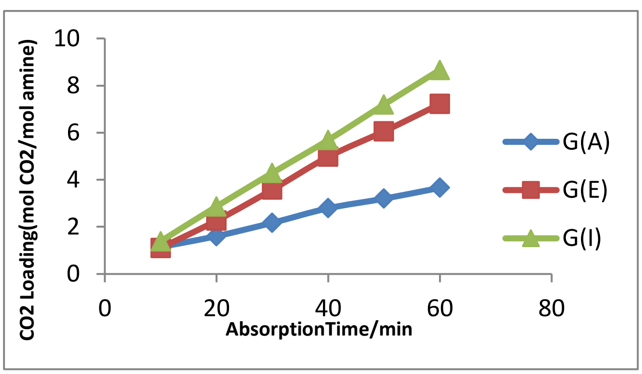

Figure 3. Maximum CO_2 absorption capacity Vs. absorption time for G(A, E, I)

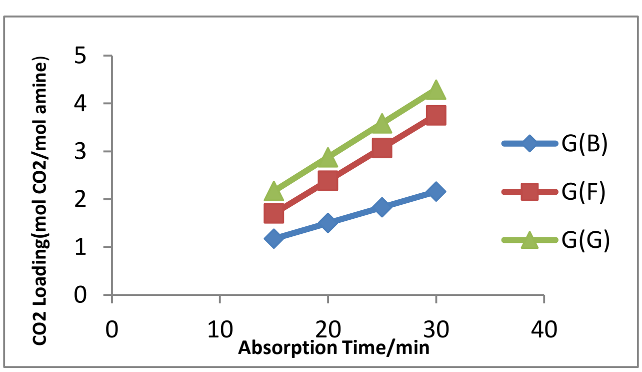

Figure 4 Maximum CO_2 absorption capacity Vs. absorption time for G(B,F,G)

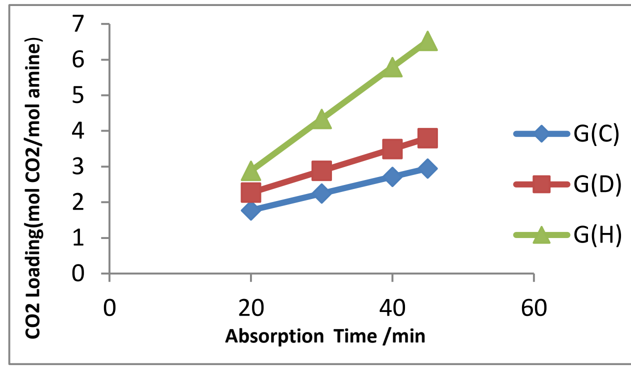

Figure 5. Maximum CO_2 absorption capacity Vs. absorption time for G(C,D,H)

When comparison of the results obtained for CO_2 loading capacity Vs. reaction time for the tested aqueous amine solution for groups (A,E,I), (B,F,G) and((C,D,H) as shown in **Figs. 3, 4 and 5** respectively. We found that increase in the CO_2 loading of an absorbent with increasing time and mean that the solubility of carbon dioxide is increased under various parameters condition.

4.1. Effect of CO_2 Loading on Overall Mass Transfer Coefficient K_{Ga}

The following figures show the main effect of CO_2loading on overall mass transfer coefficient K_{Ga} in each group. According to equation (3) calculate the overall mass transfer coefficient K_{Ga} S^{-1} (for semi-continuous process). In **Figs 7, 8, and 9** it is obvious that increasing the CO_2 load in amine solutions contributes to a reduction of the current active amine concentration, which subsequently decreases the overall mass transfer coefficient. This effect is mainly attributed to The CO_2 load in the amine solution is high. Where (MEA+DEA+TEA) blended amine solution are used in all three figures, mass transfer driving force from gas to liquid phase will be reduced. This effect is consistent with the work of each of the researchers.

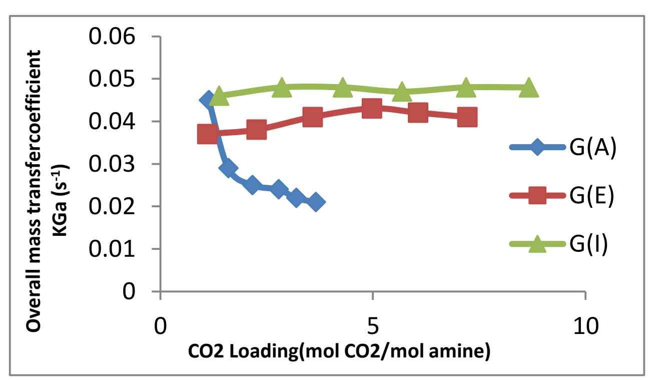

Figure 6 Progression of K_{Ga} in a function of CO_2 loading for semi-continuous processes (5% MEA+ 10%DEA+5%TEA) G(A,E,I)

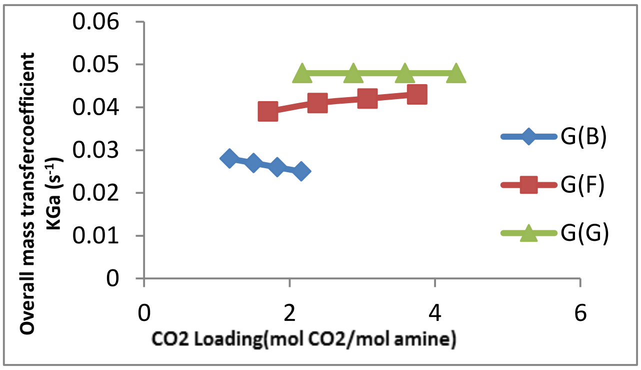

Figure 7. Progression of K_{Ga} in a function of CO_2 loading for semi-continuous processes (5% MEA+ 10%DEA+5%TEA) G(B,F,G)

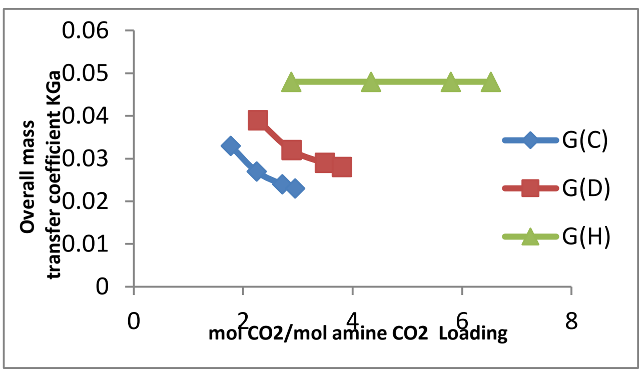

Figure 8. Progression of K_{Ga} in a function of CO_2 loading for semi-continuous processes (5% MEA+ 10%DEA+5%TEA) G(C,D,H)

The maximum value obtained from these experiments was 0.048 S^{-1} in group G,H,I at gas flow 15 L/min and liquid flow rate 400,450,500 mL/min and air flow rate 80,90,100 L/h. While Chen et al.,2015, they were found that K_{Ga} was 0.0342 S^{-1} when absorption CO_2 using MEA solution. Chen et al.,2018, absorption of CO_2 using aqueous ammonia solution K_{Ga} was 0.051 S^{-1}.

5. Conclusion

A packed bed scrubber under semi-continuous operation was successfully used for screening blended amines with the aid of the gas chromatography (GC). The overall mass transfer coefficient (K_{Ga}) of carbon dioxide absorption into blended MEA-DEA-TEA solutions was experimentally measured using a laboratory-scale packed bed. The parameter used in this study were gas flow, liquid flow, air flow and absorption time. This study showed that the increasing by the gas flow rate, the liquid flow rate, the K_{Ga} . This study showed that the increasing by the gas flow rate, the liquid flow rate, the K_{Ga} values increased. Also, increasing the CO_2 loading of amines lead to a K_{Ga} decrease.

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