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An Experimental Study of Mass Transfer coefficient of CO_2 absorption using Different Amines in a Packed Column

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Abstract

Industrial flue gas trapping carbon dioxide has become a critical environmental issue. The warming atmosphere caused by higher rates of GHG greenhouse gases poses a serious danger. The importance of this work in eliminating or reducing carbon dioxide emissions is underlined. In this work, an absorption technology was used actually to investigation the mass transfer coefficient of carbon dioxide from a gaseous mixture (air, carbon dioxide) in blended solution Monoethanolamine (MEA), Diethanolamine (DEA) and Triethanolamine (TEA) in a packed column reactor (PCR). The packed column was made of Plexiglas with 75 cm high and 10 cm inside diameter. The overall mass transfer coefficient (K_{Ga}) was evaluated at different operating conditions, gas flow rate, air Flow rate ,liquid flow rate .Where the gas flow rates were 5, 10 and 15 L /min , air flow rate 80,90 and 100 L/h , liquid flow rate 400 ,450,500 mL /min. The experimental work was conducted by using semi-continuous process with helping centrifugal pump. High-performance gas chromatographic (GC) was performed to evaluate CO_2 loading during absorption experiment. The experimental results showed that loading CO_2 in the range of 0.461–9.541 (mole CO_2 /mole amine), and the maximum value of overall mass transfer coefficient (K_{Ga}) was 0.0354 S^{-1} . It results in an increase in the CO_2 concentration consumed from the air and we are therefore trying to reduce the CO_2 concentration in the atmosphere.

Keywords : CO_2 Absorption , Mono ethanolamine (MEA), Di ethanolamine (DEA) , Tri ethanolamine (TEA), overall mass transfer coefficient (KGa) , Packed column reactor (PCR) , Taguchi method.

1. Introduction

Globally, trapping carbon dioxide from industrial flue gas has become a crucial environmental problem, according to the International Energy Agency (2012) the warming atmosphere resulting from higher rates of greenhouse gases poses a serious danger, energy-related carbon dioxide emissions will reach about 45 giga tons by 2035 in the light of current mitigation strategies and conditions, and work is becoming urgent and cost-effective carbon dioxide capture technologies are being demonstrated [1]There are three choices for reducing carbon dioxide emissions, reducing energy intensity, reducing pollution intensity and rising insulation with carbon dioxide. The first option requires energy efficiency, while the second option requires the use of non-fossil fuels, such as renewable energy and hydrogen. The third option comprises the development of carbon dioxide capture and capture techniques [2]. increasing in the carbon dioxide emission in to the atmosphere have already triggered a series of undesirable consequence such as such as global warming and climate change, which are considered to have close relationships with several environmental issues including rising sea levels, and glacier smelting, etc**[3]** Globally, the average CO_2 emission from power plants is about 40% of the total CO_2 emissions and is projected to rise by up to 60% in business as normal by the end of this century. [4] So, carbon dioxide separation technology is enjoying an increasingly larger market because of the urgent demand to response to both global warming and energy consumption . Meanwhile the most widely

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applied and effective methods for CO_2 capture is chemical absorption using novel absorbents that possess more efficient capture performance and are more energy-saving such as mixed amine solution [5]**.** In previous study 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, per fluorocarbons as well as SF_6[3]. For the purpose of preventing excessive CO_2 release in atmosphere, carbon capture and storage (CSS) is of high importance in the industry of fossil fuel .Such approach includes CO_2capturing, 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)[6]Mixed alkanoamine solvents, primary: mono ethanolamine (MEA), secondary: di ethanolamine (DEA) and tertiary: tri ethanolamine (TEA) were used to carry out this work. MEA is one of the predominant solvents because of its advantages, such as relatively low material cost, high boiling point power, low viscosity, reusability, quick absorption rate and rich industrial experience, total water solubility and solvency with other amines [7]**,**[8]**,**[9]. That employs TEA aqueous solutions in a hybrid membrane contactor by chemical reaction in one of the contactor chambers, di ethanolamine is the most appropriate secondary amine used both in the laboratory and in industry, thus reducing the regeneration energy required to reuse amine solutions [10]**,**[11]**.** The findings indicate major changes in CO_2 capture with respect to packed columns that function at room temperature.

In this paper was used a packed column reactor (PCR), with the liquid phase working in recycling mode. Because of the non-complex nature of this packed, low investment and current cost, high interfacial gas-liquid area and high absorption efficiency, which was combined with the two forms of possible amines (MEA and DEA) to test the best blended amine combination with acceptable conditions.

This study focused on improving amine solvents to achieve high CO_2 capture efficiency and low operating cost, as well as the overall mass transfer coefficients (K_{Gav}) determination, performance of the CO_2 absorption process.

2. Experimental Work and Data Analysis

A semi-continuous packed column is a powerful removal of carbon dioxide process compared to other scrubbers because it has a simple construction, higher heat and mass transfer coefficients, higher removal efficiency and efficient control of liquid residence time .So, a semi-continuous packed column reactor has become the absorber adopted by some researchers. The chemical solvent employed that used are shown in Table (1)

Table (1) Chemical materials (Amines) used in this work

Chemical Name	Abbreviation	Chemical formula	Molecular weight [g/mole]	Density g/cm^3
Monoethanol amine	MEA	C_2H_7NO	61.084	1.0117
Diethanol amine	DEA	$C_4H_{11}NO_2$	105.137	1.097
Triethanol amine	TEA	$C_6H_{15}NO_3$	149.190	1.124

Aqueous solutions were prepared with de-ionized water. The amine concentration in the mixtures was 5% volume percent for MEA, 5% volume percent for DEA, and 10% volume percent for TEA. All the experiments were performed at atmospheric pressure and at temperature equal to 298.15 K. The experimental set-up apparatus used for carbon dioxide absorption is shown in **Figure (1)**. For this experiment a packed column reactor (PCR) with the liquid phase working in recycling mode was used. The reactor is made of plexiglas with inner diameter (10) cm and the total high of the column was (75) cm.

Before introducing the solution of blended amine into packed column reactor, pure CO_2 and pure air from (compressor) were mixed where the air flow rate was regulated to an appropriate value in the range 80-100 L/h. The gas flow rate during carbon dioxide injection was used to maintain a certain flow rate, the flow rate of the feed gas (CO_2) supplied from the gas cylinder was balanced and regulated by gas flow (CO_2) rate in the range 5-15 L/min, and then it was fed through the down of the packed 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. The pressure gauges were used to show the gas and liquid inlet and outlet pressures. Before and after each test, to eliminate impurities in the reactor, de-ionized water was used for ten minute. All amine solutions were prepared using de-ionized water and volumetric glass ware, they prepared aqueous solutions using 1L of water for each experiment. The prepared absorbent liquid (solvent) was introduced at the upper section of

absorption column with the help of a liquid flow rate which was regulated to an appropriate via 400,450,500 mL/min, In order to control the liquid flow rate, a needle valve was mounted at the top of the column to create counter-current contact between gas and liquid, recycling of the exit liquid at the bottom using a Shimge centrifugal pump. Sampling of the circulating liquid and was taken from the packed column reactor at different times10 ml flask bottle. After several samples taken, the procedure of analysis was applied**.**

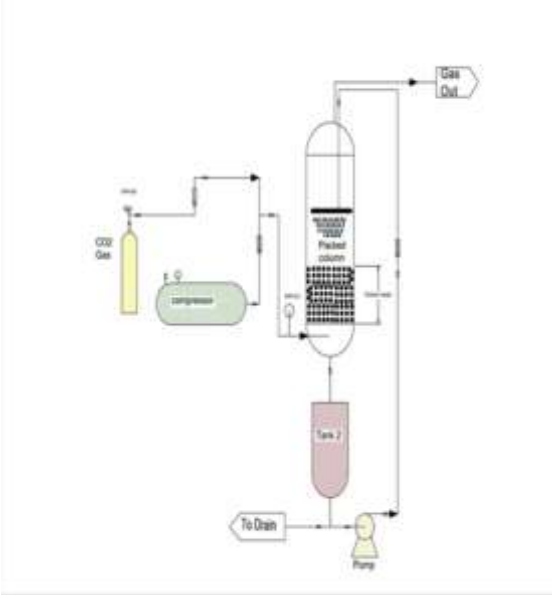

Figure (1) A schematic diagram of gas absorption experimental setup

2.1. Experimental Design

The Taguchi method was used applied as an experimental design, in order to reduce the large amount of experiments. The first step was the selection of independent variables .In this study the process parameters used in here are four parameters for semi-continuous, namely: gas flow rate ,air flow rate, liquid flow rate and time of absorption. Each factor has three levels as shown in **Tables (2).** Using the Taguchi experimental design, the orthogonal array L9(3^4) showed nine experiments, thereby reducing the number of experiments needed and the research cost by over 80%. **Table (3)** presents the combination of experiments in the orthogonal array for semi-continuous process.

Table (2) Factors and levels used in this study

Factor	Gas Flow $Lmin^{-1}$	Air Flow Lh^{-1}	Liquid Flow $mLmin^{-1}$	Amine concentration Volume percent (%)	Absorption time Min
1	5	80-100	400-500	5%MEA+5%DEA+10%TEA	30-90
2	10	80-100	400-500	5%MEA+5%DEA+10%TEA	30-90
3	15	80-100	400-500	5%MEA+5%DEA+10%TEA	30-90

Table (3) Orthogonal arrays for experimental design for semi-continuous process

Experiment No.	(gas flow) $Lmin^{-1}$	(air flow) Lh^{-1}	(Liquid flow) $mLmin^{-1}$	(Time) Min
1	5	80	400	60
2	5	90	450	30
3	5	100	500	90
4	10	80	450	90
5	10	90	500	60
6	10	100	400	30
7	15	80	500	30
8	15	90	400	90
9	15	100	450	60

2.2. Determination overall mass transfer coefficient

Mass transfer coefficient can be calculated from the equation below [12]:

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

Where:

Q_g= Gas flow rate, (L/S)

$F_1 = CO_2$ molar flux at inlet, $[\frac{mol}{s}]$

$F_2 = CO_2$ molar flux at outlet, $[\frac{mol}{s}]$

V_L = is the absorber volume of the liquid.

3. Results and Discussion

The experiments for mass transfer in a packed bed were studied based on CO2 absorption into blended solution from MEA-DEA-TEA which is in the range of (0.461–9.541) moles of CO_2 absorbed per mole of amine. Sample taken after absorption experiments

were analyzed for CO_2 and amine content .In order to determine the CO_2 loading in term 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 subtract mole of CO_2 inlet from out let and the CO_2 loading can be calculated through

$$n_{co_2,abs} = n_{CO_2,IN} - n_{CO_2,out} \quad (2)$$

$$\alpha = \frac{\text{mol of } CO_2}{\text{mol of amine}} \quad (3)$$

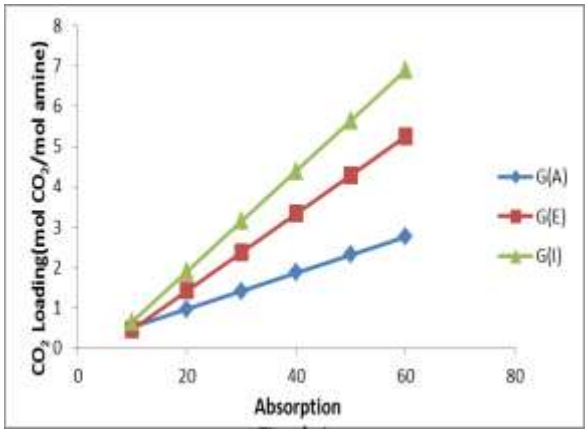

Figure (2) Maximum CO_2 absorption capacity Vs. absorption time for G (A, E, I)

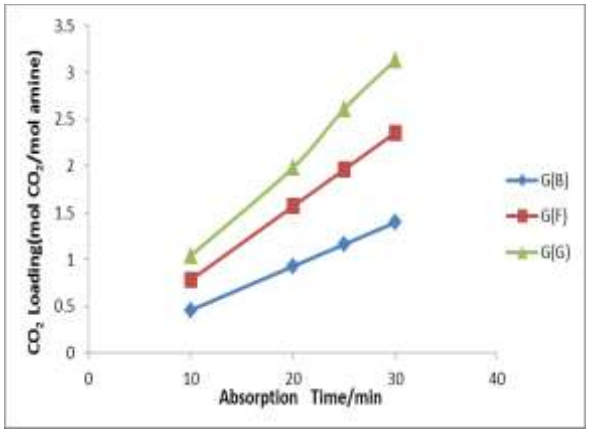

Figure (3) Maximum CO_2 absorption capacity Vs. absorption time for G (B, F,G)

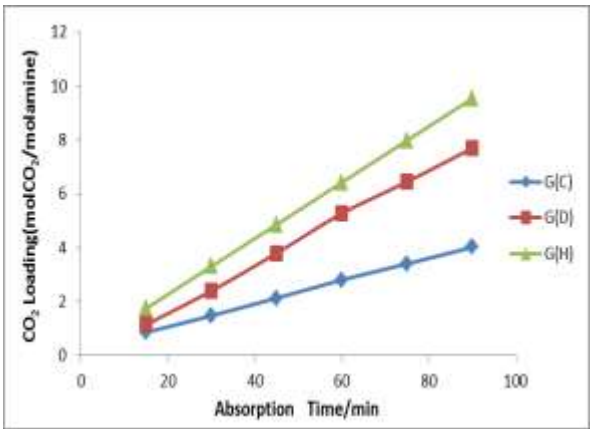

Figure (4) Maximum CO_2 absorption capacity Vs. absorption time for G(C, D, H)

when analyzing Figures (2),(3),(4) We found that An 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. The loading capacity achieved by 5% concentration by V/v % from MEA , 5% DEA and 10%TEA in Figure (2) showed the best CO_2 loading capacity in group I (6.893) mole of CO_2 per mole of amine at 60 min, the gas flow was 15 L/min ,air flow 100 L/h, and liquid flow 450 mL/min . Figure (3) showed the CO_2 loading capacity achieved by 5% concentration by V/v % from MEA , 5% DEA and 10%TEA was in group G (3.132) mole of CO_2 per mole of amine at 30 min, the gas flow was 15 L/min ,air flow 80 L/h, and liquid flow 500 mL/min. Also, Figure (4) showed theCO_2loading capacity achieved by 5% concentration by V/v % from MEA ,5% DEA and 10%TEA was in group H(9.541) mole of CO_2 per mole of amine at 90 min, the gas flow was 15 L/min ,air flow 90 L/h, and liquid flow 400 mL/min.

3.1. The effect of carbon dioxide loading capacity on overall mass transfer coefficient, (KGa)

The following figures shows the main effect of CO_2 loading on overall mass transfer coefficient $\mathbf{K_{Ga}}$ in each group. According to equation (1) calculate the overall mass transfer coefficient $\mathbf{K_{Ga}}(S^{-1})$ for semi-continuous process. In Figures (5), (6) and (7) it is obvious that an increase in CO_2 loading in amine solutions contributes to a decrease in the current active amine concentration, resulting in a decrease in the overall weight transfer coefficient. This effect is primarily due to the high CO_2 loading in the amine solution. Where (MEA+DEA+TEA) blended amine solution are used in all three figures, the mass transfer

driving force from the gas phase to the liquid phase will decrease. This effect is consistent with the work of each of the researchers.

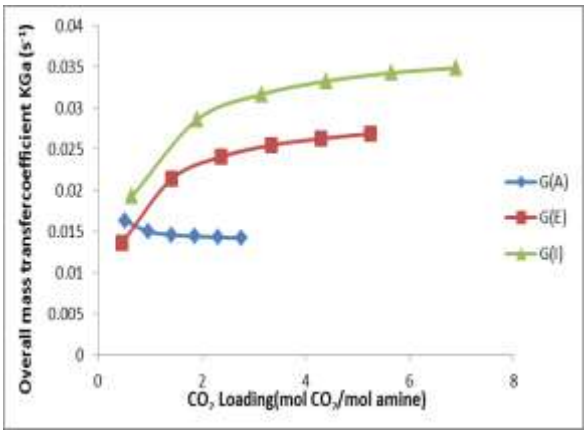

Figure (5) Progression of KGa in a function of CO2 loading for semi-continuous processes (5% MEA+ 5%DEA+10%TEA) G (A, E,I)

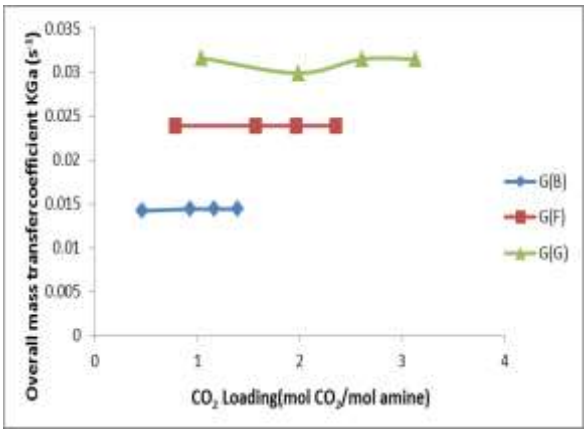

Figure (6) Progression of KGa in a function of CO_2 loading for semi-continuous processes (5% MEA+ 5%DEA+10%TEA) G (B, F, G)

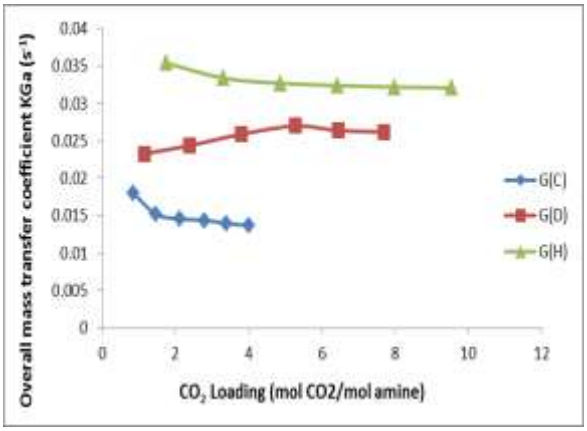

Figure (7) Progression of K_{Ga} in a function of CO_2 loading for semi-continuous processes (5% MEA+ 5%DEA+10%TEA) G (C, D, H)

The maximum value of overall mass transfer coefficient was obtained in Semi-Continuous process (0.0354 at liquid flow 400 mL/min and gas flow 15 L/min in group H for the first 15 min. While [13]they were found that $\mathbf{K_{Ga}}$ was 0.0342 $\boldsymbol{S^{-1}}$ when absorption CO_2 using MEA solution. [14] absorption of CO_2 using aqueous ammonia solution $\mathbf{K_{Ga}}$ was 0.051 $\boldsymbol{S^{-1}}$.

3.2. The effect of gas flow rate on overall mass transfer coefficient, Kga

Gas flow rate is one of the main parameters,which can affect, the efficiency of mass transfer.Many researchers have shown that K_{Ga} also grow when the gas flow rate increased.Figures (5), (6) and (7) presents the evolution of K_{Ga} in function of the time for various gas flow.

For figure (5) group I at the 60minutes of experiment, the mass transfer coefficient is higher K_{Ga} (0.0349 S^{-1}) with increasing in gas flow rate, it was 15 L/min in this group, and the air flow was 100 L/h.

For Figure (6) group G at the first 10 minutes of experiment, the mass transfer coefficient is higher K_{Ga} (0.0316 S^{-1}) with increasing in gas flow rate it was 15 L/min in this group, and the air flow was 80 L/h.

For Figure (7) group H at the first 10 minutes of experiment, the mass transfer coefficient is higher K_{Ga} (0.0354 S^{-1}) with increasing in gas flow rate, it was 15 L/min in this group, and the air flow was 90 L/h.

Also observation all Figures (5), (6) and (7) show that as more gas pass through the packed bed, more CO_2 is absorbed at each point. The turbulence in the gas phase increases as an increase in the gas flow will lead to an increase in K_{Ga}

3.3 Taguchi method of analysis

A packed bed reactor was used to study the capture. Mixed amine concentration was 5V/v% MEA+5 V/v% DEA +10V/v% TEA. We used the Taguchi approach as an experimental design to reduce the multitude of experiments [15], [16]. This study utilized three levels for the variables. When using the Taguchi method, it is important to determine a minimum number of experiments based on the following equation:

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

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 (4) shows the response of the removal efficiency factors to the S / N ratio, Figure (7) show a main effect for four variables that given a maximum values from K_{Ga} at 60 min when gas flow was 15 L/min and air flow 90 L/h and liquid flow 450 ml/min.

Table (4) Response of factors to S/N ratio for removal efficiency

Level	Gas (CO_2)flow	Air flow	Liquid flow	Time
1	-34.01	-29.86	-30.08	-30.09
2	-28.81	-29.69	-29.53	-29.49
3	-26.67	-29.94	-29.89	-29.91
Delta	7.34	0.24	0.55	0.60
Rank	1	4	3	2

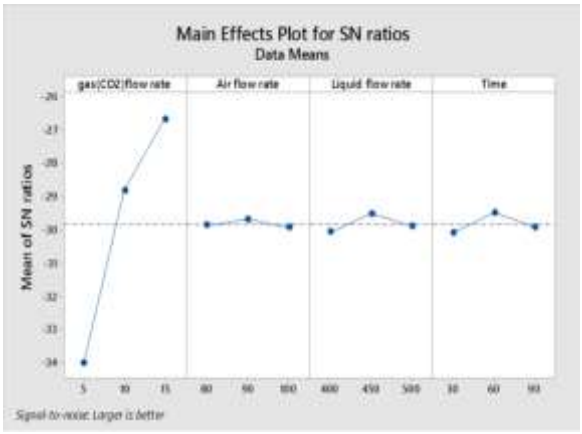

Figure (7) The optimum point of mass transfer coefficient of blended amine with 4-factor in Taguchi analysis program

4. Conclusions

The maximum carbon dioxide capture was achieved by the use of a packed bed under semi-continuous operation. The importance of this work lies in finding new alternatives to obtain maximum absorption in the column and high carbon oxide concentrations after dissolving it in the packed column. KGa 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 KGa. This study showed that the increasing by the gas flow rate, the liquid flow rate, the KGa values increased. Also,increasing the CO2 loading of amines lead to a KGa decrease. The maximum value of CO2 loading was obtained in process 9.541 (mole CO2/mole amine) at liquid flow 400 mL/min, gas flow 15 L/min and air flow 90 L/h in group I at 90 min. The maximum value of overall mass transfer coefficient was obtained in process (0.0354 at liquid flow 400 mL/min and gas flow 15 L/min in group H for the first 15 min.

5. Acknowledgments

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