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Carbon Dioxide Absorption Using Different Solvents (MEA, NaOH, KOH and $Mg(OH)_2$) in Bubble Column Reactor

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Abstract
Carbon dioxide is considered to be one of the greenhouse gases potentially responsible for climate change. The aim of this research is to reduce emissions by capturing carbon dioxide in a solution using an absorption method. The absorption capacity, absorption rate, carbon dioxide removal efficiency, and overall mass transfer coefficient of MEA (Monoethanolamin) and alkaline solvents (NaOH, KOH, Mg $(OH)_2$) were investigated using a bubble column gas absorption reactor with counter current flow. The effects of operational parameters such as solvent concentration (0.01, 0.05, and 0.25M) and solvent type were studied. The research showed that KOH, NaOH, and MEA were more efficient in capturing CO_2 than Mg $(OH)_2$ was. For all solvent types, the total mass transfer coefficient, absorption rate, and CO_2 removal efficiency were increased with the increase in the concentration of solvent. The solvent concentration is increased from 0.01 M to 0.25 M to obtain the highest KGa values for MEA, NaOH, and KOH, 3.75 1/min for MEA, 3.70 1/min for NaOH, and 3.93 1/min for KOH.The MEA, NaOH, and KOH absorption rates were maximum at 0.25 M solvent concentrations as 0.19x103 mol/Ls. The maximum CO_2 removal efficiencies for MEA, NaOH, and KOH at 0.25 M solvent concentration are greater than 60%. Absorption capacity of NaOH and KOH is 0.313 mol CO_2/mol NaOH and 0.316 (mol CO_2/mol KOH). The highest absorption capacity, 0.576 mol CO_2/mol MEA, was obtained at a solvent concentration of 0.01M MEA.

1. Introduction

The temperature of the Earth has risen especially quickly since the middle of the 20th century. Global warming emerges as one of the concerns facing the planet during the industrial revolution. Natural and human systems have undergone significant changes in response to long-term changes in the climate system caused by global warming. These changes include an increase in the frequency of catastrophic events like floods and droughts, a rise in sea level caused by glacier and polar ice cap melting, and severe ecological destruction that threatens the sustainability of the economy [1, 2].

Fossil fuels are used to generate the majority of the energy needed to meet the growing demand, which raises the atmospheric carbon concentration. Fossil fuel consumption contributes to the release of greenhouse gases such carbon dioxide (76%), methane (16%), nitrous oxide (6%), and fluorinated gases (2%) on a worldwide scale. Carbon dioxide (CO_2) is therefore frequently viewed as the main contributor to rising world average temperatures [3, 4].

The majority of today's energy needs are met by the generation of electricity from fossil fuels, carbon capture and storage (CCS) is the best option for decreasing CO_2 emissions. Pre-combustion capture, post-combustion capture, and oxy-fuel

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combustion are three groups of CO_2 capturing technologies. Among these, post-combustion capture using alkanolamines is considered as one of the most practical and widely used for the removal of CO_2 successfully [5]. There are various methods to reduce emissions of CO_2 post combustion such as chemical absorption [6], physical absorption [7], membrane separation [8], adsorption [9], cryogenic separation [10] and algal system [11]. In these processes, chemical absorption with NaOH and amine solutions are extensively used for the capture of CO_2 [12].

Aqueous MEA solution has been widely used to capture CO_2 in industrial processes due to its strong CO_2 reaction kinetics, high solubility in water, low viscosity, lower energy use, and low cost. [13]. Alkaline solvents that widely used in CO_2 removal are NaOH which has high absorption efficiency and potassium hydroxide (KOH) [14]. It is possible to enhance CO_2 absorption by utilizing different scrubber and solvent kinds. Numerous scrubbers, such as packed bed columns, sieve tray columns, and bubble columns, are used to capture CO_2.

Bubble column reactors offer a variety industrial application due to its simple design and operation, lack of moving parts, highly complicated hydrodynamic behavior, and high rates of mass and heat transfer [15]. Specifically, the packed tower and bubble column have been effectively used on pilot and industrial scales for post-combustion CO_2 capture [16].

The aim of this research is to improve the CO_2 absorption using different solvents. For this aim, the capture of CO_2 was performed using MEA and alkaline solvents (NaOH, KOH and $Mg(OH)_2$) in a bubble column. The effect of solvent types on the CO_2 removal capacity, absorption rate and overall mass transfer coefficient was determined.

2. Calculations

CO_2 Removal Efficiency

CO_2 removal efficiency is a critical consideration when assessing the performance of an absorption and calculated using Eq.1.

$$\mathbf{E} = \left(\frac{y_1 - y_2}{y_1}\right) * 100\% \quad (1)$$

y_1 = CO_2 input concentration; y_2 = CO_2 output concentration

Absorption Rate

The most crucial factor in determining solvent costs, which account for around 30% of overall capital costs, is CO_2 absorption rate (RA) [17]. Liquid holdup (and gas holdup) is considered constant throughout the column to determine the absorption rate. As a result, under steady-state operation, the rate of carbon dioxide absorption can be determined. Absorption rate is determined using Eq.2 by measuring the effluent concentration of carbon dioxide and the gas-flow rate:

$$R_A = \frac{F_{A1}}{V_L}\left[1 - \left(1 - \frac{y_1}{y_1}\right)\left(\frac{y_2}{1 - y_2}\right)\right] \quad (2)$$

FA_1 = molar flow rate of CO_2 inlet

V_L = Solvent solution volume (final volume)

y_1 and y_2 = CO_2 concentration of inlet and outlet

R_A = Absorption rate

Overall Mass Transfer Coefficient

In separation processes, the diffusion of mass from one phase to the other is occurred, and the diffusion rate is a crucial factor that effect the overall mass transfer coefficient. The two-film model, a helpful model for mass transfer between phases, is used to calculate the mass transfer coefficient and the mass transfer of CO_2. CO_2 is transferred from the bulk of the gas phase to the interface and then moved from the interface into the bulk of the liquid phase during mass transfer. The two film model assumes equilibrium at the interface (Fig. 1).

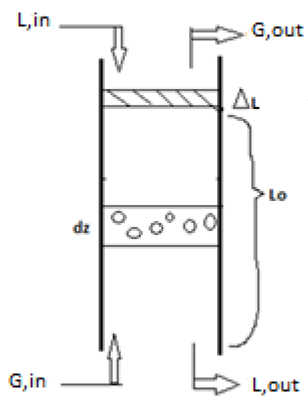

Figure 1. Schematic representation of two film theories

According to two film model, the absorption rate at a local point on both the gas and liquid side is expressed as total mass transfer coefficients and can be written as follows;

$$r_A = (K_G a)(C_g - HC_L) \quad (3)$$

With the assumption of plug flow condition at the gas phase and well mixed flow condition at the liquid phase and steady state condition, the mass equilibrium at z will be;

$$(U_g C_g)_z^S - (U_g C_g)_{z+\Delta z}^S = r_A \Delta V \quad (4)$$

$$\Delta V = \Delta z * S \quad (5)$$

Readjust the equation;

$$(U_g C_g)_z . S - (U_g C_g)_{z+\Delta z} . S = K_G a . (C_g - HC_L)(\Delta Z . S); \quad (6)$$

$$\frac{\boldsymbol{S.U(C_{g_z} - C_{g_{z+\Delta z}})}}{\boldsymbol{\Delta z}} = K_G a . (C_g - HC_L) . S \quad (7)$$

$$HC_L \simeq 0$$

$$S . U \frac{dC}{dz} = S . K_G a . (C_g) \quad (8)$$

$$Q \frac{dC}{C} = S . K_G a . dz \quad (9)$$

$$Q \int_{Cin}^{Cout} \frac{dC}{C} = \int_0^{L+\Delta L} S . K_G a . dz \quad (10)$$

$$K_G a = \frac{Q_g \ln \frac{C_0}{C}}{(\Delta L + L) . S} \quad (11)$$

Where; Cg, CO_2 gas concentration in gas phase (mol/L); CL, CO_2 gas concentration in liquid phase (mol/L); r_A, absorption rate (mol/Ls) ; $K_G a$,mass-transfer coefficient (1/min), S: column cross sectional area (cm^2), U: surface velocity (m/s) , Q, velocity of gas flow (l/min)

Absorption capacity

The area over the CO_2-time profile graph (Fig. 2) corresponds to the total absorbed CO_2. The input flow rate of CO_2 was calculated from total flow rate and the inlet concentration. The outlet flow rate of CO_2 was calculated based on the fixed flow rate of N_2 which was an inert compound and the read CO_2 concentration. The following relation was used to calculate the CO_2 outlet flow;

$$Q_{CO_{2out}} = Q_{total_{in}} \times y_{N_{2in}} \left(\frac{y_{CO_{2out}}}{y_{N_{2out}}}\right) \quad (12)$$

The volumetric flow rates were converted to molar mass flow rate using conversion factors with the assumption of ideal gas of state where each mole at standard temperature (273 K) and pressure (1 atm) occupies 22.4 L. Then it calculated again for the adjusted temperature of gas. Then the concentration (ppm)-time graph was replotted for mass flow rate-time.

The rate of absorbed CO_2 at certain time intervals was then calculated using following equation;

$$R_{CO_2} = \dot{M}_{CO_{2in}} - \dot{M}_{CO_{2out}} \quad (13)$$

The amount of absorbed CO_2 at each time interval was calculated using following equation;

$$M_{CO_{2ab}} = R_{CO_2} \times (t_2 - t_1) \quad (14)$$

The absorption capacity of the absorbent was calculated using below equation;

$$Ab. Cap = \frac{\sum_1^n M_{CO_{2ab}}}{M_{MEA}} \quad (15)$$

Where n is the number of time intervals, MCO_2 is the mass of absorbed CO_2 and MMEA is the mass of MEA in the solution. Spreadsheets in MS Excel was used for calculation procedures.

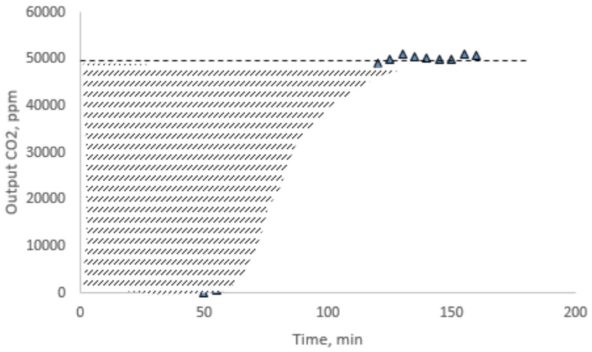

Figure 2. A sample of the CO_2 concentration profile at the output

2.1. Chemical Reaction Mechanism

NaOH

NaOH is the widely used solvent for the CO_2 capture even though, solvent is not recoverable from the reaction between CO_2 and NaOH, because it is more abundant, cheaper than MEA, and has higher CO_2 absorption capacity than MEA. Theoretically, 1.39 tons MEA and 0.9 tons NAOH required to capture one ton of CO_2, respectively. As shown below, the process by which CO_2 is absorbed by NaOH in aqueous solution [15];

Firstly, NaOH is completely ionized in water. Secondly, when gas fed into the NaOH solution, carbon dioxide is physically absorbed as aqueous carbon dioxide because NaOH is strongly alkaline.

$$CO_{2(g)} \rightarrow CO_{2(aq)} \quad (16)$$

Subsequently, aqueous CO_2 reacts with OH- as expressed in Eqs. 17 and 18 to form HCO_3^- and CO_3^{2-}

$$CO_{2aq)} + OH^-{}_{(aq)} \leftrightarrow HCO_3^-{}_{(aq)} \quad (17)$$

$$HCO_3^-{}_{(aq)} + OH^-{}_{(aq)} \leftrightarrow H_2O_{(l)} + CO_3^{2-}{}_{(aq)} \quad (18)$$

The reaction that occurring during CO_2 absorption is shown below;

$$2NaOH_{(aq)} + CO_{2(g)} \rightarrow Na_2CO_{3(aq)} + H_2O_{(l)} \quad (19)$$

The Na_2CO_3 produced in this reaction exists in solvent as ionized Na^+ and CO_3^{2-}. The NaOH solution is continuously fed CO_2, which causes CO_2 to be absorbed and deplete the OH^- level. The general absorption reaction is shown in equation 20.

$$Na_2CO_{3(aq)} + CO_{2(g)} + H_2O_{(l)} \rightarrow 2NaHCO_{3(aq)} \quad (20)$$

The net reaction of equations 2.19 and 2.20 can be summarized as equation 21 [12].

$$NaOH_{(aq)} + CO_{2(g)} \rightarrow NaHCO_{3(aq)} \quad (21)$$

KOH

In the process of removing CO_2, potassium hydroxide (KOH) is the second-most-used solvent. The reaction with carbon dioxide can be seen in Eq. 22.

$$2KOH_{(aq)} + CO_{2(g)} \rightarrow K_2CO_{3(aq)} + H_2O_{(l)} \quad (22)$$

KOH and NaOH are both used in the same chemical reaction for the absorption of CO_2. However, KOH is more expensive than NaOH, but the cost of KOH can be reduced by selling the side product of K_2CO_3 [18].

$Mg(OH)_2$

Absorption with magnesium hydroxide $Mg(OH)_2$ occur in several stages and the main reactions involved in the absorption process are as follows:

$$Mg(OH)_{2(s)} \leftrightarrow Mg^{2+}{}_{(aq)} + 2OH^-{}_{(aq)} \quad (23)$$

$$CO_{2(aq)} + OH^-{}_{(aq)} \leftrightarrow HCO_3^-{}_{(aq)} \quad (24)$$

$$HCO_3^-{}_{(aq)} + OH^-{}_{(aq)} \leftrightarrow H_2O_{(l)} + CO_3^{2-}{}_{(aq)} \quad (25)$$

$$Mg^{2+}{}_{(aq)} + CO_3^{2-}{}_{(aq)} \leftrightarrow MgCO_{3(s)} \quad (26)$$

Dissolution of solid particles in the liquid film increases the absorption rate [19].

MEA

A key solvent in the CO_2 removal process is monoethanolamine (MEA) solution due to how rapidly it reacts with carbon dioxide [20]. The reaction mechanism among H_2O-CO_2-amine differs based on the number of amine functionality. The reaction mechanism for single amine functionality like MEA has been suggested as follow [21]:

Water dissociation:

$$2H_2O \rightleftharpoons H_3O^+ + OH^- \qquad (27)$$

Carbon dioxide dissociation:

$$2H_2O + CO_2 \rightleftharpoons H_3O^+ + HCO_3{}^- \qquad (28)$$

Bicarbonate dissociation:

$$H_2O + HCO_3{}^- \rightleftharpoons H_3O^+ + CO_3{}^{2-} \qquad (29)$$

Dissociation of protonated MEA:

$$H_2O + RNH_3{}^+ \rightleftharpoons H_3O^+ + RNH_2 \qquad (30)$$

Carbamate reversion to bicarbonate:

$$H_2O + RNHCOO^- \rightleftharpoons RNH_2 + HCO_3{}^- \qquad (31)$$

3. MATERIALS AND METHOD

3.1. Chemicals

The chemicals of NaOH, KOH, $Mg(OH)_2$,MEA were purchased from Merck, Germany. All of the chemical was of the reagent grade. Double-distilled water which was obtained in the lab using a water purification equipment (Thermo Scientific, Germany) was used to prepare the aqueous solutions. The gas phase consisted of CO_2 and N_2 was prepared using CO_2 and N_2 gas cylinders (99.99% purity) obtained from Oksan LTD, Turkey.

3.2. Experimental setup

The absorption bubble column used in this study can be seen in Fig.3. It is made from plexiglass with the height of 1m and internal diameter (ID) of 5 cm. It was operated counter current flow in which downflow of the liquid and upflow of the gas were applied. All the absorption experiment were carried out at room temperature, For each experiment, the temperature of the water circulation bath was adjusted on the desired temperature and run for almost 25 min to ensure stable temperature on the vessel wall. The solutions was prepared in parallel and put over hot plate to obtain the desired temperature and quickly empty into solvent tank and left for 10 minutes to stabilize the temperature. The desired solvent concentration was prepared and poured into the feed tank. The pump is used to control the required liquid flow rate. The gas mixture was supplied using two separate mass flow controllers (ALICAT Scientific Mass Flow Controller, Range:0-10L/min, accuracy; %0,2 of full-scale) for nitrogen and carbon dioxide. The gas mixture was sent directly to the CO_2 analyzer to confirm the initial CO_2 concentration, and then the main line valves were opened to transfer the gas mixture to the column containing MEA solution. The carbon dioxide concentration in the gas phase in the output was monitored using a Vernier CO_2 gas sensor (USA). The process continued until there was no further absorption. This was confirmed by the concentration/time profile as shown in Fig. 2.

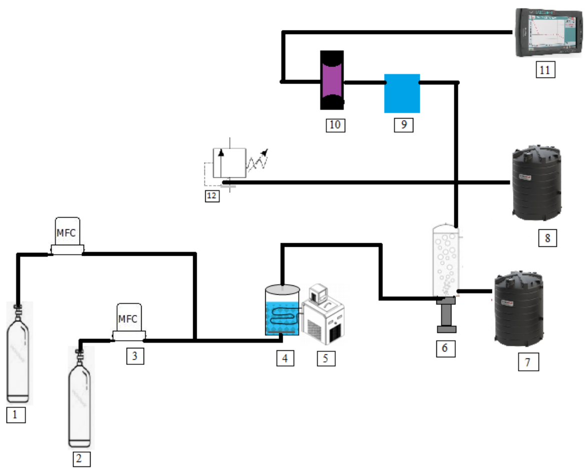

Figure 3. Experimental setup of CO_2 absorption.

(1: CO_2 cylinder, 2: N_2 cylinder, 3: Mass flow controller, 4: Humidifier, 5: Heat exchanger, 6: Column, 7: Waste tank, 8: Solvent tank, 9: Dehumidifier gas regulator, 10: Dehumidifier, 11:

CO_2 Analyzer (10.000-100.000ppm), 12: Relief valve.)

4. Results and Discussion

In this study various solvent type and concentrations were used to determine its effects on CO_2 absorption. MEA, NaOH, KOH, and Mg $(OH)_2$ solvents were used for this purpose at concentrations of 0.01 M, 0.05 M, and 0.25 M. Results are presented for a gas flow rate of 4 L/min and a liquid flow rate of 500 mL/min.

4.1. Effect of Solvent Concentration on CO_2 Removal Efficiency

The effects of MEA, NaOH, KOH and Mg $(OH)_2$ solvents concentrations (0.01-0.05-0.25M) on carbon dioxide removal efficiency were investigated at 4.0 L/min gas flow rate, 500 mL/min liquid flow rate and 5% CO_2 initial concentration (50 000 ppm) and results can be seen from Fig. 4. For all solvent types, it has been observed that the CO_2 removal efficiency increases when the solvent concentration increases. However, the difference in CO_2 removal efficiency is not very noticeable at high concentrations of NaOH and KOH. High CO_2 removal efficiencies were also obtained at low solvent concentrations. The regeneration of NaOH, KOH, and Mg $(OH)_2$ is quite difficult, in contrast to MEA regeneration. The regeneration of NaOH and KOH solvents is difficult because the final products Na_2CO_3 and K_2CO_3 are formed as a result of absorption and their regeneration is costly due to their high energy requirements [18]. The increase in solvent concentration also means the increase in the reactant amount which leeds to higher CO_2 removal [15]. High active MEA concentration in the liquid solution encourages its diffusion to the gas-liquid interface [22]. Smilarly, Yincheng et al. found that a higher NaOH concentration increases CO_2 removal efficiency [23].

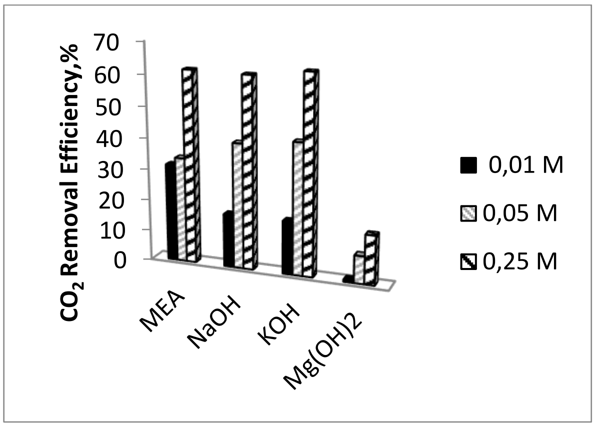

Figure 4. Effects of solvent concentrations on CO_2 removal efficiency.

4.2. Effect of Solvent Concentration on Absorption Capacity

The solvent concentration has a significant effect on absorption capacity. The impact of various solvent concentrations on absorption capacity in the bubble column is shown in Fig. 5. The gas flow rate of 4.0 L/min, solvent flow rate of 500 mL/min and the CO_2 concentration of 50000 ppm are used. As seen from Fig.5, low concentration of solvents has a better absorption capacity, which means that higher amount of CO_2 was absorbed by a mol of solvent. The highest absorption capacity was obtained at solvent concentration of 0.01M MEA as 0.576 mol CO_2/mol MEA. Absorption capacity of NaOH and KOH is 0.313 mol CO_2/mol NaOH and 0.316 (mol CO_2/mol KOH), respectively.

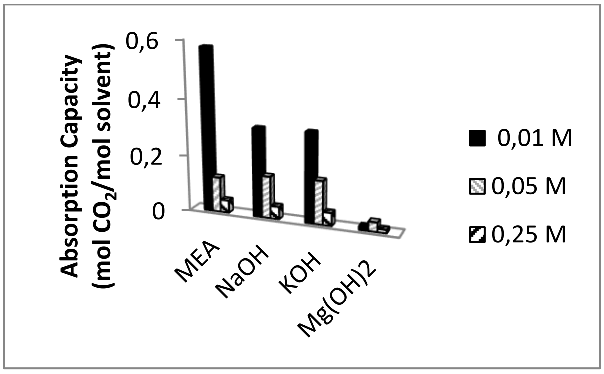

Figure 5. Effects of solvent concentrations on absorption capacity.

The increase in the solvent concentrations from 0.01 to 0.25 mol/L, decreases the absorption capacity in the bubble column. Similar trends were found in the literature [15].

4.3. Effect of Solvent Concentration on Absorption Rate

The effect of different solvent concentrations on absorption rate in the bubble column is shown in Fig. 6. The absorption rates were increased with the increase of solvent concentration and absorption rates of MEA, NaOH and KOH are almost same at 0.25M.

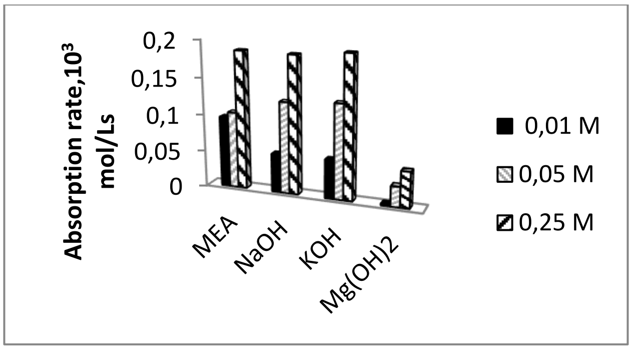

Figure 6. Effects of solvent concentrations on absorption rate.

Chen Chi obtained absorption rate of 0.0135 - 0.622 (103 mol / Ls) at 4 M MEA in bubble column [25]. Yoo et al. investigated the capacity, rate, and efficiency of CO_2 absorption by NaOH aqueous solution in a batch-style Pyrex cylindrical reactor. They found that the absorption rate increased with the concentration of NaOH [14].

4.4. Effect of Solvent Concentration on Overall Mass Transfer Coefficient

The effect of various solvent concentrations on the total mass transfer coefficient in the bubble column is shown in Fig. 7. Except for Mg $(OH)_2$, the mass transfer coefficients of the solvents were similar at 0.25 M solvent concentration, however it sharply declined at lower concentrations. As shown in Fig. 7, an increase in the solvent concentration results a higher KGa value. When the solvent concentration is increased from 0.01 M to 0.25 M, the KGa value increases from 1.47 1/min to 3.75 1/min for MEA, from 0.72 1/min to 3.70 1/min for NaOH, from 0.74 1/min to 3.93 1/min for KOH, and from 0.027 1/min to 0.67 1/min for Mg $(OH)_2$.

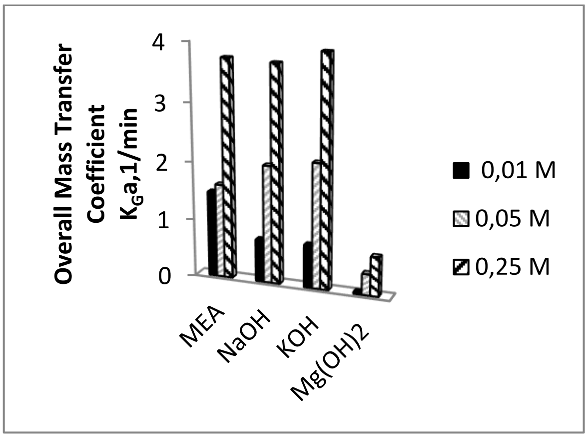

Figure 7. Effects of solvent concentrations on overall mass transfer coefficient.

Wu et al. found that when the MEA concentrations increase from 10 wt.% to 40 wt.% at a fixed 12 vol% CO_2 inlet concentration, the overall mass transfer coefficient increases from 0.2943 to 0.4044 kmol/m^3. h. kPa. This is caused by the fact that an increase in MEA concentration produces more active MEA molecules that are available to diffuse toward the gas-liquid surface and subsequently react with CO_2 molecules, which increases the reaction enhancement factor and results in better mass transfer performance [26]. According to Cheng et al., a higher Mg $(OH)_2$ concentration results in a higher mass transfer coefficient [27].

5. Conclusion

In this study, the effects of solvent type and concentration on absorption capacity (mol CO_2/mol solvent), absorption rate (mol/Ls), carbon dioxide removal efficiency (%), and total mass transfer coefficient (1/min) was investigated using a bubble column reactor with a countercurrent flow. Experiments were performed at gas flow rate of 4.0 L/min, liquid flow rate of 500 mL/min and 5% CO_2 initial concentration. Experimental results show that the aqueous solvent concentrations have a great effect on the absorption capacity, absorption rate, carbon dioxide removal efficiency and total mass transfer coefficient. Whit the increasing solvent concentration, the overall mass transfer coefficient, absorption rate, and CO_2 removal efficiency increased while the absorption capacity decreased. The CO_2 concentration of 50000 ppm was reduced to the 20000 ppm with the approximately removal efficiency of %60 using bubble column. When the solvent concentration is increased from 0.01 M to 0.25 M, the KGa value increases from 1.47 1/min to 3.75 1/min for MEA, from 0.72 1/min to 3.70 1/min for NaOH, from 0.74 1/min to 3.93 1/min for KOH, and from 0.027 1/min to 0.67 1/min for Mg $(OH)_2$. The highest absorption rates for MEA, NaOH, and KOH were obtained at 0.25 M solvent concentrations as 0.19x103 mol/Ls. The highest absorption capacity was obtained at solvent concentration of 0.01M MEA as 0.576 mol CO_2/mol MEA. Absorption capacity of NaOH and KOH is 0.313 mol CO_2/mol NaOH and 0.316 (mol CO_2/mol KOH), respectively. As a result of the study, it can be said that the removal of CO_2 from flue gases using bubble column can be achieved successfully.

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Contributions of the Authors

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Ayse Gul and Umran Tezcan Un.

Conflict of Interest Statement

There is no conflict of interest between the authors.

Statement of Research and Publication Ethics

The study is complied with research and publication ethics

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