

Carbon Dioxide Absorption Using Different Solvents (MEA, NaOH, KOH and Mg(OH)₂) in Bubble Column Reactor

Ayşe GÜL^{1*}, Umran TEZCAN UN²

¹Department of Civil Engineering, Abdullah Gul University, Kayseri, TURKEY

²Department of Environmental Engineering, Eskisehir Technical University, Eskisehir, TURKEY
(ORCID: 0000-0002-2305-6408) (ORCID: 0000-0003-3882-9175)



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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 CO₂ than Mg (OH)₂ was. For all solvent types, the total mass transfer coefficient, absorption rate, and CO₂ 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 CO₂ 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 CO₂/mol NaOH and 0.316 (mol CO₂/mol KOH). The highest absorption capacity, 0.576 mol CO₂/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 (CO₂) 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 CO₂ emissions. Pre-combustion capture, post-combustion capture, and oxy-fuel

*Corresponding author: ayse.gul@agu.edu.tr

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combustion are three groups of CO₂ capturing technologies. Among these, post-combustion capture using alkanolamines is considered as one of the most practical and widely used for the removal of CO₂ successfully [5]. There are various methods to reduce emissions of CO₂ 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 CO₂ [12].

Aqueous MEA solution has been widely used to capture CO₂ in industrial processes due to its strong CO₂ reaction kinetics, high solubility in water, low viscosity, lower energy use, and low cost. [13]. Alkaline solvents that widely used in CO₂ removal are NaOH which has high absorption efficiency and potassium hydroxide (KOH) [14]. It is possible to enhance CO₂ 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 CO₂.

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 CO₂ capture [16].

The aim of this research is to improve the CO₂ absorption using different solvents. For this aim, the capture of CO₂ was performed using MEA and alkaline solvents (NaOH, KOH and Mg (OH)₂) in a bubble column. The effect of solvent types on the CO₂ removal capacity, absorption rate and overall mass transfer coefficient was determined.

2. Calculations

CO₂ Removal Efficiency

CO₂ removal efficiency is a critical consideration when assessing the performance of an absorption and calculated using Eq.1.

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

y_1 = CO₂ input concentration; y_2 = CO₂ output concentration

Absorption Rate

The most crucial factor in determining solvent costs, which account for around 30% of overall capital costs, is CO₂ 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:

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

F_{A1} = molar flow rate of CO₂ inlet

V_L = Solvent solution volume (final volume)

y_1 and y_2 = CO₂ 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 CO₂. CO₂ 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).

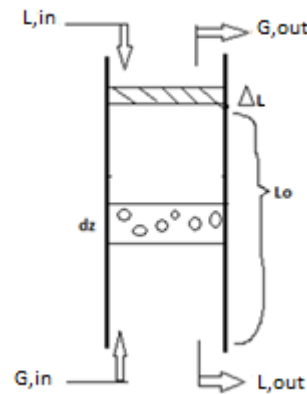


Figure 1. Schematic representation of two film theories

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;

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

$$\frac{S \cdot U (C_{gz} - C_{gz+\Delta z})}{\Delta z} = K_G a \cdot (C_g - HC_L) \cdot S \quad (7)$$

$$HC_L \approx 0$$

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

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

$$Q \int_{C_{in}}^{C_{out}} \frac{dC}{C} = \int_0^{L+\Delta L} S \cdot K_G a \cdot dz \quad (10)$$

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

Where; C_g , CO₂ gas concentration in gas phase (mol/L); CL , CO₂ 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 (cm²), U : surface velocity (m/s) , Q , velocity of gas flow (l/min)

Absorption capacity

The area over the CO₂-time profile graph (Fig. 2) corresponds to the total absorbed CO₂. The input flow rate of CO₂ was calculated from total flow rate and the inlet concentration. The outlet flow rate of CO₂ was calculated based on the fixed flow rate of N₂ which was an inert compound and the read CO₂ concentration. The following relation was used to calculate the CO₂ outlet flow;

$$Q_{CO_2 \text{out}} = Q_{\text{total in}} \times y_{N_2 \text{in}} \left(\frac{y_{CO_2 \text{out}}}{y_{N_2 \text{out}}} \right) \quad (12)$$

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 CO₂ at certain time intervals was then calculated using following equation;

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

The amount of absorbed CO₂ at each time interval was calculated using following equation;

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

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

$$Ab. Cap = \frac{\sum_1^n M_{CO_2 ab}}{M_{MEA}} \quad (15)$$

Where n is the number of time intervals, M_{CO_2} is the mass of absorbed CO₂ and M_{MEA} is the mass of MEA in the solution. Spreadsheets in MS Excel was used for calculation procedures.

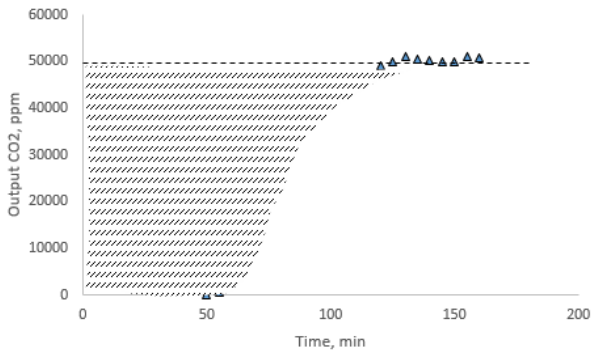


Figure 2. A sample of the CO₂ concentration profile at the output

2.1. Chemical Reaction Mechanism

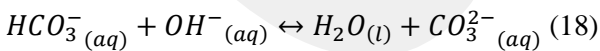
NaOH

NaOH is the widely used solvent for the CO₂ capture even though, solvent is not recoverable from the reaction between CO₂ and NaOH, because it is more abundant, cheaper than MEA, and has higher CO₂ absorption capacity than MEA. Theoretically, 1.39 tons MEA and 0.9 tons NAOH required to capture one ton of CO₂, respectively. As shown below, the process by which CO₂ is absorbed by NaOH in aqueous solution [15];

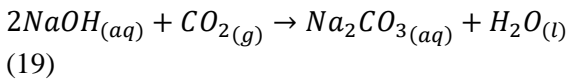
Firstly, NaOH is completely ionized in water. Secondly, when gas fed into the NaOH solution, carbon dioxide is physically absorbed as aqueous carbon dioxide because NaOH is strongly alkaline.



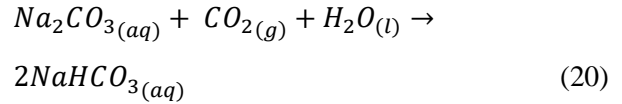
Subsequently, aqueous CO₂ reacts with OH⁻ as expressed in Eqs. 17 and 18 to form HCO₃⁻ and CO₃²⁻



The reaction that occurring during CO₂ absorption is shown below;



The Na₂CO₃ produced in this reaction exists in solvent as ionized Na⁺ and CO₃²⁻. The NaOH solution is continuously fed CO₂, which causes CO₂ to be absorbed and deplete the OH⁻ level. The general absorption reaction is shown in equation 20.

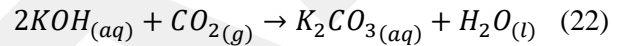


The net reaction of equations 2.19 and 2.20 can be summarized as equation 21 [12].



KOH

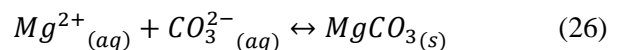
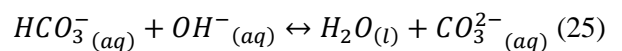
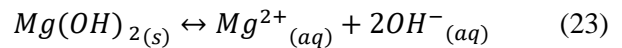
In the process of removing CO₂, potassium hydroxide (KOH) is the second-most-used solvent. The reaction with carbon dioxide can be seen in Eq. 22.



KOH and NaOH are both used in the same chemical reaction for the absorption of CO₂. However, KOH is more expensive than NaOH, but the cost of KOH can be reduced by selling the side product of K₂CO₃ [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:

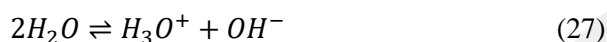


Dissolution of solid particles in the liquid film increases the absorption rate [19].

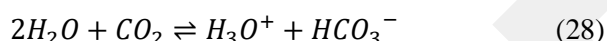
MEA

A key solvent in the CO₂ removal process is monoethanolamine (MEA) solution due to how rapidly it reacts with carbon dioxide [20]. The reaction mechanism among H₂O-CO₂-amine differs based on the number of amine functionality. The reaction mechanism for single amine functionality like MEA has been suggested as follow [21]:

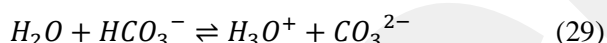
Water dissociation:



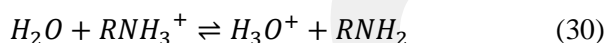
Carbon dioxide dissociation:



Bicarbonate dissociation:



Dissociation of protonated MEA:



Carbamate reversion to bicarbonate:



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 CO₂ and N₂ was prepared using CO₂ and N₂ 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 CO₂ analyzer to confirm the initial CO₂ 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 CO₂ 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.

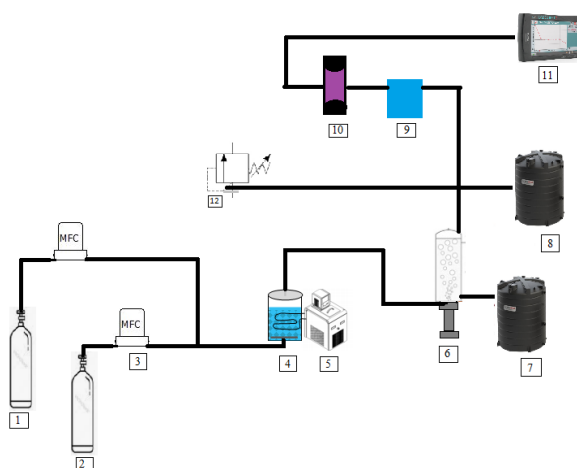


Figure 3. Experimental setup of CO₂ absorption.

(1: CO₂ cylinder, 2: N₂ 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:

CO₂ 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 CO₂ 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 CO₂ 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% CO₂ initial concentration (50 000 ppm) and results can be seen from Fig. 4. For all solvent types, it has been observed that the CO₂ removal efficiency increases when the solvent concentration increases. However, the difference in CO₂ removal efficiency is not very noticeable at high concentrations of NaOH and KOH. High CO₂ 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 Na₂CO₃ and K₂CO₃ 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 leads to higher CO₂ removal [15]. High active MEA concentration in the liquid solution encourages its diffusion to the gas-liquid interface [22]. Similarly, Yincheng et al. found that a higher NaOH concentration increases CO₂ removal efficiency [23].

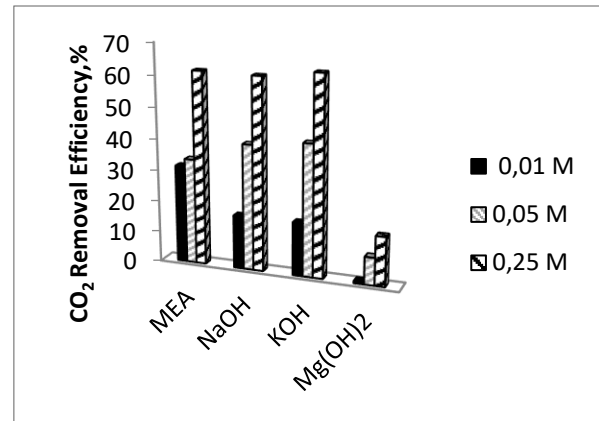


Figure 4. Effects of solvent concentrations on CO₂ removal efficiency.

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 CO₂ 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 CO₂ was absorbed by a mol of solvent. The highest absorption capacity was obtained at solvent concentration of 0.01M MEA as 0.576 mol CO₂/mol MEA. Absorption capacity of NaOH and KOH is 0.313 mol CO₂/mol NaOH and 0.316 (mol CO₂/mol KOH), respectively.

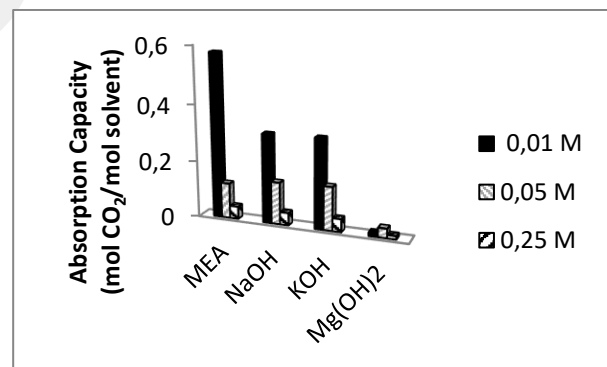


Figure 5. Effects of solvent concentrations on absorption capacity.

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.

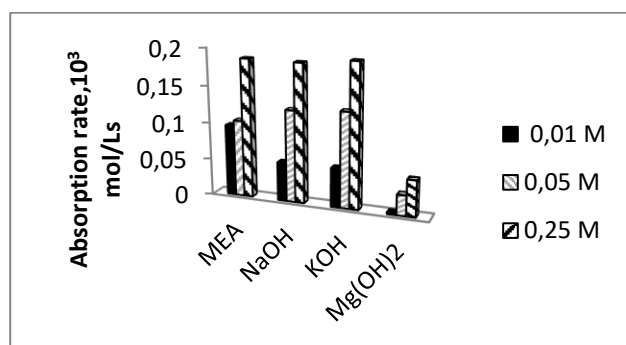


Figure 6. Effects of solvent concentrations on absorption rate.

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 CO₂ 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.

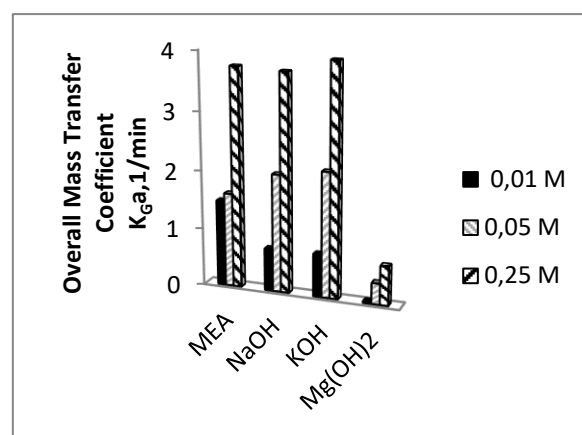


Figure 7. Effects of solvent concentrations on overall mass transfer coefficient.

Wu et al. found that when the MEA concentrations increase from 10 wt.% to 40 wt.% at a fixed 12 vol% CO₂ inlet concentration, the overall mass transfer coefficient increases from 0.2943 to 0.4044 kmol/m³. 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 CO₂ 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 CO₂/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% CO₂ 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. With the increasing solvent concentration, the overall mass transfer coefficient, absorption rate, and CO₂ removal efficiency increased while the absorption capacity decreased. The CO₂ 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 CO₂/mol MEA. Absorption capacity of NaOH and KOH is 0.313 mol CO₂/mol NaOH and 0.316 (mol CO₂/mol KOH), respectively. As a result of the study, it can be said that the removal of CO₂ 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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