Improving the activity of CO_2 capturing from flue gas by membrane gas – solvent absorption process

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Abstract

This work is focused on increasing the capturing efficiency of carbon dioxide (CO_2) through flue gas purification systems. To maximize the CO_2 capture process, many process variables such as temperature, flow rates, absorbent concentrations, and nanoparticles were investigated. This study describes the use of a polypropylene hollow fiber membrane contactor to separate CO_2 from nitrogen using different solvents, including Potassium carbonate (K_2CO_3), N-methyl diethanolamine (MDEA), and monoethanolamine (MEA). Also, the presence of silica nanoparticles and piperazine (PZ) enhances the process performance. On the other hand, the amine and mixed amino absorbents MDEA-PZ and MDEA-MEA were prepared and compared based on the absorption capacity. The optimal order of amine absorbent performance when applied to CO_2 membrane absorption is MDEA-MEA followed by MDEA-PZ. At a solute concentration of 9%, MDEA-MEA exhibits the highest CO_2 removal efficiency, which is 74.12%. However, at a concentration of 11%, MEA, MDEA-PZ, and MDEA have the highest CO_2 removal efficiencies of 80.15%, 75.13%, and 63.12%, respectively.

Keywords: Polypropylene, hollow fiber membrane contactor, CO_2 capture, nanofluid, CO_2 absorption

1. Introduction

The amount of carbon dioxide (CO_2) in the atmosphere is increasing due to the use of fossil fuels in homes and industries. This has resulted in global warming which is now widely recognized [1]. By 2050, the concentration of CO_2 is expected to double if nothing is done [2]. CO_2 capture and storage (CCS) is seen to be a viable remedy for this problem. CCS has a lot of potential in various industries, including petroleum refineries. This is because many industrial processes produce gas streams that are rich in CO_2 or, in some cases, pure CO_2. This can help reduce the cost of CCS [3]. Potassium carbonate has a strong affinity for CO_2, which makes it a highly effective absorbent for capturing the gas at large scales. Due to its excellent absorption capacity, K_2CO_3 is an attractive option for CO_2 capture applications. Moreover, it is readily available and relatively inexpensive, making it a cost-effective alternative to other absorbents. Additionally, as

stated in reference Z. Pang et al. [4], K_2CO_3 solution can be easily replenished and the trapped CO_2 can be recovered without any significant difficulties. Potassium carbonate (K_2CO_3) can help adsorb carbon dioxide (CO_2), but its effectiveness depends on various factors such as temperature, pressure, solution concentration, and duration of gas-liquid contact. Generally, lower pressures and higher temperatures can improve the adsorption process. Additionally, studies have shown that higher concentrations of K_2CO_3 in the solution can increase CO_2 capture efficiency, as reported in reference T. Moore et al. and U. Kamran et al. [5, 6]. The carbon dioxide absorption properties of various amine absorbents such as N-methyl diethanolamine (MDEA), monoethanolamine (MEA), Triethanolamine (TEA), piperazine (PZ), diethylethanolamine (DEEA) and ethylethanolamine (EEA) tend to vary. Although MEA, which is a primary amine, absorbs less CO_2, it has a higher absorption rate compared to others [1]. However, the development of mixed absorbents has also advanced to meet the high CO_2 absorption and regeneration capability, which cannot be achieved by a single *absorbent*.

Studies have found that the efficiency of carbon capture can be increased by adding nanoparticles (NPs) together with potassium carbonate (K_2CO_3). NPs provide several benefits, such as increased surface area and catalytic effect, which can enhance the absorption capacity and kinetics of carbon dioxide (CO_2) capture. The larger surface area of NPs promotes the uptake of CO_2 by improving the interaction between the absorbent and CO_2 molecules. This has been reported in literature [8-10]. It has been discovered that the presence of nanoparticles (NPs) in a K_2CO_3 solution can help reduce mass transfer restrictions by facilitating CO_2 diffusion into the absorbent solution. This is due to the fact that NPs positively affect CO_2 solubility, which in turn lowers mass transfer resistance [7]. One of the primary advantages of using nanoparticles with potassium carbonate is that they can stabilize and prevent the absorbent solution from degrading, making it more durable and ensuring its long-term effectiveness. These findings were mentioned in a reference source [11]. It is imperative to recognize that the successful incorporation of nanoparticles (NPs) in carbon dioxide (CO_2) capture processes requires careful consideration of some factors, including the appropriate NPs' selection, their dispersion in the solution, and their consistency with K_2CO_3 [12]. The selection of NPs should be based on their adsorption characteristics, stability, and cost-effectiveness. Accurate methods of characterization must be used, and the concentration of nanoparticles must be optimized to achieve the desired increases in the efficiency of carbon capture, as previously demonstrated [11,15]. The cyclic organic compound piperazine can act as a promoter or activator in the process of CO_2 absorption, leading to an enhanced. Piperazine is a reactive solvent that helps in the reaction between carbon dioxide and potassium carbonate. This reaction produces potassium bicarbonate. Piperazine increases the rate of this reaction, making carbon dioxide assimilation more effective. As a result, higher rates of carbon dioxide sequestration can be achieved in shorter contact times [5].

Studies show that piperazine not only enhances CO_2 absorption, but it also reduces the breakdown of K_2CO_3 caused by side reactions or contaminants in the flue gas[7]. For the carbon capture system to function sustainably and be financially viable over an extended length of time, stability must be maintained. However, it is imperative to take into account the potential challenges associated with the application of piperazine [16,17].

One potential issue is the instability and vapor pressure that piperazine may exhibit, which could cause it to evaporate during the capture process. To minimize any piperazine losses and maximize its effectiveness, suitable control mechanisms and system design should be implemented as advised by the source, in addition to offering intriguing advantages over traditional absorption towers, non-dispersive absorption using a gas–liquid contactor may pose a threat to the employment of supported liquid membranes and dense membranes J. Liu et al. and P. Luis et al. [18,19].

The use of membrane-based absorption has greater operational flexibility than traditional absorption carried out in scrubbers because it allows for independent control of gas and liquid flow rates, a controlled and known interfacial area, and a linear scale-up made possible by the modularity of membrane contactors. Because of the low concentration of CO_2 in the gas stream, the mass transfer of CO_2 from the gas to the liquid phase has no discernible effect on the gas flow [20]. Furthermore, in contrast to other membrane systems, the mass transfer through the membrane should be preferred over the use of dense membranes or membranes with liquid-filled pores (supporting liquid membranes). This is because the membrane pores are (theoretically) filled with gas. Since the membrane's pores are where the gas and liquid phases come into contact, the membrane's pore size and porosity are important parameters to take into account [21–22].

Recently, the use of porous hollow fiber membranes for capturing acid gases has gained significant attention in recent times [2,23]. These membranes possess high permeability, low mass transfer resistance, and exceptional chemical resistance to various chemical feed streams. Gas-liquid membrane contactors allow fluids to be contacted on opposite sides of the membrane, creating a gas-liquid interface at the mouth of each membrane pore. Diffusion across the interface enables mass transfer. The purpose of this study is to advance a novel and effective carbon capture technique. To that aim, potassium carbonate (K_2CO_3) and MDEA were chosen as the primary absorbents. Piperazine and SiO_2 nanoparticles were then added to each to form a unique carbon sequestration blend. By introducing MEA absorbent solution into the PP hollow fiber membrane, a new mixed amine absorbing liquid MDEA-MEA was created. The main focus of the study will be on temperature manipulation, K_2CO_3 concentration, and gas and liquid flow rates. By looking at how these operational parameters affect the efficiency of carbon capture.

2. Materials and Methods

A cylindrical hollow fiber module was used as a membrane contactor to investigate the CO_2 absorption performance of aqueous K_2CO_3 and MDEA with nanoparticles and piperazine.

Table 1 lists all the features of the membrane module (PP) fibers from Parsian Pooya Polymer Co. The PP membrane exhibits good hydrophobicity when the pores are gas-filled and a solvent with high surface tension is used. Fig. 1 is a schematic of a hollow fiber membrane contactor.

Table 1. Specifications of the PP hollow fiber membrane contactor.

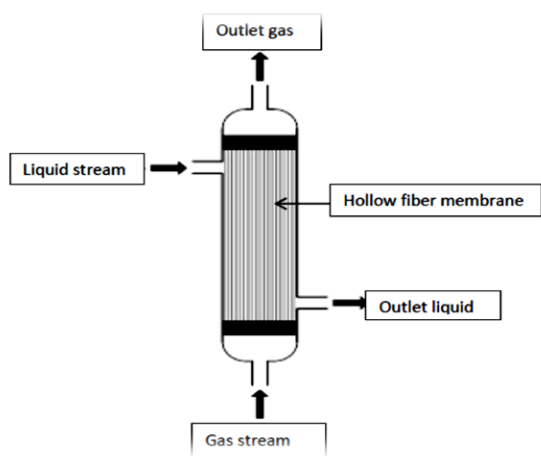| Value	Specifications
1.8	Fiber inner diameter (mm)
70.4	Membrane porosity (%)
0.4	Membrane wall thickness (mm)
25	Length of fiber (cm)
0.2	Membrane pore size (μm)

Fig.1. Schematic of ahollow fiber membrane contactor

2.1. Experimental set up

The experimental setup used in this study is shown in Fig. 2. The process involved the absorption of carbon dioxide using a PP membrane module. The setup consisted of a membrane contactor module with hollow fibers, as well as liquid and gas flow meters, regulators, pressure indicators, a temperature controller, a static mixer for gases, and a water bath. The solvent was pumped into the lumen side of the hollow fibers using a centrifugal pump.

To conduct the experiments, a gas mixture of N_2 and CO_2 was introduced into the system via compressed gas cylinders, which passed through a static mixer. The liquid was made to flow through the lumen side of the hollow fibers, which were arranged in a coiled tube structure and set up in a counter-current flow configuration. In the meantime, to make sure the temperature was controlled, the gas was directed toward the membrane module's shell after passing through a water bath.

The liquid side of the system must be kept at a little greater pressure than the gas side during the experiment to prevent a wetting issue, the pressure differential was 0.2 atm between the gas and liquid phases. By doing this, bubbles in

the mass transfer layer on the membrane surface would not form, ensuring a uniformly even mass transfer layer. The system runs for approximately 10 minutes to reached a steady state. The operation was carried out at atmospheric pressure.

To prepare the membrane for each run, the system was cleaned with distilled water to eliminate any leftover effects from the previous run. A minimum of 40 min of airflow drying ensued after this. Gas chromatography-TCD (Varian 3700) was used to examine the compositions of the gas streams entering and leaving the system. Each data point was generated by conducting the measurement at least three times and reporting the average of these values. Additional runs were performed if the results of the repeated measurements varied.

2.2. Preparation of liquid absorbent

In this study, Distilled water was used to dissolve 5% weight of (K_2CO_3 or MDEA and MEA) to create a base fluid, which was then mixed with a magnetic stirrer for 20 minutes at room temperature to create a homogeneous solution. Uses piperazine (PZ) powder of purity 99.5% at a concentrated 2% weight and is mixed into the base fluid after that. A magnetic stirrer is used to mix the suspension for ten minutes. SiO_2 nanoparticle was added to base fluid with concentration (0.05% wt.) at particle size 10nm stirred again by magnetic stirrer for 30 min to prepare nanofluid. Silica nanoparticles are hydrophilic, which means they do not need an acidic treatment to prevent clumping. The silica nanoparticles were sonicated with a water bath sonicator for 30 min to be dispersed in the base fluid in the sonicator (Tomy, ModelNo. UD 201, Japan).

3.3. Analysis methodology

The percentage of carbon dioxide removed must be calculated by comparing the amount of carbon dioxide removed to the original concentration of carbon dioxide. The
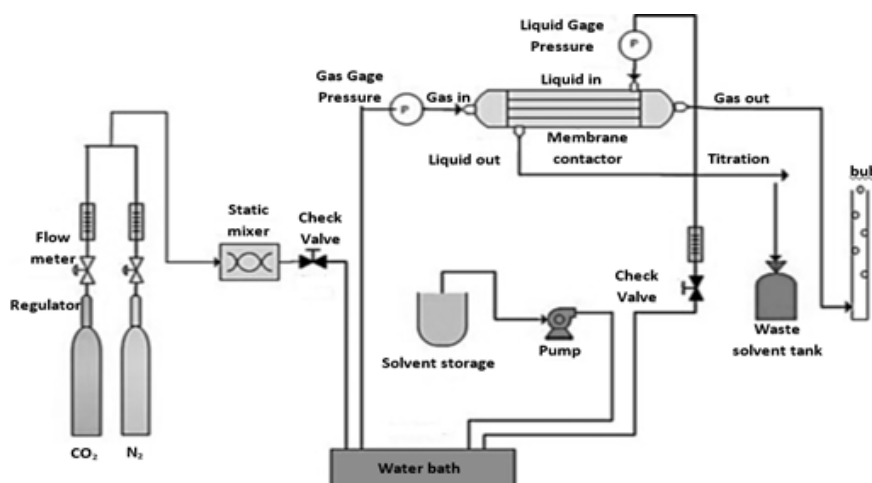

Fig. 2. The schematic diagram of the experimental apparatus

following formula was used to calculate the proportion of carbon dioxide removed:

$$Carbon\ Dioxide\ Removal\ \% =$$

$$\frac{(Initial\ CO2\ Concentration\ -\ Final\ CO2\ Concentration)}{Initial\ CO2\ Concentration} \times 100$$

3. Results and Discussion

3.1. Removal efficiency of nanofluids at various liquid flow rates

The chart in Fig. 3 shows how effective CO_2 removal is based on the liquid flow rate. The liquid flow rate ranged from 20 to 100 mL/min. The results indicate that the rate of CO2 separation increases as the liquid flow rate increases for all fluids. It was observed that the removal efficiency increased by 85% and 88% for (K_2CO_3) and (K_2CO_3/PZ/SiO_2) respectively, and by 76% and 80% for (MDEA) and (MDEA/PZ/SiO_2) respectively. As the liquid flow rate of the absorbent increases, the liquid velocity inside the shell side also increases, resulting in higher CO_2 removal efficiency. This happens as the liquid phase boundary layer at the membrane wall decreases, which in turn increases the diffusion of carbon dioxide. This led to an increase in the liquid mass transfer coefficient and an improvement in CO_2 absorption. K_2CO_3/SiO2/PZ nanofluids performed better than MDEA/SiO2/PZ nanofluids due to the higher absorption rate between CO_2 and K_2CO_3. SiO_2 with PZ to the solution increased its capacity to absorb CO_2, because the PZ leads to increased CO_2 loading, leading to an increase in the concentration gradient of CO_2 and absorption enhancement. Enhanced mass transfer in nanofluids is caused by the grazing effect, Brownian motion, and micro convection. Nanoparticles adsorb dissolved gas molecules in a thin film before releasing them into the liquid bulk as they pass through the concentration boundary layer due to the grazing effect. Additionally, nanoparticles induce micro convection via their Brownian motion, which can accelerate mass diffusion in the nanofluid. Similar results to this study were previously published by [25].
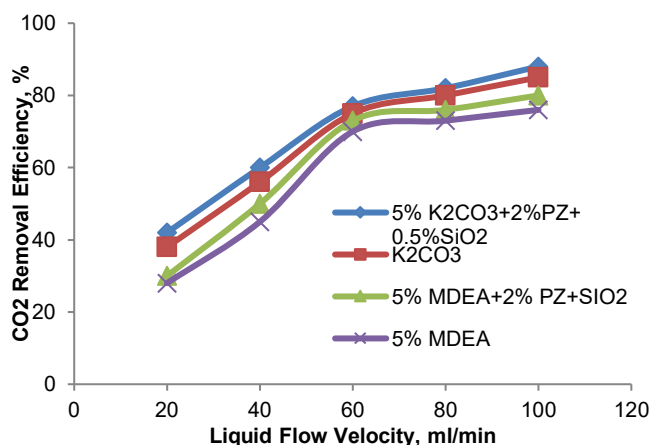

Fig. 3. Variation in The Effectiveness of CO_2 Removal Using Various Solutions and Liquid Flow Rates. Gas flow rate: 250 mL/min; CO_2 inlet concentration: 15%;Temperature: 303 K

3.2. Effects of gas flow rate on CO_2 removal

Fig. 4 illustrates the impact of the incoming gas flow rate on the efficiency of CO_2 removal. With an increase in gas volumetric flow rate from 130 ml/min to 530 ml/min, the efficiency of carbon dioxide removal decreased for all types of solutions. Specifically, the CO_2 removal efficiency decreased for K_2CO_3 from 68% to 45% and for MDEA from 63% to 38%. Furthermore, the efficiency of CO_2 removal was reduced for K_2CO_3/SiO_2/PZ from 80% to 58% and for MDEA/SiO_2/PZ from 70% to 50%.

Higher gas flow rates lead to reduced CO_2 absorption through the membrane into the solvent. This is because the residence time of CO_2 in the membrane contactor and the amount of CO_2 molecules that could penetrate through the membrane and be absorbed into the solvent are reduced with increased gas flow rates. As a result, the effectiveness of CO_2 removal significantly decreases as CO_2 flux rises. These findings align with references [27-31].
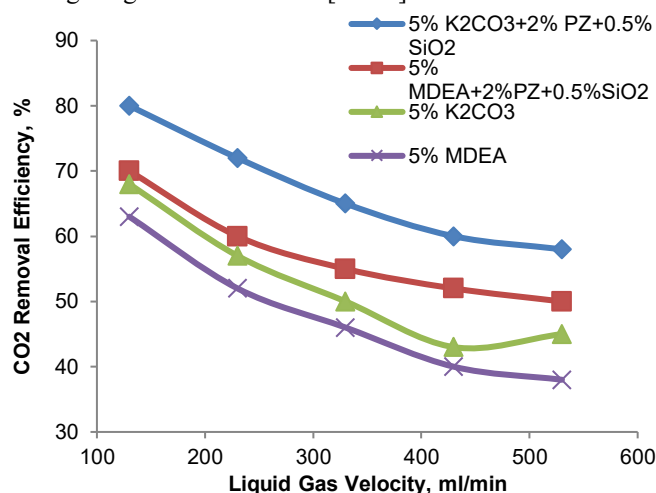

Fig. 4. Effect of Gas Flow Rate on The CO_2 Removal Efficiency of MDEA-PZ-SiO_2 and K_2CO_3-PZ-SiO_2 Nanofluids (Liquid flow rate=60 mL/min, CO_2 inlet conc.15%,T=303K)

3.3. Effects of liquid temperature

At various temperatures of 30, 35, 40, 45, and 50°C, Fig. 5 clearly illustrates how temperature (T) affects the effectiveness of CO_2 removal in a solvent reinforced by nanoparticles and PZ. There was a 250 ml/min gas volumetric flow rate and a 60 ml/min nanofluid volumetric flow rate. When the temperature rose from 30°C to 35°C, the removal efficiency for K_2CO_3/PZ/SiO_2 nanofluid improved from 70% to 77%. The rate of CO_2 removal with silica nanofluid drops to 68% when the liquid temperature is raised from 35 ºC to 50 ºC. Under the same circumstances, the MDEA nanofluid's efficiency drops from 65% to 55%. The performance of gas-liquid membrane contactors may suffer in certain ways with an increase in liquid temperature. For example, CO_2 becomes much less soluble in water as the temperature rises [32]. As temperature rises, CO_2 diffusivity increases, but so does the rate at which water evaporates, increasing the possibility that water will condense in membrane pores and preventing CO_2 diffusion [3]. According to the study's findings, temperature

has a more negative effect on the evaporation and solubility of CO_2 compared to its impact on diffusivity [32,33]. The aggregation of nanoparticles in absorption solutions and Brownian movement may be impacted by temperature changes. Higher suspension temperatures for nanofluids will result in stronger Brownian motion of suspended nanoparticles as well as more intense microconvection within the nanofluid, which further magnifies energy and mass transfer processes inside the nanofluid [35]. On the other hand, temperature can also affect the aggregation and Brownian movement of nanoparticles in absorption solutions. This manner is further passionate in a solution containing nanostructure due to the enhancement in heat transfer caused by nanoparticles [33,34].
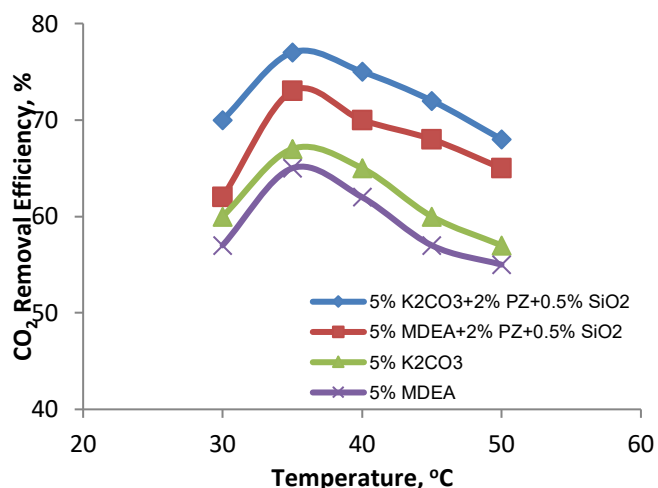

Fig. 5. Effect of liquid temperature on the CO_2 removal efficiency of MDEA-PZ-SiO$_2$ and K$_2$CO$_3$-PZ-SiO$_2$ nanofluids (Liquid flow rate=60 mL/min, Gas flow rate= 250 mL/min, CO_2 inlet conc.15%).

3.4. Effect of MDEA and K₂CO₃ concentrations

The findings depicted in Fig. 6 indicate that the CO_2 removal efficiency was found to increase from 48% to 85%, when K_2CO_3 and MDEA concentration increased from 3wt.% to 7wt.%. Similar results were observed for MDEA/PZ/SiO$_2$, where CO_2 Removal efficiency increased from 38% to 80% a K_2CO_3/PZ/SiO$_2$ shows even superior performance in terms of CO_2 removal efficiency. The fixed volumetric flow rates for gas and nanofluid were 250 ml/min and 60 ml/min, respectively. According to the findings, a higher concentration of both absorbents increases CO_2 loading, leading to an increase in removal efficiency. The solubility of CO_2 and the rate of diffusion slow down when the solvent concentration rises because of the increased viscosity of the solution [36-38]. The solvent's capacity to absorb and interact with CO_2 molecules is enhanced by an increase in K_2CO_3 concentration. As CO_2 enters the liquid and reacts with the corresponding solvent, the CO_2 concentration decreases in liquid-gas boundary layer. It enhances the CO_2 solubility rate and increases the CO_2 absorption. This event has the effect of improving the removal of CO_2 from the gaseous phase, which raises the capture efficacy overall. Increasing the concentration of solvent led to increase the viscosity of solution and result in low solubility of CO_2, low diffusion rate

of CO_2, and decreasing the removal efficiency. At high concentrations of absorbent, the liquid film resistance dominates the total mass transfer resistance, which in turn controls the overall reaction rate [39]. When SiO$_2$ NPs (silicon dioxide nanoparticles) and PZ (piperazine) are added, together with an increased concentration of K_2CO_3, the capture efficiency is increased. These additions contribute to improving the solvent's overall absorption properties or the kinetics of mass transfer [40,41].
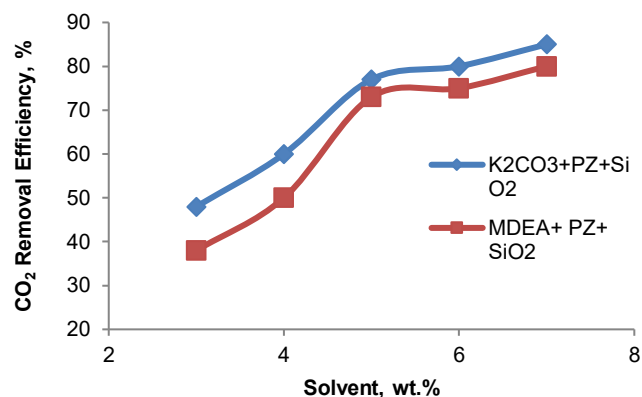

Fig. 6. Effect of K$_2$CO$_3$ & MDEA Concentration with 2%PZ and 0.05%SiO$_2$ on CO_2 Removal Efficiency (Liquid flow rate = 60 mL/min, Gas flow rate = 250 mL/min, CO_2 inlet conc.15%, T=303 K)

3.5 The effect of amine and mixed amine absorbent concentration

The three amine absorbents with mass concentrations of 3%, 5%, 7%, 9%, and 11% were prepared in 1Liter each, and the solute ratio of MDEA: MEA was 1:1, and the CO_2 removal efficiency on the hollow fiber membrane contactor was measured. The results are shown in Fig. 7. The comprehensive CO_2 removal efficiency of MEA and MDEA-MEA was 80.15% and 70.21%, respectively, which were higher than that of a single MDEA absorbent at this concentration. It can be seen that when the concentration is high (11%), the increase in the comprehensive removal efficiency of CO_2 of the mixed absorbent actually decreases, and the increase in decarbonization performance begins to slow down. This is because although the absorbent solute molecules at high concentrations are much larger than those at
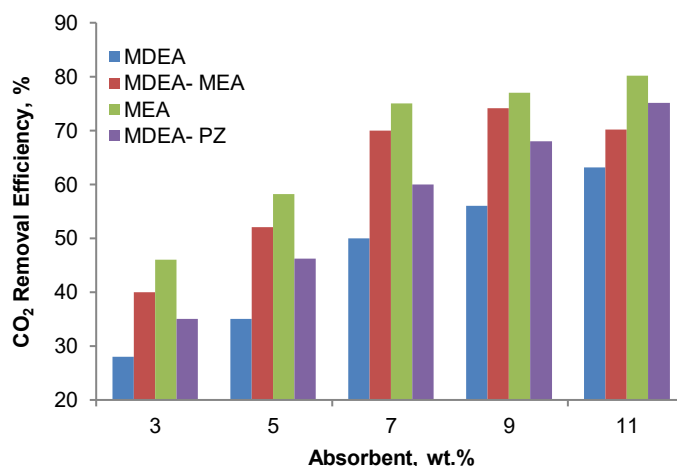

Fig. 7. CO_2 Absorption performance of amine absorbent with different concentration and comparison with single absorbent

low concentrations, the parameter mass transfer enhancement factor a directly reflects the strength of the liquid phase absorption reaction. It is foreseeable that if the concentration of the mixed solution is further increased, the efficiency of

their CO_2 membrane absorption will further decrease, so the reasonable concentration of the mixed amine absorption solution is between 9% and 10%.

Table 2. shows the previous studies related to the common use of nanoparticles and PZ for CO_2 absorption enhancement

Solvents	Process	Nanoparticle type	Concentration ofnanoparticle	Enhancement%	Reference
K_2CO_3/PZ	Bubble column	SiO_2	0.021%	12%	Kim et al., (2008)
MDEA	Membrane contactor	CNT	0. 5wt.%	13%	Yan et al., (2021)
K_2CO_3/PZ	Membrane contactor	SiO_2	0.05wt.%	88%	Present work
MDEA/PZ	Membrane contactor	SiO_2	0.05wt.%	80%	Present work

4. Conclusion

This paper applies the membrane technology method with strong application prospects and analyzes the current research status of CO_2 absorption by membrane technology. The experimental operating system has studied the CO_2 absorption performance of different absorbents experimental perspective. For mixed amine absorbents, appropriate addition of amine activators can effectively improve the CO_2 membrane absorption performance. The above research work, shows the effectiveness of carbon dioxide (CO_2) removal and the velocity of liquid flow (Q_L) are directly correlated, whereas the velocity of gas flow (Qg) and the efficiency of CO_2 removal are inversely correlated. This finding suggests that there is a range of flow rates present, which could improve CO_2 sequestration efficiency. Meanwhile, the addition of silica (SiO_2) nanoparticles at concentrations of 0.05% and 2%PZ was shown to have a substantial impact on the observed effectiveness of carbon dioxide (CO_2) removal. The investigation also revealed that 35°C was found to be the ideal temperature for CO_2 removal. The results also revealed that Potassium carbonate concentration directly impacts CO_2 removal efficiency; higher K_2CO_3 concentrations result in better operating efficiency. Finally, the CO_2 membrane absorption performance of the amine and mixed amine absorbents is in the order of MEA > MDEA-MEA > MDEA. When the concentration is higher, although the comprehensive removal rate of CO_2 in the mixed amine absorbing liquid is higher, the growth rate of the removal rate begins to slow down.

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