

Review of CO₂ Capture Using Absorption and Adsorption Technologies

Pashaei, Hassan; Ghaemi, Ahad⁺*

*School of Chemical, Petroleum and Gas Engineering, Iran University of Science and Technology,
Tehran, I.R. IRAN*

ABSTRACT: *In this work, we tried to explore the review of CO₂ capture using absorption and adsorption technologies. For this purpose, also, besides the literature review investigates the effect of some operational parameters such as temperature, pressure, amine concentration, solution flow rate, and the adsorbent loading on the CO₂ removal efficiency and CO₂ capture capacity. The results demonstrated that the flow rate of liquid and concentration of amine has a positive effect on the removal efficiency and the CO₂ flow rate has a negative effect. The results in the adsorption part indicated the pressure had a positive effect on the equilibrium adsorption capacity and the adsorbent loading and temperature had a negative effect. Moreover, the absorption of CO₂ into three ZrO₂, TiO₂, and ZnO nanofluids at Pz and DEA solution was investigated. The results represented that the loading of nanoparticles and speed of the stirrer have an optimum value for CO₂ removal efficiency. The optimum value was 0.05 wt%, 0.05 wt%, and 0.1 wt%, for ZrO₂, TiO₂, and ZnO nanoparticles, respectively. Furthermore, the CO₂ removal efficiency increased first with an increase of the stirring speed up to 200 rpm, and then begins to decrease as the stirrer speed increased above 200 rpm.*

KEYWORDS: *Absorption; Adsorption; CO₂ Capture; Nanoparticles; Adsorbent; Solvent.*

INTRODUCTION

According to the Intergovernmental Panel on Climate Change (IPCC), Global Greenhouse Gas (GHG) emissions must be reduced by 50 to 80 percent by 2050 to avoid the dramatic consequences of global warming [1, 2]. Scenarios from the International Energy Agency (IEA) indicate that the potential for reduced CO₂ emissions through enhanced energy efficiency and increased renewable energy production is limited. According to the IPCC, a delay in CO₂ emission reductions can lead to dramatic consequences, and a new strategy for reducing CO₂ emissions as soon as possible is required. CO₂ Capture is a technology with the potential for large reductions

in CO₂ emissions within 10 to 20 years. Therefore, the strategy for reducing global CO₂-emission must be a combination of (1) increased energy efficiency, (2) more renewable energy production, and (3) a wide implementation. By establishing stronger incentives favoring energy efficiency and renewable energy, global CO₂ emissions can be reduced by approximately 70 percent by 2050 compared to emissions today. Therefore, in the present study, we have tried to study all CO₂ emission methods and use the best method with optimal performance for different scenarios.

The recent importance of greenhouse gas emissions, and their potential for universal warming [3, 4], has appointed

* To whom correspondence should be addressed.

+ E-mail: aghaemi@iust.ac.ir

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concerns about the release of gases such as H_2S and CO_2 [5, 6]. With increasing environmental and political requests [7], impressive methods for CO_2 capture from the atmosphere will become increasingly significant [8-10]. Multitude types of processes produce CO_2 . A majority of these involve fossil fuel combustion, resulting in the emissions of greenhouse gases (e.g., H_2S , SO_x , NO_x , CO_2).

Given the extent of CO_2 emission processes into the atmosphere, it is crucial to recognize suitable goals for improvement [1, 11, 12]. Both human and natural sources contribute to the continuous emission of greenhouse gases [13]. While natural emissions from forest fires, volcanoes, and the decomposition of biomass are considerable, they are relatively fixed from year to year [14]. Man-made CO_2 released from automobiles, manufacturing, and power plants has increased steadily since the industrial revolution and is a contributing agent to universal warming and has become the main worry [15, 16]. Combustion of fossil fuel causes >95% of the CO_2 released annually [17]. The balance emanates from processes such as cement manufacturing, and steel, iron, and ammonia production [18].

Due to the significant value of emissions from the combustion of fossil fuel, it is effective for the analysis of this source. CO_2 emissions are represented in Fig. 1 for five point-source parts, including the agriculture, commercial and residential, electricity generation, industrial, and transportation sectors (EPA,2018).

A numerous range of processes has been extended to CO_2 removal from gas streams [19-21] including adsorption, absorption, cryogenics, and membranes [22, 23]. With membrane separation, high-purity streams are strict to get, exclusively on the scale of CO_2 removal from power plants. Membrane separation technology offered many advantages due to its inherent attributes such as energy-saving and continuous operation compared to traditional methods like absorption and adsorption processes [24]. Cryogenic separation of CO_2 causes liquid CO_2 flow, and high pressure. However, the refrigeration cost is expensive, and requires water removal, increasing the process cost. This technology is generally only intended for very concentrated CO_2 streams. Adsorption is one of the new concepts for CO_2 removal [25]. Depending on the conditions, multiple adsorbents have been used (Table 1).

Adsorption has been examined, but low CO_2 selectivity and poor capacity restrict the potential for usage to CO_2 removal [39]. Up to now, adsorption and absorption

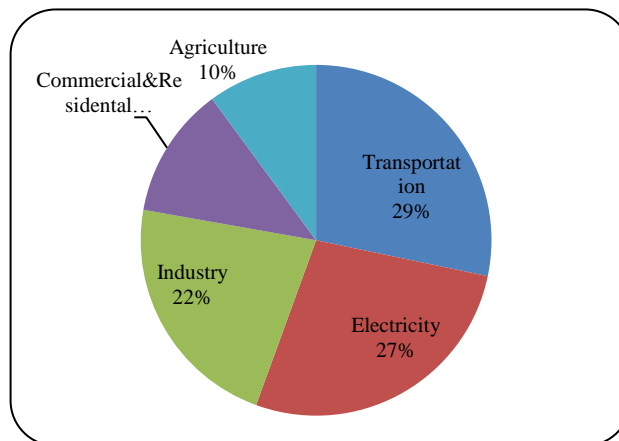


Fig. 1: CO_2 emissions from combustion of fossil fuel in the U.S.A.; (2018), total emissions: 6677 Tg CO_2 Eq.

methods provide the most economical to separate CO_2 from bulk flows [40]. That's why; this study focuses on the expansion of more effective adsorption and absorption technology [41]. The CO_2 absorption into physical [42] or chemical solvents [43] is a well-developed technology that has been used for many commercial processes [37, 44], including ammonia production and gas treatment [45, 46]. There has been a lot of research on this technology over the last 50 years, mainly on developing an understanding of the specific properties of the solvent. While a significant portion of work has been published on specific amines, little research has been outcome on comprehension or indicating complex mixtures, which are often the most effective technologies. The main purpose of this study was to explore the review of CO_2 capture using absorption and adsorption technologies [47-50]. For this purpose, we tried, in addition to literature reviewing, to investigate the effect of operational parameters such as temperature, flow rate, pressure, and the adsorbent loading on the CO_2 removal efficiency and CO_2 capture capacity. So, in the present work in addition to the effective operating parameters in both adsorption and absorption processes, the effect of nanoparticles on absorption has also been studied. Therefore, it can be a good referral for a comprehensive study of the absorption process.

RESULTS AND DISCUSSION

Carbon dioxide capture by absorption

One of the highest, and most investigated, acid gas removal technologies is an absorption process that uses a chemical solvent [51, 52]. Processes like this are usually

Table 1: CO₂ adsorption with different adsorbents.

Sorbent	Process	Temperature (K)	Capacity (mmol/g)	Ref.
NaOH	Fixed-bed reactor	298-338	2.82-29.75	[26]
Polyethyleneimine	Thermogravimetric adsorption	348	3.02	[27]
Monoethanolamine	Thermogravimetric adsorption	303	1.00	[28]
Graphene Oxide/TiO ₂ Nanocomposite	Fixed-bed reactor	298	0.77-0.95	[17]
AC/NaOH	Fixed-bed reactor	293-353	0.48-8.69	[29]
Molecular sieve 13X	Pressure swing adsorption	298	2.80-3.60	[30]
Active carbon-KOH	Pressure swing adsorption	298	-	[31]
NaOH-granular coconut shell AC	Fixed-bed reactor	332	27.10	[32]
NaOH/CaO	Temperature swing adsorption	350	3.00	[33]
Graphene	-	195	0.80	[34]
Na ₂ CO ₃	Thermogravimetric adsorption	323-343-70	2.30	[35]
Na ₂ CO ₃ /Al ₂ O ₃	Fixed-bed reactor	323-353	-	[36]
NaY zeolites/ Si/Al	Batch reactor	323	1.55-6.89	[37]
K ₂ CO ₃ /Al ₂ O ₃	Fixed-bed reactor	328-573	0.44-0.63	[38]

used in natural gas and ammonia production [53, 54]. There are multiple types of this flow sheet [55-58], including an isothermal and a temperature process. In many standard absorption processes, the variation of temperature swing (Fig. 2), a gas flow including CO₂ enters a column's bottom. The treated gas and CO₂ are released and exit the absorber's top. A solvent enters the top of the column and counter-currently contacts the gas phase in different absorbers such as packed column, tray column, and bubble column. The CO₂ removal efficiency of absorption was calculated Eq. (1) [59]:

$$\text{CO}_2\text{removal efficiency (\%)} = \quad (1)$$

$$\frac{Y_{\text{CO}_2,\text{in}} - Y_{\text{CO}_2,\text{out}}}{Y_{\text{CO}_2,\text{in}}} \times 100$$

Packed column apparatus

As shown in Fig. 3, the rich solvent, and the CO₂ are absorbed in the exit of the absorber. The rich solvent is heated in an exchanger and pumped to the top of a stripper. The CO₂ is recovered from a concentrated CO₂ flow. Somewhat heat is recovered from the lean solvent, although the solvent needs to be more cooled before its reuse in the column.

Depending on the specific usage of the process, different conditions occur. Table 2 represents some process conditions in absorber/stripper applications to CO₂ removal in a power plant setting. Generally, ammonia processing and natural gas treatment include high total pressures and concentrated CO₂ flow [60]. As well as, the outlet concentration limits are adjusted according to process requirements. Treating power plant flue gas will be the same in that CO₂ concentrations range from 10 to 15 vol% and 2 to 3 vol% for coal-fired plants and natural gas, respectively, but the pressure will be close to atmospheric. The treatment would goal about 90% removal.

Bubble column apparatus

Bubble column reactors (Fig. 4) have a vast range of industrial applications due to the easy design, selectivity for the favorable product, very involved hydrodynamic behavior, and well heat and mass transfer properties [61]. Bubble columns are applied in a variety of chemical processes, such as oxidation, alkylation, and fermentation reactions, Fischer-Tropsch synthesis, coal liquefaction, and effluent treatment [62, 63]. In the bubble reactors, the gas phase exists as a dispersed phase in a liquid phase; thus, the liquid phase's mixing behavior is usually influenced by the rising bubbles agitating action. In bubble

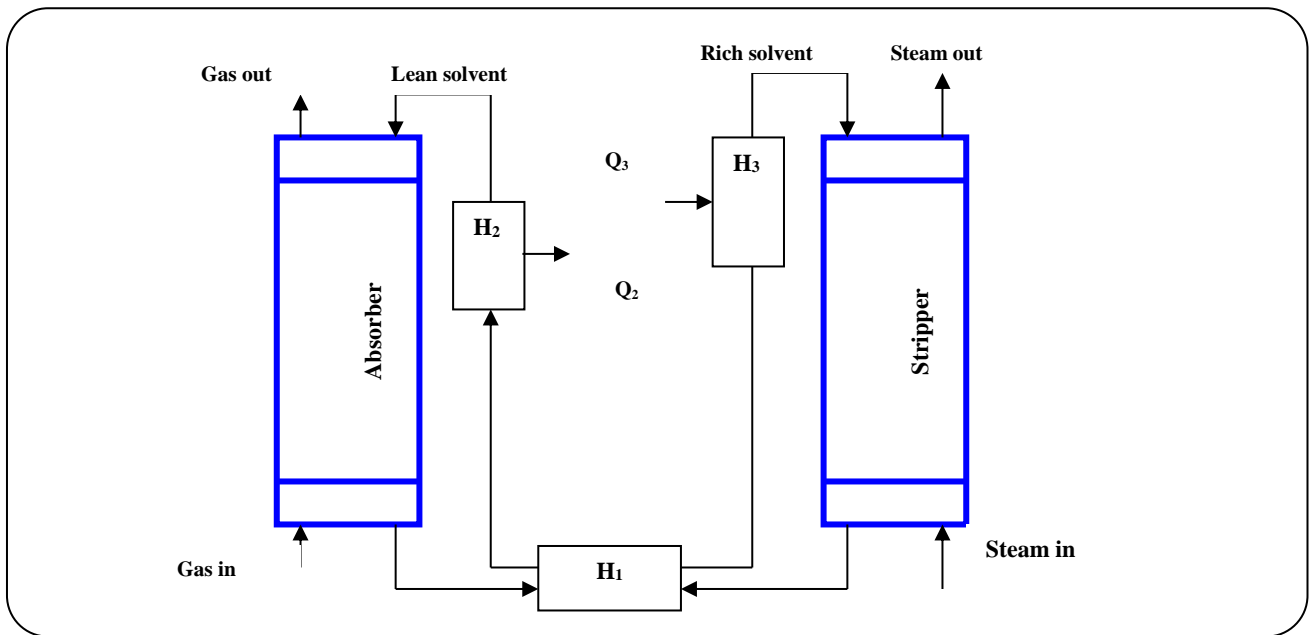


Fig. 2: Scheme diagram of absorber/stripper, H₁ (Cross Exchanger); H₂ (Cooler); H₃ (Exchanger); Q_i (Heat).

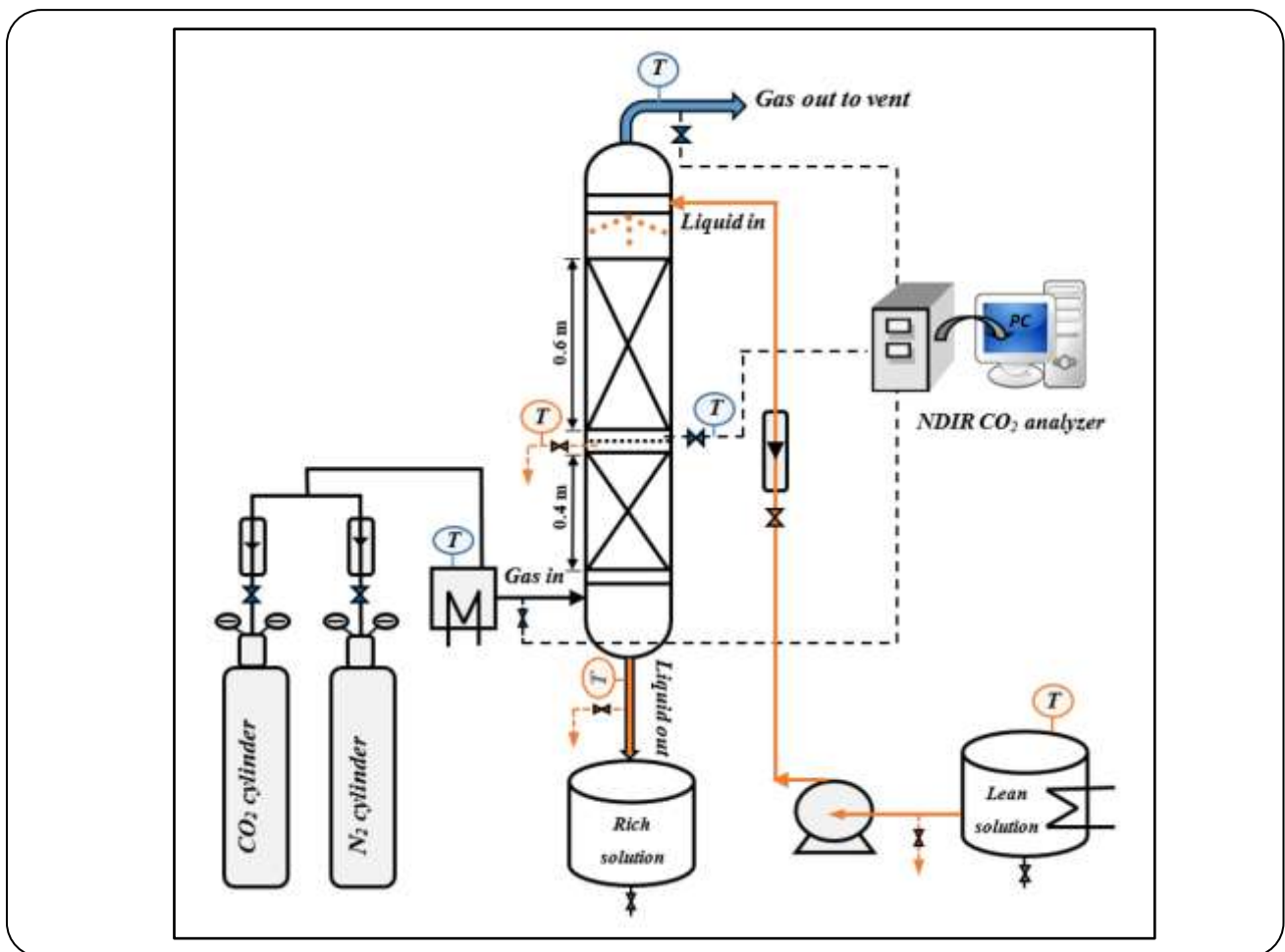
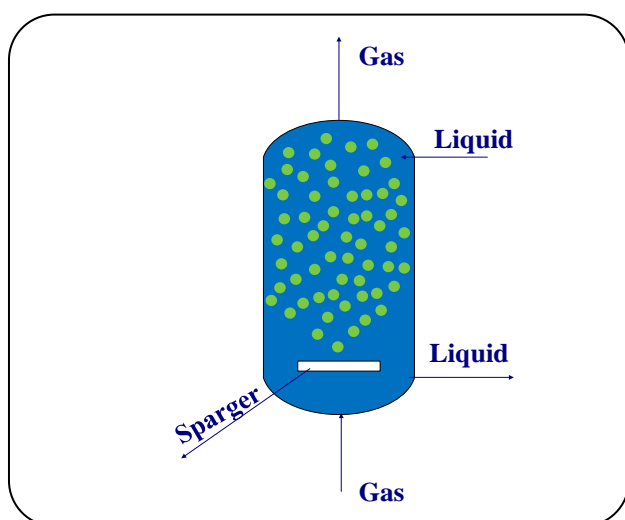


Fig. 3: Scheme diagram of packed column [57].

Table 2: Process conditions in absorber/stripper applications to CO₂ removal.

Process	Inlet CO ₂ (vol %)	Outlet CO ₂ (vol %)	P _{tot} (atm)
Natural gas	0-50	1-2	10-70
Ammonia	17-19	0.01-0.2	30
Coal power plant	10-15	1-1.5	1-1.3
Natural gas power plant	2-3	0.2-0.3	1-1.3

**Fig. 4: Scheme diagram of the bubble column.**

columns, the gas phase moves in one of the heterogeneous or homogeneous regimes, and dispersed gas is known to be inherently unsteady. A gas bubble's dispersion in liquids is the basis of many heats and mass transfer operations in chemical engineering action. The efficiency of the stirrer bubble column appertains to the physical bubbles characteristics such as bubble size, bubble size distribution, gas holdup, bubble–bubble interactions, bubble rise velocity, and mixing rate. Also, the economy of bubble columns and the design parameters are gas-liquid interfacial region, flow regime, bubble size distribution, mass transfer coefficient, and coalescence of bubbles. They are generally modeled by optimizing a plug-flow reactor with axial dispersion. The most traditional design of the bubble column prefers a high height-to-diameter ratio to allow system hydrodynamics to be in a fully developed state. Some prior work on CO₂ absorption has been published and is summarized in Table 3.

Solvents' effect on CO₂ absorption

Multitude solvents have been used to purify gas [78], but in general, the most effective is considered hot

potassium carbonate (hotpot) or aqueous amine solvents [79-81]. Amines have superiority over the hotpot process in that the absorption rate of CO₂ is fast; nevertheless [66, 81-83], the absorption heat is also high. On the contrary [61, 65, 84, 85], absorption into potassium carbonate has an absorption heat like to physical solvents but is confined by slow absorption rates. Usually, physical solvents are utilized in high-pressure usages. Some of the more common physical solvents are Purisol, Selexol, and Rectisol. Since physical solvents do not react with CO₂, the solvent is not consumed at high partial pressures [86, 87]. In addition, the absorption heat is restricted to the enthalpy of physical absorption [88], which is less than the reactive solvents [89, 90]. The processes are restricted by slow rates and selectivity of absorption [91].

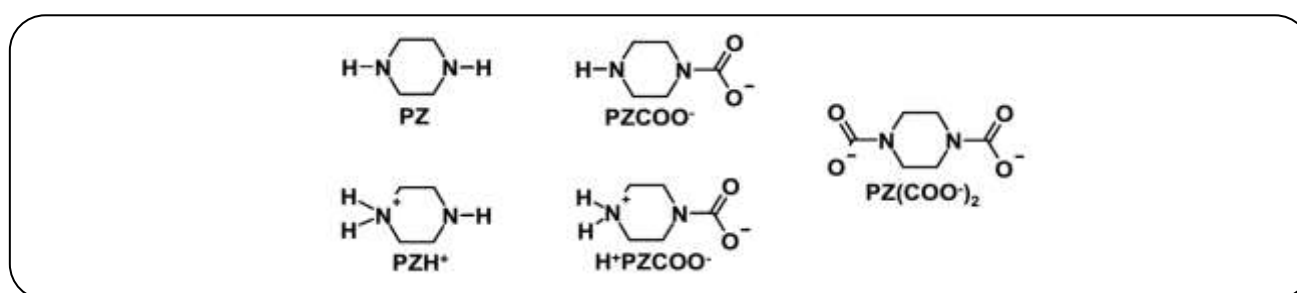
In CO₂ removal investigation from flue gas, the current advanced technology is commonly considered a 30 wt% aqueous MEA solution. MEA has a high absorption rate [91-94] and a high capacity for CO₂, but its performance is limited by multiple factors, containing corrosion issues and high absorption heat [95, 96]. One way to modify solvent performance is to combine amines or promote potassium carbonate with amines. The thought is to add a fast-reacting amine, such as MEA to a low absorption heat, such as MDEA or K₂CO₃, to take benefit of both properties. Most of this technology is used in the production of ammonia, where DEA-promoted K₂CO₃ is common.

Potassium Carbonate/Piperazine solution for CO₂ Capture

A new solvent, containing concentrated aqueous Pz and potassium carbonate, is used for the removal of CO₂ from waste gas streams. The Pz structure and its formatives in an aqueous solution with CO₂ are indicated in Fig. 5. Pz dicarbamate (Pz(COO⁻)₂) and Pz carbamate (PzCOO⁻) are products of the CO₂ reaction with Pz. protonated Piperazine carbamate (H⁺PzCOO⁻) and Protonated Piperazine (PzH⁺) are known stable molecules

Table 3: Studies on CO₂ absorption in bubble columns.

Solution	Description	Ref.
DEA	- The gas velocity, pressure, and liquid viscosity effect on gas-liquid interfacial area and gas holdup. - The positive effect of gas velocity on gas holdup - The positive effect of gas velocity and pressure on gas-liquid interfacial area	[64]
Pz/ ZnO, TiO ₂ and ZrO ₂ nanoparticles	- The nanoparticles effect on hydrodynamic, and absorption performance	[65]
Pz	- The effect of some parameter such as liquid viscosity, pressure, and gas velocity on hydrodynamic system. - Increasing gas holdup and gas-liquid interfacial area with increasing gas velocity, and pressure - The influences of absorbent and gas rate on absorption performance	[66]
Pz	- The effect of CO ₂ partial pressure, Pz concentration, temperature, and the stirrer speed upon the removal efficiency and absorption flux - Loading and removal efficiency of CO ₂ have been analyzed	[53]
Mg(OH) ₂	- Absorption kinetics has been analyzed - Offer model for mass transfer characteristics	[67]
Slurry of K ₂ CO ₃ and Li ₂ CO ₃	- The influence of slurry concentration, gas flow rate, and absorbent concentration on the absorption performance - The positive effect of absorbent and gas flow rate on the absorption performance	[68]
MDEA	- saturated the solution by increasing the gas flow rate - The positive effect of amine concentration and gas flow rate on the gas-liquid interfacial area - The initial concentration of amine to an optimum value has a positive effect on the mass transfer coefficient	[69]
TEA	- The effect of solution concentration and gas flow rate on the gas-liquid interfacial area and bubbles diameter	[70]
NaOH, Activated carbon and Alumina oxide	- The effect of solution concentration, solid loading, and gas flow rate on the gas-liquid interfacial area and mass transfer coefficient	[71]
Pyrrolidone, Methyl Pyrrolidone and Ethyl Pyrrolidone	- The effect of the solution concentration and gas velocity on the gas-liquid interfacial area and mass transfer coefficient	[72]
H ₂ O	- The influence of adding ethanol in CO ₂ absorption	[73]
DEEA/EEA	- The overall mass transfer coefficient, the absorption rate, and the determination of the optimum points	[74]
MEA/Al ₂ O ₃ , TiO ₂ , and SiO ₂ nanoparticles	- The effect of nanoparticles on hydrodynamic and absorption performance	[75]
DEA	- The effect of gas and liquid flow rate, initial concentration of amine, CO ₂ partial pressure on the removal efficiency, mass transfer flux and CO ₂ loading	[76]
Pz/ ZnO, TiO ₂ , and ZrO ₂ nanoparticles	- The nanoparticles effect on the hydrodynamic and mass transfer performance	[77]

Fig. 5: Structure of Pz in the presence of CO₂.

at moderate pH. A diprotonated Pz exists below a pH of approximately 5.5, but conditions in this work never close to low pH, so this species is out of view.

This solvent has multiple advantages over traditional amines [97]. First, due to the Pz being a diamine, the solvent can react with two CO₂ moles per one amine mole. Coupled with the potassium carbonate in the solution,

the solvent has the possibility of a higher CO₂ capacity than other amines. Also, the two functional groups of amines will appropriately affect the absorption rate. Second, the Pz has a high pK_a in comparison to MEA [98]. A high pK_a usually results in rapid absorption. Third, the high bicarbonate/carbonate value in the solution acts as a buffer, making less the protonation of the amine and making more

amine available to react with CO₂. However public studies of the Pz/K₂CO₃ solvent's performance are usual, and kinetics and thermodynamics detailed data are not accessible in the open literature [99]. And also, Pz/K₂CO₃ properties have not been previously investigated. Other work, although not specifically on Pz, is closely related to this research through modeling techniques and methods that should be mentioned. *Austgen et al.* [100] developed the rigorous thermodynamic model applied and used it to model DEA/MEA-promoted MDEA. *Glasscock* [101] began a study on the modeling of CO₂ absorption into MDEA/DEA and DEA. These researches displayed the multiple modeling techniques' ability to effectively represent amine combination over a wide range of conditions; however, none of them specifically show high ionic strength solvents.

Nanoparticles' effect on CO₂ capture

In recent years, different techniques have been applied to improve heat/mass transfer and get better absorption processes [102, 103]. The technique of these modifications is usually classified into mechanical purification, chemical treatment, and improvement of fluid properties [104-106]. Recently, nanofluids have been recognized as high-potential solvents [107] and significant mass transfer media [108]. A nanofluid is a suspension containing particles on a nano-scale that are constantly dispersed in a base fluid [109]. The nano-particle sizes are smaller than 100 nanometers. The nanoparticles are made of metals such as iron, nickel, silver, copper, and or metal oxides such as copper oxide (CuO), aluminum oxide (Al₂O₃), Fe₂O₃, FeO, ZrO₂, and TiO₂. Nanofluids having high dispersion stability are compared to base fluids and higher surface area and need lower pumping energy, and low sedimentation speed [110]. Thus, nanofluids can potentially be applied in a wide range of various usages. According to *Krishnamurthy et al.* [111] experiment, a color diffuses faster in a nanofluid than in water. They explained that the Brownian motion of the nanoparticles and shuttle effect causes convection in the nanofluids. CO₂ absorption is one of the most significant applications of nanofluids in gas absorption. Many studies have been performed to increase CO₂ absorption. *Pineda et al.* [112] considered the Al₂O₃/methanol and SiO₂/methanol nanoparticles' effect using a tray column on CO₂ absorption. Their results represented that the absorption capacity of the Al₂O₃ and

SiO₂ nanofluids increased up to 9.4% and 9.7%, respectively, at the 0.05 vol% nanoparticles loading. In another research, *Pineda et al.* [113, 114] investigated the TiO₂, Al₂O₃, and SiO₂ nanoparticle's effect on CO₂ absorption in an Annular Contactor (AC) with a trays column. The results of their research indicated that the absorption rate of TiO₂, Al₂O₃, and SiO₂ increased up to 5%, 6%, and 10%, respectively. *Lee et al.* [115] studied showed an increase in CO₂ absorption performance in a bubble column. The results showed that the absorption performance of the Al₂O₃ and SiO₂ nanofluids was enhanced up to 4.5% and 5.6%, respectively, at 0.01 vol% nanoparticles loading in comparison to methanol-based fluids. *Jung et al.* [116] studied the effect of Al₂O₃ nanoparticles in methanol-based fluids on CO₂ absorption rate using a bubble column. They indicated that the absorption performance increased to 8.3% in comparison to pure methanol. *Pang et al.* [117] experimentally researched the NH₃ absorption intensifies in the silver nanofluid during the bubble column. They found that the absorption rate was enhanced to 55% at 0.02 wt% nanoparticles loading. *Ma et al.* [118] and *Lee et al.* [119] studied the NH₃ heat and mass transfer in the absorption process using the Carbon NanoTube (CNT) and Al₂O₃ nanoparticles in a bubble column. Their results demonstrated the CNT and Al₂O₃ lead to increasing the mass and heat transfer rates significantly. *Sumin et al.* [120] experimentally studied the Al₂O₃ and CNT nanoparticles' influence on CO₂ absorption in a stirred thermostatic reactor. *Samadi et al.* [121] proposed a wetted-wall column with external magnets. They experimentally understood that the Fe₃O₄ nanofluids could increase the mass transfer flux and mass transfer coefficient to 22.35% and 59%, respectively. *Salimi et al.* [122] studied the CO₂ absorption in the NiO (at 0.01 vol %) and Fe₃O₄ (at 0.005 vol %) nanofluids in the presence of a magnetic field. They represented that the mass transfer rate of the NiO and Fe₃O₄ nanofluids was enhanced up to 9.5 wt% and 12 wt%, respectively, in comparison to based-fluids. *Kim et al.* [123] investigated the CO₂ absorption performance in the silica (at 0.021 wt%) nanofluid in a bubble column. The results displayed that the absorption rate in the nanofluids was enhanced 24 % in comparison to pure water-based fluids. After absorption of gas in the nanofluid, the contact area was increased because of broken gas bubbles into small bubbles with stable nanoparticles and finally led

Table 4: Some studies of used various nanoparticles for CO₂ absorption.

Absorbent-Solution	System	Remarks	Ref.
MDEA- Magnetite nanoparticles	Wetted wall column	- Surrender the effects of the input area - Removing creeping resistance by adding surfactant - Absorption increased with the solid concentration up to 0.02 vol%.	[124]
Al ₂ O ₃ nanoparticles	Stirred reactor	-The absorption rate and heat transfer rate in nanofluid containing 0.02 vol% particles were found to be 18% and 29% higher than those base fluid, respectively.	[119]
2-ethyl-1-hexanol, noctanol and 2-octanol, Cu, CuO, and Al ₂ O ₃	Bubble column	-The appending of nanoparticles and surfactants enhances the absorption performance up to 5.32 times.	[125]
Methanol, SiO ₂ , and Al ₂ O ₃ nanoparticles	Bubble column	-by adding 0.01 vol% of Al ₂ O ₃ and SiO ₂ nanoparticles, the absorption rate is increased up to 4.5% and 5.6%, respectively, in comparison to methanol-based fluids.	[115]
Methanol, Al ₂ O ₃ and SiO ₂ nanoparticles	tray column	-The maximum absorption rate is enhanced up to 9.4% and 9.7% at 0.05 vol% of Al ₂ O ₃ and SiO ₂ nanoparticles, respectively, in comparison to methanol-based fluids.	[112]
MEA, SiO, and Al ₂ O ₃ nanoparticles	Bubble column	-The absorption rate is enhanced up to 8% compared to the pure MEA solution.	[126]
CNT and Al ₂ O ₃ nanoparticles	Stirred reactor	-The absorption rate is weak and enhanced. - The stirring speed has a positive effect on the enhancement factor in CNT nanofluid.	[120]
MEA, MDEA, SiO ₂ , TiO ₂ , Al ₂ O ₃ , and MgO nanoparticles	Bubble column	-The performance of MDEA and MEA solutions was similar. -Most nanoparticles could remarkably improve the mass transfer. -The enhancement factor increased with the increase of the nanoparticles concentration up to an optimum value and then decreased.	[127]
NaCl, and Al ₂ O ₃ nanoparticles	Bubble column	- The optimum nanoparticles loading of Al ₂ O ₃ was 0.01 vol%. -The nanoparticles' consequence on the enhancement factor is more considerable in the unsaturated state area than in the saturated state area.	[128]
Methanol, and Al ₂ O ₃ , nanoparticles	Bubble column	-The absorption rate is enhanced up to ~8.3%.	[129]
ZnO, and SiO ₂ nanoparticles	Stirred reactor	-The absorption rate is increased up to 14% and 7% at 0.1 wt% of ZnO and SiO ₂ nanoparticles, respectively. - The absorption performance decreases with increasing temperature.	[130]
Methanol, Al ₂ O ₃ , SiO ₂ and TiO ₂ nanoparticles	tray column	-The absorption performance is increased up to 10%, 6%, and 5% of Al ₂ O ₃ , SiO ₂ and TiO ₂ nanoparticles, respectively.	[113]
Fe ₃ O ₄ and NiO nanoparticles	packed column	-The mass transfer rate is increased up to 12% and 9.5% of Fe ₃ O ₄ and NiO, respectively. - The optimum nanoparticles loading of Fe ₃ O ₄ and NiO were 0.005 wt%, and 1.5 wt%, respectively	[122]
Pz, TiO ₂ , ZnO, and ZrO ₂ nanoparticles	Bubble column	- The optimum nanoparticles loading of TiO ₂ , ZnO, and ZrO ₂ were 0.01 wt%, 0.05 wt% and 0.01 wt%, respectively. -The effect of nanoparticles loading, nanoparticle type, CO ₂ partial pressure, and stirrer speed on the absorption and hydrodynamic performance were investigated. -The absorption rate of TiO ₂ , ZnO, and ZrO ₂ is enhanced up to 14.7%, 16.6%, and 3.7%, respectively.	[77]
MDEA, Pz- SiO ₂ nanoparticles	Stirred reactor	- Absorption rate increased with the solid concentration up to 0.02 vol%. - The optimum nanoparticle loading was 0.001 vol%.	[131]

to an increased absorption rate in the nanofluid. Some open literature has been available on the effect of nanofluids to increase CO₂ absorption, as shown in Table 4.

Fig. 6 indicates the variation of CO₂ removal efficiency with the loading of ZnO, TiO₂, and ZrO₂ nanoparticles in the Pz and DEA base fluid. All experiments were performed in a continuous bubble column reactor. As shown in this figure; the removal efficiency of all nanofluids is higher than 57.1%; it is certainly implied that nanofluids can increase the absorption rate. It can also be concluded

from the trend that the removal efficiency increases with increasing particle concentration up to 0.05 wt% of ZnO and TiO₂ nanoparticles and then reduced. In comparison, the removal efficiency increased with the increase of ZnO nanoparticles loading in all-solid concentration. The maximum removal efficiency for Pz/ZnO nanofluid (at 0.1 wt %) is 80.6. In the other words, the average CO₂ removal efficiency in the base fluid and optimum concentration (at 0.05 wt %) of TiO₂, ZnO, and ZrO₂ nanofluids was carried out 4.6%, 65.3%, 65.6%, and 60.8%, respectively. These results mean that the CO₂ removal

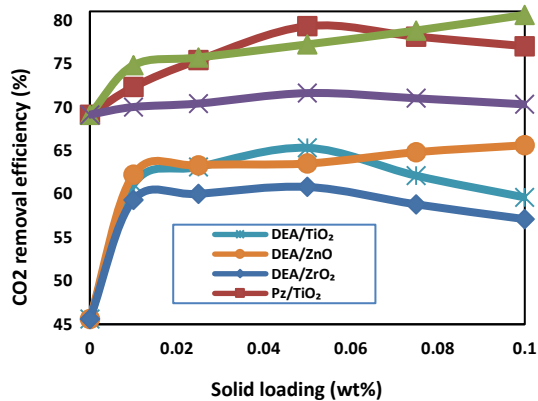


Fig. 6: Variation of CO₂ removal efficiency vs. the nanoparticles loading in nanofluids.

efficiency of DEA/ZnO, DEA/TiO₂, and DEA/ZrO₂ nanofluids in comparison with DEA base fluid was increased by 43.8%, 43.2%, and 33.3 %, respectively. It is undoubted that nanoparticles can increase absorption performance. The nanofluid mechanism increases the CO₂ removal efficiency may be for three reasons as follows. Due to the Brownian motion, nanoparticles can cause micro-convection in nanofluids. This micro-convection can improve the CO₂ absorption performance of the nanofluids. Nanoparticles in the nanofluids can cause the grazing (or shuttle) effect. However, when the nanoparticles number in the solution is numerous, the nanoparticles accumulate together that caused a reduction in the Brownian motion effect. Besides, with more increase in nanoparticles, the viscosity of nanofluid increased and caused a decrease in the diffusion coefficient of CO₂ and eventually decreased the CO₂ removal efficiency.

Fig. 7 presents the variation of CO₂ removal efficiency with the stirrer speed in different nanofluids (at 0.05 wt%). As shown trends in this Figure; the removal efficiency increases with an increase in the stirrer speed up to 200 rpm because of broken bubbles to small bubbles and increasing contact area, and then starts to decrease by increasing above 200 rpm. The increasing and decreasing reasons for the removal efficiency by the stirrer speed are explained in detail in the previous work [65, 66, 87]. By increasing in stirring speed above 200 rpm due to the broken of the bubbles and formation of coalescence phenomenon and accumulation bubbles in the center of the column, increasing in stirring speed had a reverse effect. When the stirrer speed was zero, removal efficiency for ZnO, TiO₂, and ZrO₂ nanofluids was 58.5%, 61.7%, and 55.5%,

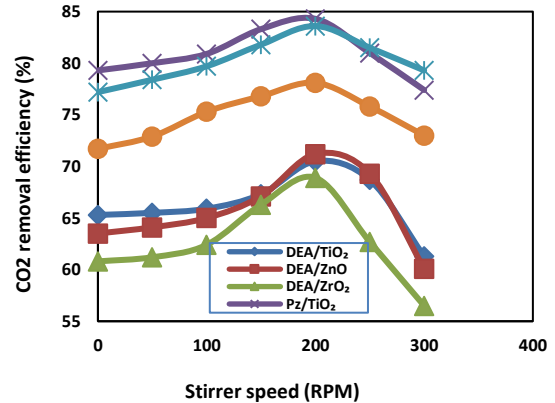


Fig. 7: The stirrer speed effect on CO₂ removal efficiency in various nanofluids.

respectively. The highest removal efficiency in ZnO, TiO₂, and ZrO₂ nanofluids was about 71.2%, 70.5%, and 68.9%, respectively, at 200 rpm.

Amine concentration effect on CO₂ absorption

Fig. 8 presents the variations of CO₂ removal efficiency in different Pz and DEA nanofluids. All trends showed at 15 min of the beginning time, the removal efficiency is high, and the slope changes are great. Nevertheless, after that time, the decline is small. There are two reasons for this tendency: the first, at the beginning of the process, CO₂ concentration is high and causes the contact time at the unit volume of gas-liquid to get lesser in the column. Second, increasing the volume fraction of CO₂ causes an increase in the reaction rate of CO₂ absorption at the specified amount. These two agents make CO₂ removal efficiency slowly reduced over time. Also, as shown in Fig. 8, the CO₂ removal efficiency performance at a high concentration of amine is better than that low concentration for both Pz and DEA solutions. In other words, increasing the concentration of amine in both Pz and DEA solutions has a positive effect.

Effect of liquid flow rate on CO₂ absorption

In this section, the volume flow rate effect of the absorbent on the CO₂ removal efficiency was examined. In Figure 9, the CO₂ removal efficiency profiles are drawn versus different solution flow rates. By increasing the liquid flow rate, CO₂ removal efficiency increases. With the increase of liquid flow rate, the liquid mass transfer resistance and contact time are decreased, and this gives a higher speed of CO₂ diffusing into the liquid.

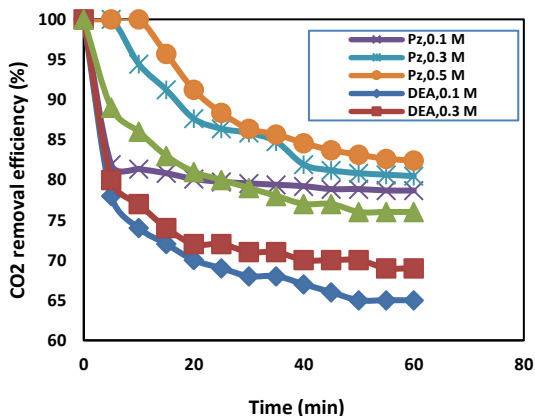


Fig. 8: Variations of CO₂ removal efficiency at the process and the effect of amine concentration.

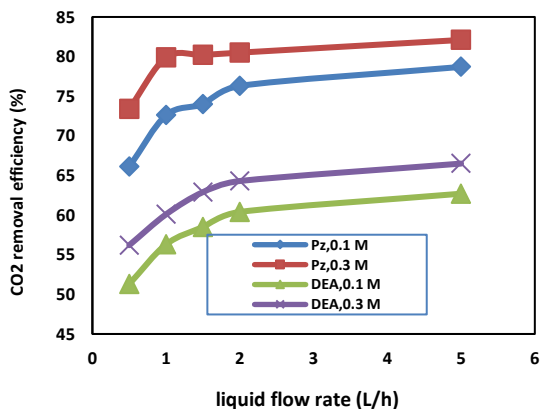


Fig. 9: The effect of solution flow rate on CO₂ removal efficiency.

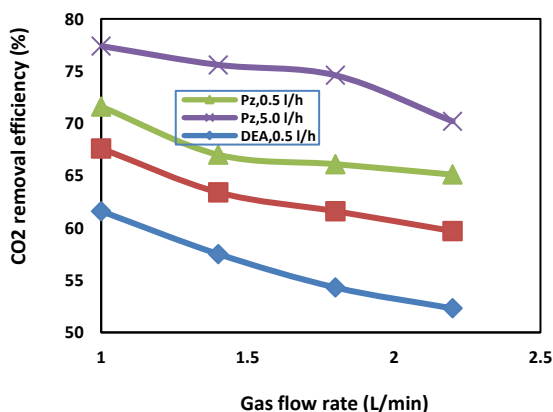


Fig. 10: Variations of CO₂ removal efficiency with the gas flow rate.

Effect of gas flow rate on CO₂ absorption

Fig. 10 displays the profiles of the gas flow rate effect on CO₂ removal efficiency at the different solutions of flow rates. Figure 10 demonstrated that the gas flow rate

has a considerable influence on CO₂ removal efficiency. It was evident that CO₂ removal efficiency had decreased with increasing the gas flow rate. By increasing the gas flow rate, the CO₂ that remained in the solution increased. So, the reaction between the limited quantity of free amines and an excess amount of CO₂ molecules resulted in the CO₂ removal efficiency decreasing. Another reason to decrease removal efficiency is to reduce the contact time at high gas flow rates.

Effect of Temperature on CO₂ absorption

Effects of Pz concentration, CO₂ partial pressure, and temperature on absorption efficiency are presented in Fig. 11. As seen in the figure, in a constant Pz concentration, CO₂ partial pressure, and stirrer speed, the removal efficiency reduces with increasing temperature. It is known that when temperature increases, equilibrium absorption capacity decreases according to the exothermic nature of reactive absorption, and the reaction rate increases according to the Arrhenius equation. Hence, a decrease in absorption efficiency for aqueous Pz solution with temperature seemed to be more affected by a reduction in absorption capacity than an increase in reaction rate. Fig. 11 shows two apparent issues, too. First, when temperature and amine concentration are fixed, an increase in the partial pressure of CO₂ causes a decrease in removal efficiency. Secondly, in a constant temperature, and CO₂ partial pressure the removal efficiency is increased with increasing the Pz concentration.

CARBON DIOXIDE CAPTURE BY ADSORPTION

Carbon dioxide adsorption using amine-modified adsorbents involves a chemical reaction between amino groups and CO₂ [132, 133]. The Pz is an alkanol amine; its reaction with CO₂ can be explained according to the zwitterions mechanism [134]. This mechanism includes the reaction of CO₂ with Pz to form an ionic middle compound of zwitterion (PzH⁺COO⁻) [135]. Then a proton is separated from it by an alkali such as Pz, PzH⁺, or PzCOO⁻ in the reaction environment, and Pz carbamate (PzCOO⁻) and protonate Pz (PzH⁺) are produced. This mechanism is shown in Figure 12.

The adsorption percentage of adsorbent was calculated using Eq. (2) [26]:

$$\text{Adsorption}(\%) = \frac{P_i - P_e}{P_i} \times 100 \quad (2)$$

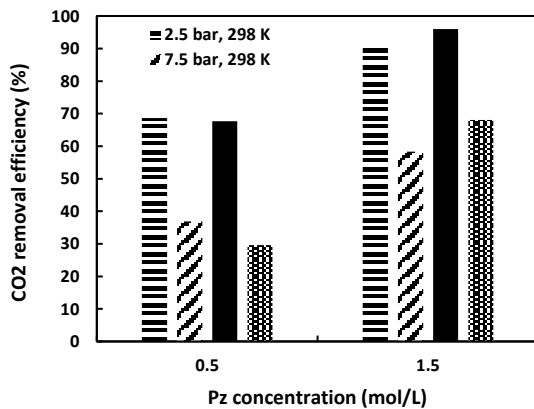


Fig. 11: Effects of Pz concentration, CO₂ partial pressure, and temperature on absorption efficiency.

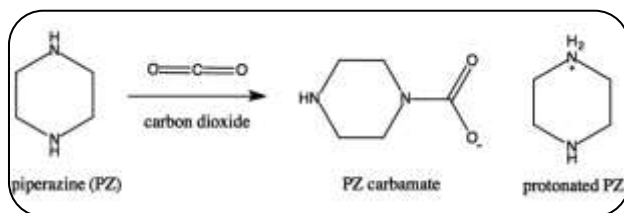


Fig. 12: Piperazine reaction with CO₂ [91].

The adsorption capacity of the adsorbent was calculated with Eq. (3) [26]:

$$q_e \text{ (mg/g)} = \frac{(P_i - P_e)VM_{CO_2}}{RTm} \times 1000 \quad (3)$$

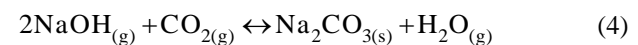
Effect of adsorbent amount

Saeidi *et al.* [26], Fashi *et al.* [91], and Karbalaei *et al.* [29] studied results on NaOH [136, 137], and AC sorbent for CO₂ capture from the air on the batch reactor is indicated in Figs. 13 (A) and 13 (B). As shown, adsorbent dosage increases, keeping all the other parameters at constant CO₂ adsorption capacity decreases, which are shown in Fig. 13. At lower adsorbent dosage, the numbers of active sites are higher. With the increase in adsorbent dosage, aggregation of particles takes place; consequently, the available adsorption sites may decrease; as a result, CO₂ adsorption capacity decreases. This means that the adsorbent dosage has a negative effect on CO₂ adsorption capacity.

Temperature effect on CO₂ adsorption

The effect of different temperatures on the CO₂ adsorption capacity for Activated Alumina (AA) and AA/NaOH [138], AC/NaOH [29], NaOH [26], and AA/Pz [91] adsorbents are shown in Fig. 14. As can be seen from

the trend to AA, AA/NaOH, AA/Pz, and AC, the increase in temperature from 293 to 353 K, reducing the CO₂ adsorption capacity for the adsorbents, and the maximum value of CO₂ adsorption capacity was observed at 293 K. Also the result indicates that the CO₂ adsorption process is highly exothermic, and CO₂ adsorption capacity decreases by increasing the temperature. However, a trend in the NaOH indicates that the optimum temperature for CO₂ removal is 308 K. To describe the perceived trend in this Figure, it can be noted that below 308 K, an increase in temperature leads to an increase in the carbonation reactions (i.e., forward reactions in Equations (4) and (5) [139]). Nevertheless, above 308 K, the reverse NaHCO₃ decomposition reaction becomes thermodynamically favorable and slows down the overall carbonation reaction. Consequently, the optimum temperature for the present CO₂ removal system is 308 K. also; Fig. 14 indicates that NaOH has better CO₂ adsorption capacity than other adsorbents [140, 141]. There are two main steps for this reason. First, NaOH loading leads to an increase in CO₂ removal rate due to direct growth in the reactant amount in the reaction system along with an increase in the gas-solid contact surface. The second is the pH effect. In the beginning, CO₂ gas instantly reacts with hydroxide ions (OH⁻) and is converted to carbonate (CO₃²⁻). With increasing the solution conversion, the concentrations of [Amine]H⁺ and HCO⁻³ rise, this can be described *via* a base catalyst mechanism. At the lower conversion, the carbamate is formed rapidly and then decreases slowly at the higher conversion. When the pH value becomes low sufficient, the Amine conversion to HCO⁻³ becomes higher and postpones the formation of [Amine]COO⁻. Therefore due to the conversion of Amine to Amine carbamate and protonated Amine, the concentration of free Amine becomes low but the value of Amine is still important for the absorption of CO₂.



$$\Delta H_{298\text{K}} = -127.5 \text{ kJ}$$



$$\Delta H_{298\text{K}} = -131.5 \text{ kJ}$$

Pressure effect on CO₂ adsorption

The effects of different adsorption pressures including 2, 4, 6, and 8 bar, on CO₂ adsorption by activated alumina (AA) and AA/NaOH [138], AC/NaOH [29], NaOH [26], and AA/Pz [91] adsorbents, are presented in Fig. 15. The adsorption capacity at 2 bar is low, and it gradually

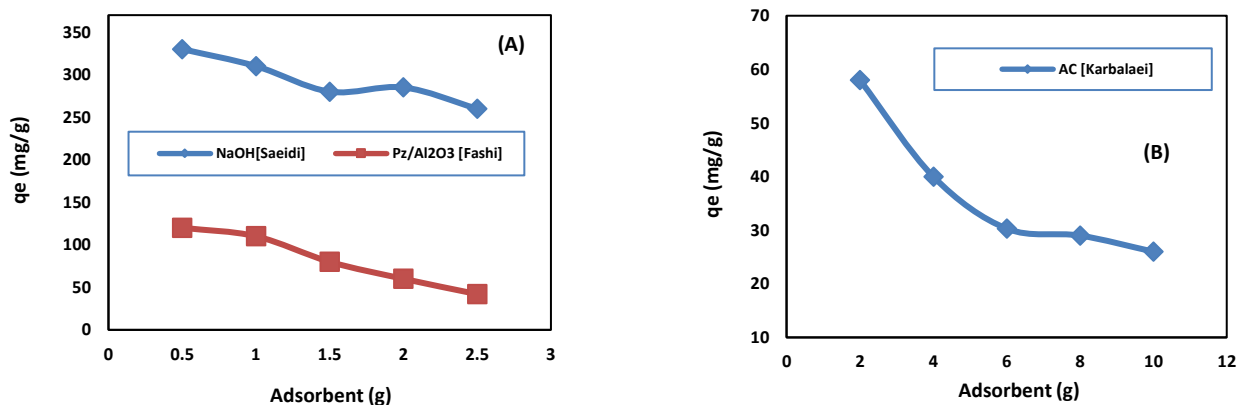


Fig. 13: Effect of the adsorbent on CO₂ adsorption capacity.

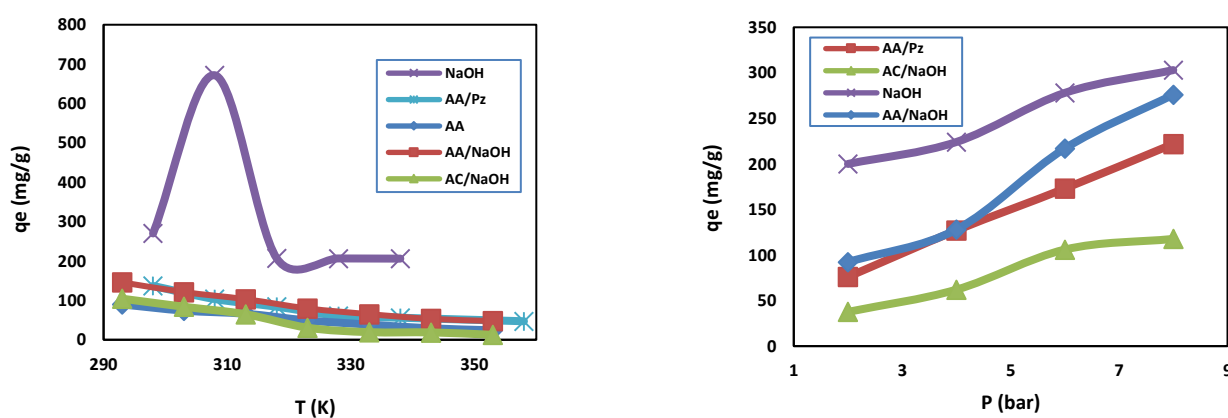


Fig. 14: Effect of temperature on CO₂ adsorption capacity.

Fig. 15: Effect of pressure on CO₂ adsorption capacity.

increases with increasing pressure from 2 to 8 bar. Some graph such as AC/NaOH shows that maximum adsorption capacity was carried out at 6 bar and after this pressure increasing pressure, there is no impressive effect on CO₂ adsorption capacity. Nevertheless, the maximum CO₂ adsorption capacity for all adsorbents is near the pressure of 8 bar.

CONCLUSIONS

This research consists of two absorption and adsorption parts. In the absorption section, it was found that data for the CO₂-Pz-H₂O and CO₂-DEA-H₂O system, the CO₂ removal efficiency increased with an increase in the liquid flow rate and amine concentration and decreased with an increase in the CO₂ flow rate. The CO₂ removal efficiency increases with the solid loading of TiO₂ and ZrO₂ nanoparticle up to a maximum value and then decreases. But, the removal efficiency of ZnO nanoparticles increased with the increase in the solid loading. It was observed that the effect of stirrer speeds up to 200 rpm is positive on the CO₂ removal

efficiency. In the adsorption section, CO₂ capture by some adsorbents such as AA, AA/NaOH, AA/NaOH, AA/Pz, and NaOH was evaluated. The effects of numerous operating parameters, such as adsorbent loading, temperature, and pressure on the CO₂ removal rate were considered. It was found that CO₂ adsorption capacity is in direct and indirect proportion to the pressure and temperature, respectively, which enhances CO₂ adsorption capacity by increasing pressure and decreasing the temperature. This study has also shown that NaOH has a better CO₂ adsorption capacity than other adsorbents due to their high pH. The analysis data of NaOH is important and can be deliberate as a baseline for a comparison with the created NaOH in the future.

Nomenclature

AA	Activated alumina
AC	Activated Carbone
Abbr.	Abbreviation
AMP	Isobutanolamine

DEA	Diethanolamine
DGA	Diglycolamine
DIPA	Diisopropanolamine
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
m	Mass of adsorbent, g
M_{CO_2}	Molar mass of carbon dioxide, g/mol
P_i	Initial pressure, bar
P_e	Equilibrium pressure, bar
Pz	Piperazine
q_e	Equilibrium adsorption capacity, mg/g
R	Universal gas constant, 8.314 J mol/K
T	Temperature of the reactor, K
TEA	Triethanolamine
V	Volume of the reactor occupied by the CO ₂ gas, mL
y_{CO_2-in}	Mole fraction of inlet CO ₂
y_{CO_2-out}	Mole fraction of outlet CO ₂

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