RESEARCH ARTICLE



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Insight into CO₂ capture by aqueous solutions of N,N-diethylethanolamine promoted with potassium salts of amino acids

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Abstract

N,N-Diethylethanolamine (DEEA) is a potential high-capacity CO₂-capturing solvent. The CO₂ reactivity of DEEA can be improved by the addition of rate promoters. Both equilibrium CO₂ solubility and desorption rate are also influenced by promoters. In this work, the effect of promotion of DEEA with three amino acid salts, potassium arginate (PA), potassium prolinate (PP), and potassium glycinate (PG), was investigated. In a stirred cell reactor, CO2 reactivity of the promoted solutions was studied at 303 K. Rate promotion with PA was most effective; this was then followed by PP and PG. The value of the liquid-side mass transfer coefficient (0.005 cm/s) for CO₂ absorption in water inside the stirred cell was found. Equilibrium CO₂ solubility in the promoted mixtures was measured. Empirical equations that predicted solubility data (accuracy 99%) were proposed. Desorption trials were performed at 363 K. PA, PP, and PG lowered sensible energy constraint by 59%, 32%, and 30%. PA was most-suited for faster desorption of aqueous solutions of DEEA. Overall, potassium salts of arginine, proline, and glycine were promising candidates for improving the performance of the tertiary amine DEEA. Finally, catalytic desorption of loaded solutions of DEEA was studied and it was found that alumina was a promising catalyst for faster desorption.

KEYWORDS

absorption, carbon dioxide, desorption, N,N-diethylethanolamine, solubility

1 | INTRODUCTION

CO₂ capture from the flue gas of coal-fired power plants is crucial. Absorption, adsorption, and membrane separation are candidate options for post-combustion carbon capture technologies in power plants. Especially, absorption in chemical solvents is the most mature and popular option. Additionally, this technique is also applied in natural gas processing, ammonia manufacturing, coal gasification, and ethylene oxide production. For instance, amine-based solvents are applied in a closed-loop absorption–desorption

system in gas processing units.^[1,2] Amines are classified as primary (such as monoethanolamine, MEA), secondary (such as diethanolamine, DEA) or tertiary (e.g., triethanolamine, TEA, or N-methyl diethanolamine, MDEA). MEA and DEA react quickly with CO₂ but their loading capacity is low and regeneration cost is high. Contrarily, MDEA and TEA are less reactive and easily regenerable, and they load more CO₂.^[3,4] Piperazine and MEA are suitable rate promoters for MDEA.^[5–7]

N,N-Diethylethanolamine (DEEA), which is a tertiary amine comprising two ethyl groups attached to nitrogen,

has recently attracted much attention from academia. [8-10] For instance, the kinetic and thermodynamic features of DEEA were reported in our earlier works. [11,12] Kinetics analysis was also reported in the past works of others. [13,14] Between 293 and 353 K, Luo et al. [15] reported equilibrium CO₂ solubility in DEEA in the 1-4 M range of molarity. Ethanol, producible from renewable biomass, can be used for manufacturing DEEA. [16] Additionally, DEEA reacts faster than MDEA, and thus is more advantageous. [17] In the past, several rate promoters were added to DEEA, for example, piperazine, [11,13,17,18] N-ethylethanolamine, [8,16] hexamethylene diamine, [11] N-(2-aminoethyl) ethanolamine, [11] and ethylene diamine.

Alkaline salts of natural amino acids (AAsH) are a special class of eco-friendly solvents that can promote CO₂ absorption in DEEA. Life cycle assessment studies suggested that the ecological footprint of CO₂ separation can be lowered by applying AAsH. [21-25] In this way, the CO₂-capturing process can be sustainable. In the past, MDEA was promoted with potassium glycinate (PG) and potassium arginate (PA). [27-29] Previously, we showed that PG is an effective promoter in DEEA solutions. [30] Interestingly, PG, PA, and potassium lysinate (PL) catalyzed the desorption of loaded MEA solutions, according to Li et al. [31] Clearly, further investigation on absorption and desorption features of DEEA/AAsH mixtures will be useful.

Ramezani et al.^[32] reviewed useful properties of sodium and potassium salts of 20 amino acids by analyzing data published in past works. Gusnawan et al.^[33] proposed bio-solvents derived from soybean and moringa protein, which are rich in glycine, alanine, aspartic acid, glutamic acid, and leucine. Recently, we showed that animal and plant protein are candidate resources for renewable amino acid-based solvents.^[34] Especially, PG, PA, and potassium prolinate (PP) were most reactive among nine different amino acid salts investigated in our work.

In this work, absorption and desorption features of DEEA/PA, DEEA/PP, and DEEA/PG mixtures were investigated. The influence of these salts on the absorption and desorption process was studied. Kinetic trials were performed in a stirred cell and rate constants were measured. Equilibrium measurements were performed in an equilibrium cell and empirical equations that predicted solubility data were proposed. Loaded solutions were desorbed in a batch apparatus and cyclic capacity, desorption rates, and reduction in sensible energy constraint were measured. Amine regeneration can be improved by using transition metal oxide catalysts. They accelerate the desorption process of CO₂-rich amines. During the past decade, much effort was focused on the

optimization of catalytic desorption of MEA among other amines. In this work, the efficacy of AAsH on the catalytic desorption of DEEA was compared to that of alumina, which is renowned for improving amine regeneration.

2 | THEORY

2.1 | Absorption theory

The pathway for the reaction between CO_2 and DEEA was described in many past works.^[5,8,12,35,36] Typical of tertiary amines, DEEA promotes CO_2 hydrolysis in an aqueous medium. This reaction is represented in Equation (1):

$$DEEA + H_2O + CO_2 \xrightarrow{k_{2-DEEA}} DEEA^+H + HCO_3^-$$
 (1)

Amino acids are protonated in aqueous medium and exist as zwitterions.^[30] These zwitterions are transformed into amino acid salts in alkaline medium (see Equation (2)):

$$R - CH(NH2) - COOH + KOH$$

$$\stackrel{aq.}{\rightarrow} R - CH(NH2) - COO^{-}K^{+} + H2O$$
 (2)

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Here, R varies with the type of amino acid {arginine: $HN=C(NH_2)-NH-(CH_2)_3$, glycine: H, proline: $(CH_2)_3$ }. The pathway for the reaction between CO_2 and potassium salts of amino acids is in line with the zwitterion mechanism. [37,38] According to this mechanism, CO_2 and amino acid salts combine to produce intermediate zwitterion that is protonated by a base to form carbamate. This mechanism is shown in the reactions represented by Equations (3) and (4):

$$CO_2 + AAsH \xrightarrow{k_{2-AAsH}} AAsH^+COO^-$$
 (3)

$$AAsH^{+}COO^{-} + B \xrightarrow{k_B} AAsCOO^{-} + BH^{+}$$
 (4)

Here, the salt is denoted by AAsH, while AAs represents ^+K —OOC—HC(HN)—R.

The rate of reaction is obtained by applying the steady-state principle to the intermediate zwitterion:

$$R_{\text{CO}_2-\text{AAsH}} = \frac{k_{2-\text{AAsH}}(\text{CO}_2)(\text{AAsH})}{1 + \frac{k_{-1}}{k_{\text{B}}(\text{B})}}$$
 (5)

When the base AAsH, OH^- , and H_2O contribute significantly to zwitterion deprotonation, Equation (5) is transformed into Equation (6):

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$$R_{\text{CO}_2-\text{AAsH}} = \frac{k_{2-\text{AAsH}}(\text{CO}_2)(\text{AAsH})}{1 + \frac{k_{-1}}{k_{\text{AAsH}}(\text{AAsH}) + k_{\text{OH}} - (\text{OH}^-) + k_{\text{H}_2O}(\text{H}_2O)}}$$
(6)

On the whole, AAsH contributes to the absorption of CO_2 and enhances the overall rate of absorption. The overall rate of all reactions in an aqueous mixture of AAsH and DEEA is represented by the following:

$$\begin{split} R_{CO_2-DEEA/AAsH} &= \frac{k_{2-AAsH}(CO_2)(AAsH)}{1 + \frac{k_{-1}}{k_{AAsH}(AAsH) + k_{OH^-}(OH^-) + k_{H_2O}(H_2O)}} \\ &\quad + \Big[k_{H_{2O}}(H_2O) + k_{OH^-}(OH^-) \\ &\quad + k_{2-DEEA}(DEEA)\Big](CO_2) \end{split} \label{eq:RCO_2-DEEA} \end{split}$$

The observed reaction rate constant is defined by the following:

$$k_{\text{obs}} = R_{\text{CO}_2} / (\text{CO}_2) \tag{8}$$

From Equation (7), k_{obs} is given by the following:

$$\begin{split} k_{obs-DEEA/AAsH} = & \frac{k_{2-AAsH}(AAsH)}{1 + \frac{k_{-1}}{k_{AAsH}(AAsH) + k_{OH^-}(OH^-) + k_{H_2O}(H_2O)}} \\ & + k_{H_2O}(H_2O) + k_{OH^-}(OH^-) \\ & + k_{2-DEEA}(DEEA) \end{split} \tag{9}$$

The contribution of $k_{\rm H_2O}$ in Equation (9) can be neglected. The concentration of OH^- depends on the pKa value of the amine. At low OH^- concentration, it is known that the contribution of $k_{\rm OH^-}(OH^-)$ in Equation (9) can also be neglected. When deprotonation is almost instantaneous as compared to the reverse reaction in Equation (3) (i.e., $k_{\rm B}(B)$ is very large due to the high value of $k_{\rm AAsH}$), Equation (9) is expressed as follows:

$$\mathbf{k}_{\text{obs-DEEA/AAsH}} = \mathbf{k}_{\text{2-AAsH}}(\mathbf{AAsH}) + \mathbf{k}_{\text{2-DEEA}}(\mathbf{DEEA}) \tag{10}$$

Thus, the overall reaction comprised two parallel reactions of CO_2 with DEEA and AAsH.

2.2 | Estimation of kinetics

The CO_2 content in the bulk liquid could be neglected at the reaction conditions chosen for this work. According to Doraiswamy and Sharma, ^[40] the value of R_{CO_2} for the fast reaction regime is given by Equation (11):

$$R_{CO_2} = k_L(CO_2)E \tag{11}$$

In this reaction regime, the Hatta number is equal to the value of the enhancement factor due to chemical reaction E:

$$E = \mathrm{Ha} = \frac{\sqrt{\mathrm{D}_{\mathrm{CO}_2} k_{\mathrm{obs}}}}{\mathrm{k}_{\mathrm{I}}} \tag{12}$$

The inequality $10 < \text{Ha} < < (E_i - 1)$ is satisfied in the fast reaction regime. Here, E_i is the enhancement factor for an instantaneous reaction, whose value as per the film theory is given by Equation (13):

$$E_i = 1 + \left[\frac{(Am)}{z (CO_2)} \frac{D_{Am}}{D_{CO_2}} \right]$$
 (13)

Here, z denotes the stoichiometric coefficient. It is evident from Equations (11) and (12) that R_{CO_2} is given by the following:

$$R_{CO_2} = \sqrt{D_{CO_2} k_{obs}} \tag{14}$$

According to Henry's law, the saturation concentration of CO_2 is given by $(CO_2) = P_{CO_2}H_{CO_2}$. It is evident from Equations (10) and (14) that rate is given by the following:

$$R_{CO_2} = P_{CO_2} H_{CO_2} \sqrt{D_{CO_2} [k_{2-AAsH} (AAsH) + k_{2-DEEA} (DEEA)]} \eqno(15)$$

Vaidya and Kenig^[41] reported a method for determining the volumetric rate of CO_2 absorption in a stirred cell reactor. In this method, the fall in pressure inside the reactor enables the determination of the gradient dP_{CO_2}/dt , and hence, the rate (see Equation (16)):

$$R_{CO_2} = \left(-\frac{dP_{CO_2}}{dt}\right) \left(\frac{V_G}{V_I RT}\right) \tag{16}$$

2.3 | Desorption theory

The cyclic capacity is a useful feature of CO_2 -capturing solvents. A high value of cyclic capacity suggests low regeneration cost. [42] Patil et al. [43] showed that the cyclic capacity is found from the difference between the loading (mol CO_2 / mol amine) of the rich and lean solvent (see Equation (17)):

$$Q_{\text{Cyc}} = (\alpha_{\text{rich}} - \alpha_{\text{lean}}) \tag{17}$$

From knowledge of the cyclic capacity, another useful desorption parameter, viz. the amount (moles) of CO_2 desorbed can be found (see Equation (18)):

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$$n_{\text{CO}_2} = (\alpha_{\text{rich}} - \alpha_{\text{lean}}) \times C \times V_{\text{L}} = Q_{\text{Cyc}} \times C \times V_{\text{L}}$$
 (18)

According to Bhatti et al., $[^{44}]$ the rate of desorption of CO_2 can be found from the cyclic capacity using Equation (19):

$$r = \frac{(\alpha_{\text{rich}} - \alpha_{\text{lean}})}{t} = \frac{Q_{\text{Cyc}}}{t}$$
 (19)

A high value of desorption rate suggests fast regeneration and low heat duty (for an equal supply of thermal energy). The efficiency of desorption can be calculated using Equation (20):

$$\eta = \frac{(\alpha_{\text{rich}} - \alpha_{\text{lean}})}{\alpha_{\text{rich}}} = \frac{Q_{\text{Cyc}}}{\alpha_{\text{rich}}}$$
(20)

The regeneration energy comprises the heat of desorption, the heat of vaporization and the sensible heat. If the values of heats of desorption and vaporization are low, the sensible heat provides the regeneration energy constraint (see Equation (21)):

$$Q_{\rm S} = \frac{C_{\rm p}(T_i - T_{\rm o})}{(\alpha_{\rm rich} - \alpha_{\rm lean}) \, x_{\rm Am} \, M_{\rm CO_2}} = \frac{C_{\rm p} \Delta T}{Q_{\rm Cyc} \, x_{\rm Am} M_{\rm CO_2}}$$
(21)

3 | EXPERIMENTAL METHODOLOGY

3.1 | Materials

DEEA, arginine, glycine, proline, sodium hydroxide, and potassium hydroxide were purchased from Molychem Pvt. Ltd., Mumbai. The minimum purity of these chemicals was 97%. Potassium salts of the amino acids were prepared by neutralizing an aqueous solution of the amino acid with an equimolar quantity of potassium hydroxide under continuous cooling. CO₂, N₂, and N₂O gas cylinders (purity 99.95%) were purchased from Inox Air Products Ltd., Mumbai. The molecular structures of DEEA and the amino acids used in this work are shown in Figure 1.

3.2 | Experimental section

3.2.1 | Trials for absorption kinetics

A stirred cell reactor, which was provided by Sharad Autoclave Engineers, Mumbai, was used for studying absorption kinetics. It was described in our past works.^[11,45] Typically, the jacketed reactor was purged

with N_2 , evacuated, filled with fresh absorbent (400 mL), and maintained at 303 K. Next, CO_2 was introduced in a gas reservoir, which was also maintained at 303 K using a water circulation bath. Thereafter, CO_2 was introduced inside the reactor and gas and liquid phases were separately stirred. The agitation speeds were 1000 (gas phase) and 65 rpm (liquid phase). It was ensured that the gas–liquid interface was flat and undisturbed. The drop in the pressure reading was monitored for 20 s using a pressure transmitter and data acquisition system. Using Equation (16), the absorption rate was measured at different concentrations of DEEA and promoter. The results were reproducible (error <5%).

3.2.2 | Equilibrium CO₂ solubility measurements (VLE)

A high-pressure vapour-liquid equilibrium (VLE) setup was used (see Figure 2A). It was supplied by BEEM Industries Ltd., Nashik. It was made of SS316 and could be used up to 10 MPa pressure. It comprised reservoirs for gas (capacity 1.86 L) and liquid, equilibrium cell (capacity 0.7 L), vacuum pump, and safety line (with backpressure release valve and rupture disc). Both reservoirs and equilibrium cell were maintained at 303 K using a water circulation bath. They were equipped with temperature indicator and controller (TIC) and pressure indicator (PI), which were supplied by Selec Control, Navi Mumbai. The gas and liquid phases inside the equilibrium cell were stirred using an agitator. The equilibrium cell could be isolated from the reservoirs using needle valves. In every trial, the absorbent (235 mL) was charged from the liquid reservoir into the equilibrium cell. Next, CO2 from the gas reservoir was charged inside the cell. Both phases were agitated at 1000 rpm and the fall in pressure was recorded. Typically, the pressure inside the cell was almost constant after 0.5 h, thereby suggesting the attainment of equilibrium. From knowledge of the pressure readings in the gas reservoir and the equilibrium cell, the respective moles of CO2 were determined using the van der Waals equation (see Equation (22)):

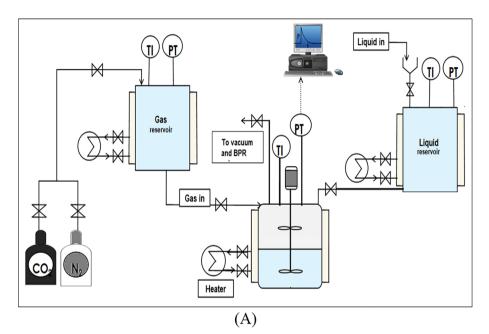
$$\left[P_{\text{CO}_2} + \frac{\text{an}_{\text{CO}_2}^2}{V_{\text{G}}^2}\right] \times \left[V_{\text{G}} - n_{\text{CO}_2}b\right] = nRT_{\text{i}}$$
(22)

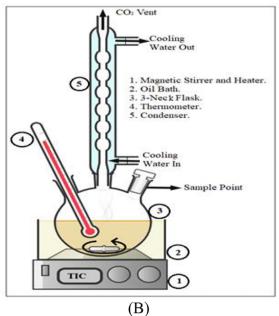
The values of the constants a and b in Equation (22) for CO_2 are 0.364 J-m³/mol² and 4.27 × 10⁻⁵ m³/mol.^[46] Knowing the values of the moles of CO_2 transferred from the gas reservoir (n_T) and the residual moles of CO_2 upon the attainment of equilibrium ($n_{CO_2}^*$), the moles of CO_2 absorbed were determined using Equation (23):

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FIGURE 1 Molecular structures of N,N-diethylethanolamine (DEEA) and amino acids used in this study.

FIGURE 2 Schematics of (A) highpressure vapour–liquid equilibrium (VLE) setup and (B) batch desorption apparatus.





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$$n_{\rm CO_2} = n_{\rm T} - n_{\rm CO_2}^* \tag{23}$$

Using Equation (24), equilibrium solubility was found:

$$\propto \left(\frac{\text{mol CO}_2}{\text{mol amine}}\right) = \frac{n_{\text{CO}_2}}{n_{\text{Am}}}$$
 (24)

The error (<3%) in measuring VLE data was found by repeating a few trials.

| CO₂ desorption trials

The batch setup for amine regeneration, earlier used by Zhang et al., [47] was modified and used (see Figure 2B). It comprised an oil bath, thermometer, three-necked glass desorption reactor (capacity 1000 mL, Make: Borosil), reflux condenser (with arrangement for circulation of cooling water), magnetic stirrer, and heater (Remi Instruments, Mumbai).

The absorbent was loaded with CO₂ in the stirred cell, thereby ensuring a constant initial CO2 loading for the desorption trials. The molar volume of the solution was used to calculate the moles of amine for the desired loading (moles of CO₂/moles of amine), while the equation of state (van der Waals equation) was used to calculate the moles of CO₂. After the desired loading (0.25 mol CO₂/mol amine) was achieved, the CO₂-rich solution (400 mL) was transferred to the batch desorption setup. The desorber was heated to 363 K. A temperature lower than the usual values in industrial regenerators and reboilers was chosen in this work to especially highlight the influence of promoters on the desorption process. The desorption time, also chosen earlier by Zhang et al., [48] was 9 h. However, the equilibrium CO2 loading of aqueous DEEA solutions did not significantly change after 3 h. As a result, desorption time was chosen as 3 h. Liquid samples were collected at 0.5, 1, 1.5, 2, and 3 h, and their CO₂ content was determined using the method of titration reported by Altabash et al. [49] An auto-titrator (877 Titrino Plus Auto titrator, Metrohm India Private Limited, Bangalore) was used. During catalytic trials, 1.5 g of alumina was initially added to 400 mL of solution inside the desorption reactor and the aforesaid procedure was then followed.

In this method, pH of 50 mL methanol was adjusted to a value of 11 using an aqueous 0.5 M sodium hydroxide solution. A known volume of the desorbed sample was then injected into the aforesaid methanol. This lowered the pH value, and the mixture was back-titrated to pH = 11 by adding sodium hydroxide. This additional amount of sodium hydroxide was noted and used to compute the equilibrium CO_2 loading (\propto). [49] The acid-base

neutralization was used as the basis for computation. The error in measuring loading was <2%. The calculated values of CO2 lean solutions were further used for the estimation of desorption rates and other desorption properties. More details were provided in our recent work. [46]

RESULTS AND DISCUSSION

Estimation of physical properties

The density and viscosity of the DEEA-AAsH systems were measured using a specific gravity bottle and capillary viscometer, respectively. The uncertainty in these values was found by repeating the measurements thrice. The average absolute deviation was estimated to be 2%. At 303 K, the diffusion coefficients for N₂O $(2.03 \times 10^{-9} \text{ m}^2/\text{s})$ and CO_2 (2.15 × 10⁻⁹ m²/s) in water were reported in a past work.^[50] From the viscosity measurements of this work at 303 K and modified Stokes-Einstein correlation, [51] N₂O diffusivity in DEEA-AAsH solutions was determined using Equation (25):

$$\left(D_{N_2O}\mu^{0.8}\right)_{amine} = Constant = \left(D_{N_2O}\mu^{0.8}\right)_{water}$$
 (25)

The values of D_{CO2} and H_{CO2} in DEEA-AAsH solutions were estimated using the N₂O analogy method. ^[50] The estimated values of various physical properties for DEEA, AAsH, and their blends are listed in Table 1. For instance, the measured value of viscosity of DEEA $(1.5 \,\mathrm{M})$ was 1.66 mPa-s at 303 K, while the value of $\mathrm{H}_{\mathrm{CO}_2}$ was 2.38×10^{-4} kmol/(m³ kPa). Knowledge of these parameters is essential for estimating the values of reaction rate constants.

Estimation of liquid-side mass 4.2 transfer coefficient (k_L)

The value of k_L was estimated using a method reported by Littel et al. [52] From a mass balance of the solute gas in both the gaseous and liquid phases, the following equation was derived in their work:

$$\ln \left[\frac{P(t) - P_{\text{final}}}{P_{\text{initial}} - P_{\text{final}}} \right] = - \left[\frac{(\widehat{m}V_{\text{L}}) + V_{\text{G}}}{V_{\text{L}}V_{\text{G}}} \right] k_{\text{L}} A_{\text{i}} t$$
 (26)

A plot of $ln\left[\frac{P(t)-P_{final}}{P_{initial}-P_{final}}\right]$ versus t was drawn using the outcome of a trial on the physical absorption of CO_2 in water at 303 K in the stirred cell. The values of \widehat{m} $(=0.59 \, \text{mol/mol}), \ V_{\rm G} \ (=1080 \, \text{mL}), \ V_{\rm L} \ (=400 \, \text{mL}), \ \text{and} \ A_{\rm i}$ $(=45.4 \text{ cm}^2)$ were known. It was found that k_L equals

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 $5.45 \times 10^{-5}\,\text{m/s}$. More details were provided in a recent work. ^[51] This value is identical to the k_L values for this model contactor in general.

4.3 | Estimation of reaction kinetics

Kinetics trials were performed at 303 K and the results are represented in Table 2. First, the absorption rates in DEEA (1.5, 2, and 2.5 kmol/m³) were measured. Typical of the concentrations of the benchmark amine MEA (30 wt.%) often used in laboratory-scale kinetic studies, the highest DEEA concentration used in this work was 2.5 M (or 29.2 wt.%). The rate values increased with the rise in molarity in solution. Especially, the values of rate for 2 M

 $(0.87\times10^{-6}~\text{kmol/(m}^2~\text{s}))$ and 2.5 M DEEA $(1.27\times10^{-6}~\text{kmol/(m}^2~\text{s}))$ were identical to those reported in our past works. The values of a lumped kinetic parameter $H_{CO_2}\sqrt{D_{CO_2}k_2}$ were also determined. The second order rate constant k_2 for 1.5 M DEEA (67 m³/(kmol s)) was in agreement with the value reported in the past works of Kierzkowska-Pawlak and Kruszczak [53] ($k_2=65~\text{m}^3/(\text{kmol s}))$) and Nozaeim et al. [54] ($k_2=62~\text{m}^3/(\text{kmol s}))$.

Next, trials were performed with PP, PA, and PG solutions (0.5 M). To ensure conformation of the reaction system to the fast reaction regime systems, higher promoter concentrations were not applied in this work. As evident from the rate values, these salts were significantly more reactive than DEEA. Thus, they are candidate promoters for DEEA. In general, the reactivity of salts was of the order

TABLE 1 Solution property data for DEEA, AAsH, and their blends at 303 K.

	Concentrat	ion (M)	Density	$\mu imes 10^3$	$\mathrm{D_{CO_2}} imes 10^9$	$\mathrm{H_{CO_2}} imes 10^4$
Amine system	DEEA	AAsH	(kg/m^3)	(Pa-s)	(m^2/s)	(kmol/(m³ kPa))
DEEA	1.5	0	974	1.66	1.20	2.38
DEEA	2	0	968	2.15	0.97	2.41
DEEA	2.5	0	963	2.74	0.81	2.44
PP	0	0.5	1022	1.52	1.28	2.63
PA	0	0.5	1036	0.97	1.84	2.47
PG	0	0.5	1027	0.88	1.99	2.58
DEEA + PP	2	0.5	1004	2.04	1.01	2.70
DEEA + PA	2	0.5	1010	1.76	1.14	2.65
DEEA + PG	2	0.5	1002	1.69	1.18	2.69

Abbreviations: AAsH, alkaline salts of natural amino acids; DEEA, N,N-diethylethanolamine; PA, potassium arginate; PG, potassium glycinate; PP, potassium prolinate.

TABLE 2 Kinetic data for DEEA, AAsH, and their blends at 303 K.

	Concentration (kmol/m³)		P_{CO_2}	$R_{CO_2} imes 10^6$	$\rm H_{\rm CO_2} \sqrt{D_{\rm CO_2} k_2} \times 10^7$	k ₂ (m ³ /	k _{obs}		
Amine system	DEEA	AAsH	(kPa)	$(\text{kmol}/(\text{m}^2 \text{s}))$	$(\text{kmol}^{1/2}/(\text{m}^{1/2} \text{ s kPa}))$	(kmol s))	(1/s)	E = Ha	E_i
DEEA	1.5	0	5.8	0.48	0.68	67	101	6	1083
DEEA	2	0	5.5	0.87	1.12	220	441	12	1508
DEEA	2.5	0	5.2	1.27	1.53	492	1229	18	1961
PP	0	0.5	6.1	5.90	13.63	2095	10,475	67	157
PA	0	0.5	7.4	6.80	13.09	15,211	7605	69	138
PG	0	0.5	7.6	8.43	15.69	18,579	9289	79	129
DEEA + PP	2	0.5	7.0	2.92	2.63		2351	28	1322
DEEA + PA	2	0.5	7.4	3.83	3.28		3362	36	1282
DEEA + PG	2	0.5	8.5	3.57	2.65		2071	29	1094

Abbreviations: AAsH, alkaline salts of natural amino acids; DEEA, N,N-diethylethanolamine; PA, potassium arginate; PG, potassium glycinate; PP, potassium prolinate.

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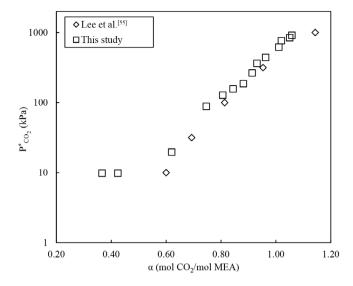


FIGURE 3 A comparison of vapour-liquid equilibrium (VLE) data of this work with Lee et al.[55] (monoethanolamine (MEA) = 1 M, T = 313 K).

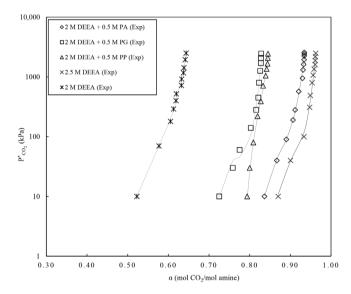


FIGURE 4 A plot of equilibrium partial pressure of CO₂ versus loading capacity at 303 K (solid lines denote model predictions). DEEA, N,N-diethylethanolamine; PA, potassium arginate; PG, potassium glycinate; PP, potassium prolinate.

PP > PG > PA. The k_2 value for PP was the highest (20,950 m³/(kmol s)). Upon the promotion of DEEA (2 M) with PP, PA, and PG (0.5 M), the enhancement in absorption rate was observed. The observed rate constants for DEEA/PP, DEEA/PG, and DEEA/PA were 2351, 2071, and 3362 1/s, correspondingly. From the values of E and Ha, it was confirmed that the investigated systems belonged to the fast pseudo-first order reaction regime systems.

Equilibrium solubility of DEEA-AAsH systems

The dependence of equilibrium CO2 partial pressure (P*CO2) on the CO2 loading capacity of solutions (mol CO₂/mol amine) was studied. Trials were first performed at 313 K using MEA solutions (1 kmol/m³). The results for MEA were in good agreement with the outcome of a past work of Lee et al. [55] (see Figure 3). Further trials were performed using DEEA/promoter mixtures (2/05 kmol/m³) at 303 K. The results are represented in Figure 4. The loading capacity of DEEA was enhanced upon promoter addition. It is evident from Figure 4 that the loading capacity varied between 0.523 and 0.962 mol CO₂/mol amine. For a given partial pressure, the loading capacity increased in the order: 2 M DEEA < DEEA/PG < DEEA/PP < DEEA/PA < 2.5 M DEEA. For example, the values of α for 2 M DEEA, DEEA/ PG, DEEA/PP, DEEA/PA, and 2.5 M DEEA were 0.523, 0.725, 0.794, 0.836, and 0.87 mol CO₂/mol total amine at $P_{CO_2}^* = 10 \text{ kPa}$. Empirical polynomial-type model for DEEA and DEEA-AAsH systems was developed to represent VLE data. It is represented by Equation (27):

$$\log(P_{CO_2}^*) = A'\alpha^3 - B'\alpha^2 + C'\alpha - D'$$
 (27)

Using least square regression, the values of A', B', C', and D' were estimated from the data between 10 and 2500 kPa. The model accurately predicted equilibrium solubility for different DEEA-AAsH systems used in this study. The error in the predictions was estimated by the

DEEA-AAsH system	\mathbf{A}'	\mathbf{B}'	C'	\mathbf{D}'	% AAD
•			-		
2 M DEEA	1411.9	2404.2	1377.8	263.94	0.52
2.5 M DEEA	6087.5	16,524	14,961	4516.66	0.29
DEEA-PG	6301.2	14,515	11,149	2553.85	0.91
DEEA-PP	16,589	41,037	33,873	9326.88	0.40
DEEA-PA	3653.8	9585.3	8395.5	2453.345	0.14

TABLE 3 Estimated values of the constants in Equation (27) and percent average absolute deviation (AAD).

Abbreviations: AAsH, alkaline salts of natural amino acids; DEEA, N,N-diethylethanolamine; PA, potassium arginate; PG, potassium glycinate; PP, potassium prolinate.

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percent average absolute deviation (%AAD) described in Equation (28):

$$\%AAD = \pm \sum_{i=1}^{N} \left(\frac{P_{CO_{2,i}}^{\text{model}} - P_{CO_{2,i}}^{\text{exp}}}{P_{CO_{2,i}}^{\text{exp}}} \right) \times 100$$
 (28)

The error in prediction was <1%. The results are shown in Figure 4 and Table 3.

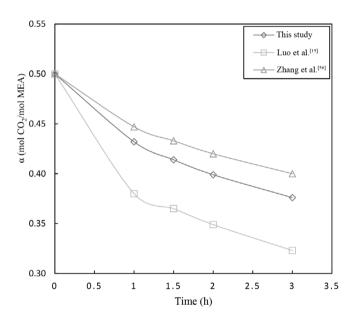


FIGURE 5 Plots of loading capacity vs. time for 5 M monoethanolamine (MEA) at 368 K and comparison with the works of Luo et al. [15] and Zhang et al. [56]

TABLE 4 Desorption features of DEEA and DEEA-AAsH systems at 363 K.

Amine system	Q _{cyc} (mol CO ₂ /mol total amine)	η	$r \times 10^2$ (mol CO_2 /(mol total amine h))	n _{CO2} (mol)
DEEA (1.5 M)	0.17	0.66	5.53	0.1
DEEA (2 M)	0.15	0.60	5.03	0.12
DEEA (2.5 M)	0.11	0.46	3.79	0.11
$DEEA+Al_2O_3 (2.5 M)$	0.13	0.51	4.28	0.13
DEEA + PP (2 + 0.5 M)	0.12	0.48	3.99	0.12
DEEA + PA (2 + 0.5 M)	0.20	0.80	6.73	0.20
DEEA + PG (2 + 0.5 M)	0.12	0.46	3.84	0.12

Abbreviations: AAsH, alkaline salts of natural amino acids; DEEA, N,N-diethylethanolamine; PA, potassium arginate; PG, potassium glycinate; PP, potassium prolinate.

4.5 | Desorption trials

Several useful solvent features such as cyclic capacity, amount of CO_2 desorbed, rate of CO_2 desorption, CO_2 desorption efficiency, and regeneration energy requirements were determined to study the overall desorption performance. The experimental setup was validated using 5 M MEA at 368 K with the past works of Luo et al. [15] and Zhang et al. [56] The results are represented in Figure 5. The cyclic capacity for MEA obtained in this work (0.124 mol/mol) was between the values reported by Zhang et al. [16] (0.1 mol/mol) and Luo et al. [15] (0.177 mol/mol). The outcome of desorption trials for DEEA and DEEA–AAsH systems at 363 K is represented in Table 4. The cyclic capacity of DEEA, desorption

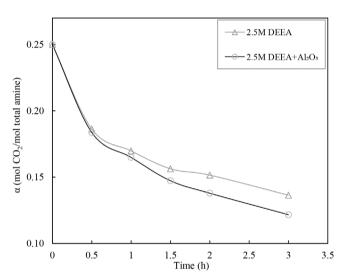


FIGURE 6 Plot of loading capacity of 2.5 M N,N-diethylethanolamine (DEEA) versus time at 363 K without and with Al_2O_3 catalyst (loading 1.5 g).

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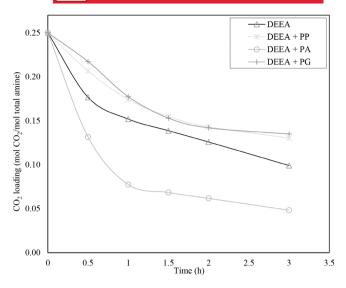


FIGURE 7 Plot of loading capacity versus time at 363 K for N,N-diethylethanolamine (DEEA) and DEEA–alkaline salts of natural amino acids (AAsH) systems.

efficiency, and rate of desorption decreased with the rising molarity of DEEA in solutions.

The solid metal oxide catalyst Al_2O_3 is effective for improved desorption of CO_2 -rich amines. [57,58] In our recent work, [46] we discussed the role of metal oxide catalysts in the desorption process. This catalyst (loading 1.5 g) was also chosen in the present work for accelerating desorption of DEEA (2.5 M) at T=363 K. The results are shown in Table 4 and Figure 6. It was found that desorption efficiency and rate of desorption improved when Al_2O_3 was used, thus resulting in reduced energy requirement (4%).

The influence of AAsH (0.5 M) on the desorption process for DEEA (2 M) was also studied at 363 K. The results are represented in Figure 7 and Table 4. As evident from Figure 7, PP and PG lowered the cyclic capacity of DEEA. However, the addition of PA improved the cyclic capacity of DEEA. Additionally, it is evident from Table 4 that DEEA/PA mixtures exhibited the highest values of desorption efficiency (0.8) and rate of desorption $(6.73 \times 10^{-2} \text{ mol CO}_2/(\text{mol amine h}))$. The effect of promoter addition on the sensible energy constraint was also studied at 363 K. It was found that PG, PP, and PA (0.5 M) lowered the sensible energy requirement of DEEA (2 M) by 30%, 32%, and 59%.

5 | CONCLUSIONS

The tertiary amine DEEA, which is producible from renewable resources, has high loading capacity but low reactivity with CO₂. The addition of promoters improves reactivity;

additionally, the loading capacity and desorption features of DEEA are influenced too. In this work, the performance of DEEA solutions promoted with three salts of amino acids, viz. PA, PP, and PG, was comprehensively studied. Solution properties of DEEA, DEEA/PA, DEEA/PP, and DEEA/PG (2/0.5 kmol/m³), such as viscosity and CO₂ diffusivity, were determined and absorption rates were measured in a stirred cell. The pseudo-first order rate constants at 303 K were measured. It was found that k_L for a stirred cell equals 5.45×10^{-5} m/s. The dependence of CO₂ equilibrium partial pressure on loading capacity of the promoted DEEA solutions was studied at 303 K and empirical polynomialbased models that accurately predicted solubility data were proposed. Using a batch setup, desorption properties such as cyclic capacity, desorption rates, and efficiency of desorption were determined at 363 K. PP and PG lowered the cyclic capacity of DEEA. However, the addition of PA improved the cyclic capacity of DEEA. DEEA/PA mixtures exhibited the highest values of rate and efficiency of desorption. It was found that PG, PP, and PA (0.5 M) lowered the sensible energy requirement of DEEA (2 M) by 30%, 32%, and 59%. Finally, it was found that desorption efficiency and rate of desorption improved when Al₂O₃ was used.

NOMENCLATURE

 (H_2O)

Ha

a	van der Waals constant in Equation (22)
	(J m ³ /mol ²)
A_i	gas-liquid interfacial area (m²/m³)
A'	constant in Equation (27)
AAsH	amino acid salt
(AAsH)	concentration of amino acid salt (kmol/m³)
Am	amine system
b	van der Waals constant in Equation (22)
	(m^3/mol)
В	base assisting in zwitterion deprotonation
(B)	concentration of base B in bulk liquid
	(kmol/m³)
B'	constant in Equation (27)
C'	constant in Equation (27)
(CO_2)	concentration of carbon dioxide (kmol/m³)
$c_{ m p}$	specific heat capacity (J/(mol K))
D'	constant in Equation (27)
D_{Am}	diffusivity of amine in liquid (m²/s)
$\mathrm{D}_{\mathrm{CO}_2}$	diffusivity of CO ₂ in liquid (m ² /s)
$\mathrm{D}_{\mathrm{N_2O}}$	diffusivity of N ₂ O in liquid (m ² /s)
(DEEA)	concentration of DEEA (kmol/m³)
E	enhancement factor due to chemical reaction
E_i	enhancement factor for an instantaneous
	reaction

concentration of water (kmol/m³)

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Ц	Henry's law constant (kmol/(m³ kPa))
H_{CO_2}	forward reaction rate constant in Equation (3)
k_{2-AAsH} k_{-1}	reverse reaction rate constant in Equation (3)
	rate constant for deprotonation by AAsH
k _{AAsH} k _B	rate constant for deprotonation by AASIT
	rate constant for deprotonation by H ₂ O
k _{H2O}	rate constant for deprotonation by H ₂ O
k _{OH} - k _L	liquid side mass transfer coefficient (m/s)
_	observed reaction rate constant (1/s)
$\widehat{\mathrm{m}}$	dimensionless solubility (mol/mol)
	nitrous oxide
N ₂ O	
$n_{ m Am}$	moles of amine (mol) moles of CO ₂ (mol)
n_{CO_2}	moles of CO ₂ (mol) moles of CO ₂ at equilibrium (mol)
$n_{\mathrm{CO}_2}^*$	moles of CO ₂ at equilibrium (mor) moles of CO ₂ transferred to the reactor (mol)
$n_{ m T}$ (OH $^-$)	concentration of hydroxide ion (kmol/m ³)
P_{CO_2}	
1 CO ₂	partial pressure of CO ₂ in the bulk gas phase (kPa)
D *	equilibrium CO ₂ partial pressure (kPa)
$P^*_{CO_2}$ $P(t)$	partial pressure of solute gas at time t (kPa)
P _{final}	final partial pressure of solute gas (kPa)
P _{initial}	initial partial pressure of solute gas (kPa)
Q_{Cyc}	cyclic capacity (mol CO ₂ /mol amine)
$Q_{\rm S}$	sensible heat (kJ/g)
$Q_{ m V}$	heat of vaporization (kJ/g)
r	rate of CO_2 desorption (mol CO_2 /(mol
•	amine h))
R	ideal gas law constant (kPa m³)/(kmol K)
R_{CO_2}	specific rate of absorption of CO ₂ (kmol/
11002	$(m^2 s)$
T	temperature (K)
t	time (h)
$V_{ m G}$	volume of gas phase (cm ³)
$V_{ m L}$	volume of liquid phase (cm ³)
Z	stoichiometric coefficient
α	CO ₂ loading (mol CO ₂ /mol amine)
μ	viscosity (Pa s)
η	CO ₂ desorption efficiency
•	- 1

AUTHOR CONTRIBUTIONS

Hitesh D. Rawate: Methodology; investigation; validation; writing - original draft. Prakash D. Vaidya: Conceptualization; funding acquisition; resources; writing review and editing; supervision.

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CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could

have appeared to influence the work reported in this paper.

PEER REVIEW

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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