

Advanced Amino Acid-Based Technologies for CO₂ Capture: A Review

Vida Sang Sefidi[®] and Patricia Luis*

Materials & Process Engineering (iMMC-IMAP), UCLouvain, Place Sainte Barbe 2, 1348 Louvain-la-Neuve, Belgium

Supporting Information

ABSTRACT: Capture and utilization or storage of CO₂ have been considered as the primary options to reduce the impact on the climate caused by CO₂ emissions. Several technological approaches have been proposed depending on the point of actuation (precombustion, postcombustion, or oxyfuel combustion), but absorption processes are still the most extended solution implemented in the industry. Alkanolamines are typical solvents used for CO₂ capture, even though they have a noticeable negative impact on the environment. Amino acids and their salts have been appointed as alternative solvents since their functional group is similar to that of alkanolamines but presenting better properties. In addition, they show very good performance in CO₂ capture. This



manuscript reviews the state of the art of the use of amino acids in CO₂ capture and the main technologies involving the application of these solvents.

INTRODUCTION

Emission of greenhouse gases has a significant impact on the environment.¹ The rise of global temperature has already caused an increase in the polar ice melting and provoked a change in the climate.² Carbon dioxide (CO_2) is the main contributor in terms of emitted quantities, mostly caused by human activities.³ Technologies focused on precombustion, postcombustion capture and oxyfuel combustion have been developed to reduce CO₂ emissions.⁴ In precombustion processes, CO₂ is retained from a reformed fossil fuel gas stream called synthesis gas (syngas), which is mainly CO_2 and hydrogen (H_2) ;⁵ in postcombustion, the CO₂ is removed from existing flue gas, involving the separation of CO₂ from a nitrogen-rich stream.^{6,7} In oxyfuel combustion, fuel is burnt in a mixture of oxygen and flue gas that is recycled, therefore the outlet stream is rich in CO_2 .⁸ The pre- and postcombustion processes are mature technologies implemented in several commercial plants, but oxyfuel combustion technology is still under development.⁹ Figure 1 shows an overview of main CO_2 separation technologies used in each case.

Absorption processes have been widely implemented in the industry by using absorption columns and alkanolamine solvents, such as monoethanolamine (MEA) and diethanolamine (DEA), among others.^{10,11} This conventional process is very energy consuming, which causes a wide range of environmental impacts.¹² Those alkanolamine solvents are used due to a number of appropriate characteristics for CO₂ capture such as high absorption rate, low solvent cost, high resistance to thermal degradation, low molecular weight, and low solubility for hydrocarbons.^{13,14} However, they have some

disadvantages that cannot be neglected, such as low CO₂ loading, high energy requirements for regeneration, degradation through oxidation of the amine, vaporization losses due to high vapor pressure, and high viscosity.^{15,16} In addition, they can cause operational problems such as corrosion, foaming, and fouling of the process equipment.¹⁷ Therefore, researchers have been looking for alternative solvents to replace the conventional ones. As the amine group is the main reason for CO₂ capture in alkanolamines, researchers are studying other solvents with the amine group and by preference nonvolatile. One example is the polyamideoamine dendrimer Gen. 0.^{18,19} This solvent has a high selectivity toward CO₂ and showed promising results in combination with membrane technology.^{20,21} Other solvents such as ammonia, amino acids, and ionic liquids are also being considered as replacement of conventional amines. Ammonia has high loading capacity, no corrosion problems, and no absorption degradation. On the other hand, ammonia is a toxic solvent, its high volatility produces solvent losses at the top of the absorption columns, it is more expensive than other conventional solvents,²² and it produces a considerable amount of CO₂ emissions during its production process.²³ Ionic liquids are also candidates for CO₂ capture as they present good characteristics such as thermal stability and recyclable compared to other chemicals like amines, and they are considered as "green solvents" due to

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Pre-combustion	Post-combustion	Oxyfuel combustion
 Chemical adsorption Physical adsorption Chemical/physical absorption Chemical looping Membrane technology 	 Oxygen transport membrane boilers Ceramic auto-thermal recovery process Ion transport and membrane technology Cryogenic oxygen Chemical looping Chemical/physical absorption 	Chemical adsorption Physical adsorption Chemical absorption Cryogenics separation Membrane technology Bio chemical methods Ionic liquids

Figure 1. Classification of CO₂ capture technologies

their negligible vapor pressure. However, ionic liquids present some disadvantages, such as higher viscosity, which leads to a lower CO_2 absorption rate.^{24,25} Regarding amino acids, they have recently attracted the attention of the scientific community as solvents for CO_2 capture, since they have a similar functional group than alkanolamines.^{26,27} Amino acids solvents have been commercialized by BASF and Siemens Co. for acid gas removal. However, there is scope for further development.^{28,29} Amino acids have certain advantages over alkanolamines, such as higher surface tension, an almost nonvolatile nature, resistance to degradation in oxygen-rich flue gas, and better absorption.^{30–33} In addition, they could be synthesized from biosources.^{34,35} These characteristics make them a very attractive candidate in a global scenario for CO_2 capture.

Using environmentally friendly solvents is the first step in the development of a novel better process for CO₂ capture, but the selection of the technology to perform the capture is not less important.³⁶ As indicated above, absorption columns are typically the conventional technology for the absorption step. A second desorption step using heating is typically proposed, which has high energy consumption and is the main share in the capital cost of a CO₂ capture plant. Implementation of an economically feasible CO₂ capture plant has led to the introduction of membrane technology (e.g., membrane contactors) to capture CO₂ and produce other compounds of interest, such as (bi)carbonate salts. Furthermore, the recovery of the (bi)carbonate salts could be done via conventional crystallization or using membrane distillation/ crystallization. $^{37-40}$ The stripped CO₂ or the products like (bi)carbonate salts can be reused in other industries such as chemical and oil, food, mineralization, and power, instead of storing the CO_2 in the ground.⁴

In this review, a general overview of the use of amino acids for CO_2 capture as well as the different technologies proposed is presented. The amino acids kinetics for CO_2 capture and their performance are explained and a comparison of the performance of pure amino acids with the performance of blends with other solvents is introduced. Finally, the efficiency and performance of technologies is discussed and compared.

2. AMINO ACIDS AS CO₂ CAPTURING AGENT

Amino acids are organic compounds containing amine $(-NH_2)$ and carboxyl (-COOH) functional groups, along with a side chain (R group) specific to each amino acid. There are 20 different types. Amino acids are categorized in different aspects such as hydrophobicity or type of enantiomers.⁴² Figure 2 shows the general structure of amino acids. Based on their stereochemistry, amino acids are defined by D and L



Figure 2. General structure of amino acids.

enantiomers. The 20 amino acids are L-isomers, and their enantiomeric D-isomers are rarely found in nature. In Table 1, the list of amino acids can be seen.

Table 1. List of Amino Acids

name	3-letter symbol	molecular weight	molecular formula
alanine	Ala	89.1	C ₃ H ₇ NO ₂
arginine	Arg	174.2	$C_6H_{14}N_4O_2$
asparagine	Asn	132.12	$C_4H_8N_2O_3$
aspartic acid	Asp	133.11	$C_4H_7NO_4$
cysteine	Cys	121.16	$C_3H_7NO_2S$
glutamic acid	Glu	147.13	C ₅ H ₉ NO ₄
glutamine	Gln	146.15	$C_5H_{10}N_2O_3$
glycine	Gly	75.07	$C_2H_5NO_2$
histidine	His	155.16	C ₆ H ₉ N3O ₂
hydroxyproline	Нур	131.13	C ₅ H ₉ NO ₃
isoleucine	Ile	131.18	$C_6H_{13}NO_2$
leucine	Leu	131.18	$C_6H_{13}NO_2$
lysine	Lys	146.19	$C_6 H_{14} N_2 O_2$
methionine	Met	149.21	$C_5H_{11}NO_2S$
phenylalanine	Phe	165.19	$C_9H_{11}NO_2$
proline	Pro	115.13	$C_5H_9NO_2$
pyroglutamatic	Glp	139.11	C ₅ H ₇ NO ₃
serine	Ser	105.09	$C_3H_7NO_3$
threonine	Thr	119.12	$C_4H_9NO_3$
tryptophan	Trp	204.23	$C_{11}H_{12}N_2O_2$
tyrosine	Tyr	181.19	$C_9H_{11}NO_3$
valine	Val	117.15	$C_5H_{11}NO_2$

2.1. Amino Acids Production. In CO_2 capture, the origin of the solvent is of utmost importance. Due to the global problematic of CO_2 emissions, the environmental impact and CO_2 emissions generated during the production and use of the solvent have to be taken into account. Thus, using environmentally friendly and biosourced solvents is essential.

Amino acids are produced by extraction from protein hydrolysates, enzymatic processes, fermentation with aid of microorganism, and chemical synthesis.^{34,43} As an example, Figure 3 shows an overview of the microorganism synthesis. In the production of amino acids via extraction, industrial



Figure 3. Overview of production of amino acids in microorganism synthesis process. Reproduced with permission from ref 34. Copyright 2016 Elsevier.

byproducts and waste such as hair, meat extracts, and plant hydrolysates could be used to produce amino acids in large scale, but it will result in nondesired byproducts such as salts that may have negative impacts on the environment if they are not recovered.³⁵ However, processes such as fermentation methods, chemical synthesis and enzymatic processes are environmentally friendly for production of some amino acids. Furthermore, in the chemical synthesis, CO₂ is used to produce some amino acids such as methionine.^{35,43} However, they need to be optimized operationally and economically to be used in large scale production.

One of the key aspects of amino acids is the ability to react with CO_2 . Some amino acid salts show similar or even better affinities toward CO_2 than alkanolamines due to the presence of identical amino functional groups in their molecules.^{44–46} In the next section, the reaction kinetics of amino acids with CO_2 is explained.

2.2. Kinetics and Reaction. Amino acids in water form a zwitterion, as indicated in eq $1.^{47,48}$ The distribution of the base, neutral, and zwitterion forms at different pH values can be seen in Figure 4.⁴⁹ Based on the reaction mechanism of the CO₂ and amino acids, the base form is the active form. Therefore, amino acids need to be deprotonated to react with CO₂ at neutral pH. Some of the bases considered to neutralize amino acids are MEA, KOH, and NaOH. The deprotonation step is necessary in order to make the amine group reactive toward CO₂.^{50–53}

$$HO_{2}CRNH_{3}^{+} \leftrightarrow H^{+} + ^{-}OOCRNH_{3}^{+}$$
$$\leftrightarrow 2H^{+} + ^{-}OOCRNH_{2}$$
(1)

Due to the similar functional group between amino acids and alkanolamines, the reaction mechanism of the amino acids can be considered as those of alkanolamines.^{54,55} Two mechanisms have been proposed to demonstrate the reaction kinetics of CO_2 with the amine group: the zwitterion mechanism and the termolecular mechanism.^{56–58} The reactions of the zwitterion mechanisms are shown in reactions 2–4, and the termolecular reaction of amino acids with CO_2 is described in reaction 5. The zwitterion mechanism considers



Figure 4. Distribution of glycine at different pH values. Reproduced with permission from ref 50. Copyright 2013 American Chemical Society.

the formation of a zwitterion, and this is followed by the removal of a proton by a base. On the other hand, in the termolecular mechanism, an amine reacts simultaneously with one molecule of CO_2 and one molecule of a base.⁵⁹ There are several studies on the kinetic reaction of amino acids, which show that the zwitterion mechanism explains better the kinetic reaction of amino acids.⁶⁰ However, the termolecular mechanism better fits the reaction of arginine with CO_2 .^{61,62} In Table 2, a list of studies that has been done on the kinetics of amino acids is shown.

Zwitterion reaction mechanism:

$$2^{-}OOCRNH_{2} + CO_{2}$$

$$\leftrightarrow ^{-}OOCRNHCOO^{-} + ^{-}OOCRNH_{3}^{+} \qquad (2)$$

$$^{-}OOCRNHCOO^{-} + ^{-}OOCRNH_{3}^{+} + H_{2}O$$

$$\leftrightarrow ^{-}OOCRNH_{3}^{+} + HCO_{3}^{-} + ^{-}OOCRNH_{2} \qquad (3)$$

Table 2. Summary of the Studies Done on Kinetics of CO₂ Absorption by Amino Acid Salts

name of amino acid	counter ion	experimental technique	condition	kinetic equations	ref
alanine	potassium	stirred cell reactor	T = 293.15 - 313.1 K, concentration = $1 - 3$ M	$K_{\rm ov} = 4.518 \times 10^8 \exp\left(\frac{-3845}{T}\right) \exp(0.5706C_{\rm AAS})C_{\rm AAS}$	63
arginine	potassium	wetted wall column	<i>T</i> = 313–342 K	$K_{2-\text{Arg}} = 2.58 \times 10^{16} \exp\left(\frac{-8645}{T}\right)$	26
proline	potassium	stirred cell reactor	T = 298 K	$K_{\rm ov} = 2.09 \times 10^4 C_{\rm AAS}^{1.08}$	64
proline	potassium	wetted wall column	<i>T</i> = 303–323 K	$K_{2-\text{Pro}} = 2.42 \times 10^{11} \exp\left(\frac{-4384}{T}\right)$	65
proline	sodium	stopped flow	<i>T</i> = 298–313 K	$K_{2-\text{Pro}} = 5.28 \times 10^5 \exp\left(\frac{-1440}{T}\right)$	66
proline	potassium	stirred cell reactor	T = 298 K, concentration = 0.5 kmol/m ³	$K_{2-\text{Pro}} = 3.69 \times 10^{12} \exp\left(\frac{-5211}{T}\right)$	51
glycine	potassium	stirred cell reactor	<i>T</i> = 293–303 K	$K_{\rm ov} = 2.42 \times 10^{16} \exp\left(\frac{-8544}{T}\right) \exp(0.44C_{\rm AAS})C_{\rm AAS}$	39
glycine	potassium	wetted wall column	<i>T</i> = 298–335 K	$K_{2-\text{Gly}} = 1.95 \times 10^{13} \exp\left(\frac{-7670}{T}\right)$	51
glycine	sodium	wetted wall column	<i>T</i> = 303–323 K	$K_{2-\text{Gly}} = 1.95 \times 10^{13} \exp\left(\frac{-7670}{T}\right)$	67
sarcosine	potassium	stirred cell reactor	T = 298 K, concentration = 0.5 kmol/m ³	$K_{\rm ov} = 9.77 \times 10^3 C_{\rm AAS}^{1.41}$	68
sarcosine	potassium	wetted wall column	<i>T</i> = 315–335 K	$K_{2-\text{Sar}} = 6.24 \times 10^{10} \exp\left(\frac{-1699}{T}\right)$	69
sarcosine	potassium	string of disk contactor	<i>T</i> = 298–335 K	$K_{2-\text{Sar}} = 2.6198 \times 10^9 \exp\left(\frac{-915.8}{T}\right)$	68
sarcosine	potassium	stirred cell reactor	<i>T</i> = 298–308 K	$K_{2-\text{Sar}} = 8.67 \times 10^8 \exp\left(\frac{-3127}{T}\right)$	69
taurine	sodium	stopped flow	<i>T</i> = 298–313 K	$K_{2-\text{Tau}} = 5.44 \times 10^{11} \exp\left(\frac{-5780}{T}\right)$	66
taurine	potassium	stirred cell reactor	<i>T</i> = 285–305 K	$K_{2-\text{Tau}} = 3.23 \times 10^{12} \exp\left(\frac{-5700}{T}\right)$	26
taurine	potassium	wetted wall column	<i>T</i> = 323–353 K	$K_{2-\text{Tau}} = 2.7 \times 10^{12} \exp\left(\frac{-6074}{T}\right)$	70

 $-OOCRNH_3^+ + HCO_3^- + -OOCRNH_2$

$$\rightarrow 2^{-}OOCRNH_3^{+} + CO_3^{2-}$$
(4)

Termolecular reaction mechanism:

$$CO_2 + OOCRNH_2 \cdots B \leftrightarrow OOCRNHCOO \cdots BH^+$$
(5)

2.3. Performance of Amino Acids as Solvent in CO₂ Capture. 2.3.1. Organic and Inorganic Salts of Amino Acids. Amino acids themselves have a slow reaction rate and low CO₂ loading in neutral pH. However, when they are neutralized with a base, their performance enhances significantly. Among the known counterions, KOH shows a higher activity and solubility limit than NaOH and LiOH.71 Other counterions that can neutralize amino acids are MEA and methylamino-propylamine (MAPA). When amino acids are neutralized with MEA, the amine group of the amino acid is the active agent to capture CO_{2i} and the amine group of MEA acts as a counterion to deprotonate the amino acids.⁷² Ciftja et al.⁵² studied the deprotonation step of amino acids with MEA. The result showed that the complete neutralization of amino acids did not occur with MEA. In addition, when the amine group of MEA was unable to neutralize the amino acids, the

MEA. Moreover, the carbamate formation of the amino acids was higher than the MEA. Aronu et al.⁷² studied the potential of amine amino acid salts for CO₂ capture. Methylaminopropylamine (MAPA) was mixed with several amino acids. It was found that the organic salts of the amino acid had similar CO₂ absorption properties than MEA at the same concentration. In addition, the amine-amino acid salts were more stable than MEA. However, amino acid salts of an inorganic base, KOH, showed a lower performance in CO₂ absorption than the amine-amino acid salts. Thus, the base added in the solution has an important impact in the final CO₂ absorption capacity of the solvent. In addition, other factors such as temperature of the solution and CO₂ partial pressure also have an effect on the CO_2 loading of amino acids^{73,74} since the solubility of CO2 decreases with the increase in temperature.^{75,76} CO₂ absorption is exothermic in nature and an increase in temperature should decrease the extent of chemical absorption in accordance with Le Chatelier's principle.^{77,78} In addition, with the increase in the CO₂ partial pressure, the CO₂ loading of a solution increases.^{79,80}

carbamate that was formed came from the amine group of

Ho-Jun Song et al.⁴⁴ studied net cyclic capacity of 16 different amino acids in comparison to conventional solvents

amino acids	K_2CO_3 concentration	amino acid concentration (M)	T (K)	acceleration ^a	ref
arginine	35	0.077	322	0.44	59
arginine	35	0.387	322	1.35	59
histidine	35	0.104	322	1.54	59
glycine	30	1	333	22	88
sarcosine	30	1	333	45	88
proline	30	1	333	14	88
^{<i>a</i>} acceleration =					
CO abaa	mition rate in promoted V CO, solution				

Table 3. Performance of Different Amino Acids in Promoting Potassium Carbonate

 CO_2 absorption rate in unpromoted K₂CO₂ solution at same absorption condition

such as monoethanolamine (MEA) and piperazine (Pz). The difference of the initial absorption and desorption rates could be due to the structure of amino acids. It should be noted that as amino acids have a closer carboxyl and CO_2 -bound amino groups and a bulkier substituted side group, the performance of the desorption step improves but the initial absorption rate becomes slower. Initial desorption rates were lower for sterically hindered amino acids = polyamino acids > linear amino acids > cyclic amino acids.⁴⁴

2.3.2. Inorganic Amino Acids Salts Blended with Alkanolamines. In the absorption process, piperazine (Pz) is normally used to enhance the reaction rate and extend the absorption capacity of a solution.^{81,82} Blending of piperazine with several different amino acids has been researched.^{45,83,84} Regarding the properties of the solvent, adding piperazine to the amino acid solvents decreases the surface tension of the solution, which is an important property for some technologies, like those using membrane contactors (a porous membrane is used as the physical barrier between the gas and the solvent), leading to the undesired wetting phenomena.⁴⁴ Furthermore, adding piperazine involves a decrease in viscosity with an increase of temperature and an increase in viscosity with an increase of the piperazine concentration.⁶⁷

Regarding the performance of the amino acids to capture CO_{2} , it was observed that the net cyclic capacities of piperazine-promoted alanine, aminobutyric acid, and serine were at least 25% higher than MEA.⁴⁴ Ramezani et al.⁸⁵ worked on improving the performance of MEA by blending it with potassium lycinate (KLys). A similar trend for the blend of the amino acids and piperazine was observed for the physical properties of MEA and potassium lycinate. CO_2 loading, density, and viscosity of the solvent decreased with the increase of temperature. Additionally, the corrosion rate raised with the increase of amino acid in the solution.

2.3.3. Blends of Amino Acid with Carbonate Salts. Carbonate salts are used extensively for absorption of CO_2 due to their low cost and availability.⁸⁶ The major challenge of carbonate salts is their slow reaction with CO_2 .⁵⁸ Under low temperature and low partial pressure, the absorption kinetics are relatively slow. Several promoters are considered to improve the absorption rate of the carbonate salts.³¹

The most widely studied promoters for potassium carbonate solutions are amines such as MEA, DEA, piperazine, etc.⁸⁷ Recently, blends of amino acids and carbonate salts attracted significant attention. Table 3 shows the performances of different amino acid salts combined with potassium carbonate.^{59,86,88}

2.3.4. Inorganic Amino Acids Blended with Ammonia. Ammonia shows a very good absorption rate and capacity for CO_2 absorption. Yang et al.⁸⁹ studied the vapor loss of NH_3 during the absorption. It was observed that addition of amino acids to the ammonia solvent had a positive effect on the vapor loss. They investigated different amino acids like taurine, sarcosine, and proline. Among them, taurine showed promising results.

Xiang et al.⁹⁰ worked on a blend of ammonia and sodium sarcosinate to enhance the performance of ammonia for CO_2 capture. The results presented that the CO_2 loading of the solution increased from 1.9 to 3.3 times than using only ammonia.

2.3.5. Amino Acids Based Ionic Liquids. Ionic liquids (ILs) are salts and liquids at low and moderate temperatures.⁹¹ They are a large set of chemically different solvents and liquid classes with remarkably different characteristics.²⁴ ILs are promising solvents due to the high CO_2 loading capacity, low cost, nontoxicity, and availability.⁹² An important drawback much discussed in the case of ionic liquids is their high viscosity.⁹³ Recently, amino acid-based ILs have been prepared in order to improve CO_2 sorption capacity. In the amino acids based ILs, the amino group is acting as the anion. Thus, amino acids are the parts that react with CO_2 .²⁵ The kinetic reaction of the amino acids based ILs can be explained via the zwitterion mechanism.^{94,95}

In some cases, the interaction between amino acids and IL liquids is impossible. Therefore, not all amino acids and ILs can be blended.²⁵ Some ILs that have been studied with amino acids are 1-hydroxyethyl-3-methylimidazolium [C₂OHmim],⁹⁴ trihexyl(tetradecyl)phosphonium [P66614],⁹⁶ 1-butyl-3-methylimidazolium [BMIM],⁹⁷ tetrabutylphosphonium [P(C₄)₄],⁹⁸ and 1-aminoethyl-2,3-dimethylimidazolium [aemmim].⁹⁹ Moreover, Jian-Gang Lu et al.^{100,101} synthesized a new amino acid functionalized protic IL, in which the MEA and DEA act as the proton. The structure of the salts is shown in Figure 5.



Figure 5. Molecular structure of [MEA][Gly].

Amino acid-based ILs have been studied for CO_2 capture.⁹⁴ Lv⁹⁴ used a cell reactor to study the absorption rate of the $[C_2OHmim][Gly]$. It was observed that the CO_2 absorption rate decreases with time when approaching the saturation point. Additionally, the stability study of the amino acid-based IL showed that under the condition of absorption and desorption, the solvent is stable and there is no mass loss.⁹⁹ Amino acid-based ILs have higher CO_2 loading (i.e., 0.2–0.8 mol of CO_2/mol of IL) in comparison to the MEA (i.e., 0.5 mol of CO_2/mol of amine) and inorganic salts of amino acids



Figure 6. Typical chemical absorption system for CO₂ capture. Reproduced with permission from ref 11. Copyright 2016 Elsevier.



Figure 7. Overall mass transfer coefficient-temperature relationships for all the data reported in the literature for the conventional systems.

(i.e., 0.1–0.7 mol of CO_2/mol of solvent). In contrast to the positive aspects of the ILs, it has to be mentioned that at high concentrations, amino acids start to salt out.¹⁰² Amino acid ILs are good candidates to replace the conventional solvents. However, there is not sufficient data about their properties and performance. Table S1 shows several amino acid ionic liquid capacities as reference.

3. CURRENT TECHNOLOGY USING AMINO ACID SOLUTIONS FOR CO₂ CAPTURE

Conventional technology such as gas absorption/desorption columns as well as advanced separation systems such as membrane gas absorption and membrane gas separation have been studied with amino acid-based solutions as the absorption solvent.^{103,104} The overview of these technologies and the

implications related to the use of amino acids are presented in this section.

3.1. Absorption Columns. Gas absorption columns have been commercialized and used in the industry for decades.⁴¹ There are different designs of gas liquid absorption columns, i.e., wetted wall columns, packed columns, bubble columns. Wetted wall column are intensively used for mass-transfer research.¹⁰⁵

In the columns, the overall absorption of carbon dioxide can be divided into two steps, physical and chemical absorption. The driving force is the gradient of the chemical potential, which can be translated to the concentration gradient.^{106,107} Equations 6 and 7 show the overall flux of CO₂ in the liquid and gas sides. In the equations, k_g and k_l are the mass transfer coefficients for the gas and liquid films, respectively.²⁸



Figure 8. Liquid phase boundary layer resistance: (a) nonwetted mode and (b) wetted mode.

$$N_{\rm CO_2,g} = k_{\rm g} (P_{\rm CO_2,bulk} - P_{\rm CO_2,i})$$
(6)

$$N_{\rm CO_2,l} = ak_{\rm l}(C_{\rm CO_2,i} - C_{\rm CO_2,bulk}) = k_{\rm g}'(P_{\rm CO_2,i} - P_{\rm CO_2,bulk}^*)$$
(7)

Figure 6 shows a typical chemical absorption system. In absorption columns, the CO_2 is removed by reaction between solvent and CO_2 . Then it goes to desorption column in which CO_2 is removed by an increase in the temperature.¹¹ The energy penalty and solvent losses associated with this technology have addressed some of the recent research toward the optimization of the absorption/desorption process by using other solvents that require less energy for regeneration.

Amino acid solutions are an example since they require less regeneration energy, being less corrosive and having fewer solvent losses during the process. Several studies have been done on the performance of the amino acid solvents with wetted wall columns.^{27,51,58,59,65,89,107–110} An increase of the amino acids concentration and temperature has a significant impact on the overall mass transfer coefficient depending on the type of amino acids.

Figure 7 shows the overall mass transfer coefficient versus temperature for all the research that reported the mass transfer coefficient. The purpose of this figure is to show the state of the art of using amino acid-based solutions for CO_2 capture for conventional systems, such as wetted wall columns, bubble columns, etc. The cloud of points is represented regardless solvent concentration, gas feed concentration or the system fluid dynamics.^{27,60,63,65,73,78,109–113}

Amino acid-based solvents for CO₂ capture have been studied in wetted wall columns^{27,60,78} and stirred cells.^{63,110,111,113} Due to the high viscosity of amino acid solutions, increasing temperature has a positive effect on the diffusion of molecules within the liquid phase. Nevertheless, a good mass transfer at lower temperature can also be achieved as shown by Shen et al.¹¹³ The highest mass transfer coefficient in Figure 7 was achieved by Knuutila et al.¹¹⁴⁻¹¹⁶ and Kim et al.⁶³ evaluated the physical properties and absorption rate of potassium alanine and potassium glycine with a stirred cell reactor. Potassium glycine had a higher absorption rate due to the fact that potassium alanine is a steric hindrance amino acid. However, in the case of potassium alanine, the total energy consumption was less than the potassium glycine because of the faster desorption rate of the potassium alanine solution. Knuutila et al.^{114,115} studied the performance of 3.5 M potassium sarcosine (KSAR) and 30 wt % of MEA with a pilot plant of the conventional process. The simulation and experimental results showed a high absorption rate for the potassium sarcosine. Based on the condition of the process, it

was found that the energy consumption of the potassium sarcosinate was higher than MEA. Thus, further optimization for potassium sarcosinate solvent is needed to be used in the conventional process.

BASF Co.²⁵ commercialized three solvents for acid gas removal which are called alkalacid: sodium alanine, diethyldimethylglycine, and sodium phenolate. Sodium alanine solvent removes hydrogen sulfide (H₂S) and carbon dioxide (CO₂) from the flue gas. On the other hand, diethyldimethylglycine selectively removes H₂S from gases containing CO₂. Alkalacid solutions were found to be less selective than tripotassium phosphate for acid gas removal. Alkalacid solutions are noncorrosive, which is one of the main advantages since process equipment require less special material for construction.²⁵

It needs to be mentioned that CO_2 capture with a high concentration of amino acids and high CO₂ loadings leads to precipitation of the amino acid or (bi) carbonates or a mixture of both.¹¹⁷ In the absorption process, the precipitation could have a positive effect since eqs 2-4 would be displaced to the production of more carbonate. However, the presence of a solid may lead to a severe operating process and certain adjustment of the equipment is required to successfully design a viable process. For example, the absorber should be able to handle slurries.^{118,119} A new process called DECAB has been proposed.¹²⁰ The design of the process is based on the precipitation of the amino acids solution during the absorption process. A separator is used after the absorption column to separate the precipitates, and then they are recycled back to the absorption column. In the process, the total energy consumption is reduced. Moreover, two solvents are used, MEA and the potassium salt of taurate. Usage of taurate salt reduces the overall energy consumption by 15% in comparison to the MEA baseline. Therefore, the precipitation of amino acid salts can be used to increase the CO₂ capture capacity of the process and reduce the energy consumption.^{120,12}

Siemens Co.²⁹ also developed a process based on amino acid salt solutions for CO_2 capture using packed columns. The operation conditions of the process are adjusted to avoid crystallization of the amino acids salts. Therefore, the temperature of the solution is above the crystallization temperature. It was observed that the degradation of the amino acids (thermal degradation, oxidation) was less than 1%. In addition, 2.7 MJ/kg of CO_2 captured was required for solvent regeneration.

3.2. Membrane Technology. Membrane technology combined with amino acid solutions has appeared very recently, and it is taking an important position in the development of novel processes for CO_2 capture. Concretely,

Review



Figure 9. Comparison of the overall mass transfer coefficient based on different temperatures and liquid velocity for sodium glycinate: (a) T = 303.15 K, concentration 0.5 M. Adapted with permission from ref 126. Copyright 2015 Elsevier. (b) Concentration of solvent = 0.5 M, gas flow rate = 100 cm³/min, liquid flow rate = 25 mL/min. Adapted with permission from ref 60. Copyright 2015 Elsevier.

membrane gas absorption (MGA) uses membrane contactors as the device to allow the contact between the gas phase and the absorption solvent. This technology provides a highly specific surface area, independently controllable gas and liquid flow rates, a compact and energy efficient separation unit, and a linear scale-up design.³²

Unlike other contactors in which there are only two mass transfer resistances caused by the gas and the liquid phases, the presence of the membrane includes the membrane itself as an additional resistance to the mass transfer, i.e., gas phase boundary layer resistance, membrane diffusion resistance, and liquid phase boundary resistance.¹⁰⁵ Three resistances can be seen in Figure 8a. Compared to the liquid phase boundary layer resistance, the gas phase boundary layer resistance is relatively small and may be neglected. Normally, porous membranes are used. Thus, the resistance of the membranes may be also relatively small compared to the liquid phase boundary layer resistance as long as the porous membrane is not wetted by the absorption liquid.¹²² However, if the pore gets wet, the membrane resistance becomes very noticeable. Figure 8b shows the boundary layer resistances when

membrane pores are wetted.¹²³ The wettability of the membrane pores depends on the type of materials used for the fibers of the membrane. There are two types of membrane, hydrophobic or hydrophilic. To avoid this problem, hydrophobic are better options for the gas absorption process using aqueous solutions. Additionally, solvents with high surface tension can be utilized.¹²⁴

The transmembrane flux for the three regions (i.e., gas, membrane and liquid) can be expressed by 32

$$N = k_{\rm g}(P_{\rm gb} - P_{\rm gm}) = k_{\rm m}(P_{\rm gm} - P_{\rm lm}) = k_{\rm l}(C_{\rm lm} - C_{\rm lb})$$
(8)

where k_{g} , k_{m} , and k_{l} are the three individual mass transfer coefficient for the gas, membrane, and liquid phases, respectively. *P* and *C* are the gas partial pressure and liquid concentration. The subscripts gb, gm, lm, and lb represent gas bulk, gas-membrane interface, liquid-membrane interface, and liquid bulk, respectively.

In the membrane gas absorption process, typically an increase in the velocity of the liquid solvent increases the overall mass transfer coefficient, which can be seen in Figure



Figure 10. Overall mass transfer coefficient-temperature relationships for all the data reported in the literature for the membrane contactors.

9.¹²⁵ In the case that the gas phase has a significant resistance to mass transfer, the gas velocity would have the same effect on the overall mass transfer coefficient. In addition, with the increase in purity of the gas, the overall mass transfer coefficient would also increase.¹²⁶ However, the performance of the amino acid solution may decrease at higher concentrations of amino acid due to an increase of the viscosity of the solution. This would lead to higher mass transfer resistance in the liquid phase.³²

Figure 10 shows the overall mass transfer coefficient versus temperature for all the research that reported the mass transfer coefficient. In Figure 10, the state of the art of using amino acid-based solutions for CO₂ capture for membrane contactors is presented. Those clouds of points are represented regardless of solvent concentration, gas feed concentration or the system fluid dynamics¹²⁷⁻¹³⁰ It can be observed that, in each type of the membrane, the mass transfer coefficient increases with the increase of the temperature, regardless of other operational conditions. Among all the studies, de Montigny et al.¹³¹ have achieved the highest mass transfer coefficient. They have studied two types of the membranes: polypropylene (PP) and polytetrafluoroethylene (PTFE) for CO₂ capture. The maximum mass transfer coefficient was observed with the PTFE membrane contactor, having less surface area in comparison to the PP membrane contactor. Membrane porosity presented a significant effect on the mass transfer coefficient.

In membrane contactors, the percentage of porosity of the membrane, membrane material and membrane thickness could also effect the mass transfer coefficient.³⁶ As it was mentioned before, hydrophobic membranes present better mass transfer for CO_2 capture since the amino acid solvents are prepared in aqueous solutions.¹²⁴ However, there is still the possibility of wetting in long-term use.¹³² In Figure 10, it can be seen that membrane contactors also present a good mass transfer at low temperature. Since using membrane contactors is still in an early stage of development, there is large room for increasing the mass transfer coefficient by optimizing the operating conditions as well as the contactor and membrane characteristics.

Yan et al.¹³³ studied the wetting phenomena of different solvents such as MEA, 3,4-methylenedioxy-N-ethylamphet-

amine (MDEA), and potassium glycinate. The results presented better performance for potassium glycinate due to higher surface tension. Aqueous potassium glycinate solution had a lower potential of membrane wetting after a continuously steady operation.

In Figure 11, the membrane gas absorption system for CO_2 absorption is shown. Temperature is one effective factor in



Figure 11. CO_2 gas absorption and the stripping system via membrane contactors. Reproduced with permission from ref 125. Copyright 2010 John Wiley and Sons.

improvement of the process as a rise of the temperature increases the CO₂ permeation through the membrane in the absorption process. The effect of the temperature on the process performance when using MEA and sarcosine has been studied.¹³⁴ As it was mentioned before, solutions of amino acids salts have a more stable carbamate and (bi)carbonate products in solution; therefore, more energy is needed for desorption of CO₂. This leads to some limitation in using this process at a large scale. To reduce the cost and energy consumption and furthermore to utilize the CO₂ instead of storing it under the ground, other technologies such as crystallization of the carbonate salts by using membrane contactors are under research.^{135–137}

4. CONCLUSIONS

Amino acids are potential candidates for CO_2 capture. Current studies mainly focus on the thermodynamic properties and kinetic reaction of amino acids with CO_2 . Along with the

determination of the properties of the amino acids, the performance of several amino acids in small-scale technologies have been studied. Amino acids performance is similar or in some cases better than the conventional solvents. However, to have a better understanding of the chemical process of the amino acids with CO_2 , further studies on solubility, viscosity, and surface tension are required.

From the technology point of view, amino acids show good characteristics for absorption of CO_2 . The use of amino acids in conventional columns requires an extensive study and optimization to improve the performance and energy consumption of the process. Additionally, when the conventional process is adjusted with properties of amino acids, i.e., precipitation of amino acids during the process, the energy consumption decreases and the CO_2 capacity of the solvent increases. Furthermore, amino acids with gas membrane absorption show a good performance, indicating that the use of amino acids for CO_2 capture is a suitable alternative, even though it is still in an early stage of development.

ASSOCIATED CONTENT

S Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.iecr.9b01793.

Table S1, amino acid ionic liquids capacities (as Supporting Information for section 2.3.4) (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: patricia.luis@uclouvain.be.

ORCID [®]

Vida Sang Sefidi: 0000-0001-9972-0048

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest. **Biographies**



Patricia Luis is a Professor at UCLouvain, Belgium, in the division of Materials and Process Engineering. Her main research interests address CO_2 capture and recovery using membrane contactors and process intensification in the chemical industry by applying advanced separation technology and exergetic and environmental analyses. She authored more than 100 publications in these fields with more than 2000 citations. She is member of the Editorial Boards of the *Journal of*

Chemical Technology and Biotechnology, Separation and Purification Technology, Clean Technologies, C-Journal, and Frontiers in Chemical Engineering–Separation Processes. She has authored the book Fundamental Modelling of Membrane Systems, published by Elsevier in 2018.



Vida Sang Sefidi is a Ph.D. student at the UCLouvain, Belgium, in the division of Material and Process Engineering. During her Master studies, she became interested in the in the green energies and activities related to the reduction of emission of greenhouse gases. She did her Master's thesis on the production of hydrogen. In 2018, she started her Ph.D. thesis on CO_2 capture using amino acid-based solutions and membrane technology, aiming at converting the CO_2 into a valuable end product for further use in industry.

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ABBREVIATIONS

 CO_2 = carbon dioxide MEA = monoethanolamine DEA = diethanolamine KGly = potassium glycinate IL = ionic liquidsKOH = potassium hydroxide NaOH = sodium hydroxide LiOH = lithium hydroxide MAPA = methylamino-propylamine Pz = piperazine KLys = potassium lycinate NH₃ = ammonia [aemmim] = 1-aminoethyl-2, 3-dimethylimidazolium [C₂OHmim] = 1-hydroxyethyl-3-methylimidazolium [P66614] = trihexyl(tetradecyl)phosphonium [BMIM] = 1-butyl-3-methylimidazolium [P(C4)4] = tetrabutylphosphonium Ktau = potassium taurate NaGly = sodium glycinate K asparaginate = potassium asparaginate K prolinate = potassium prolinate KSar = potassium sarcosinate WWC = wetted wall column MGA = membrane gas absorption MDEA = 3,4-methylenedioxy-*N*-ethylamphetamine

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