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SCREENING OF THE AMINO ACID-BASED SOLVENTS FOR CARBON DIOXIDE ABSORPTION

BY

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Abstract. Post-combustion separation and capture of CO_2 from flue gases resulting from the burning of fossil fuels is a challenging technique due to the low pressure and low concentration of carbon dioxide, the impurities in the flue gases and the cost of regenerating the absorber. Researchers have developed various CO_2 capture technologies such as absorption, adsorption, and membrane separation cryogenic technology. Of these, absorption was the most widely used. A wide variety of organic and inorganic substances have been proposed as solvents for CO_2 uptake. But, these solvents have some major disadvantage such as: low absorption rate for K_2CO_3 solutions, high energy consumption for the amine solutions, high volatility and toxicity of ammonia. These can be solved by using another selective solvent for carbon dioxide. This paper provides information available in the literature on current solvents and the potential for amino acid salts to be used as absorbent for CO_2 capture.

Keywords: acid gas, chemisorption, energy efficiency, equimolar blend, greenhouse gases.

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1. Introduction

The industrial revolution meant increasing mankind's dependence on fossil fuels. The main elements of fossil fuels are carbon and hydrogen. Through the process of burning fuels, the heat mainly used for energy is released. The amount of carbon dioxide in the gas phase depends on the carbon content of the fuel. Over time, it has been observed that the Earth's temperature depends directly on the concentration of CO_2, and an increase in global average temperature has been recorded since the early 1900s. An increase of a few degrees Celsius is expected to significantly affect the climate. As a result, there is an urgent need to address the issue of high CO_2 emissions.

Carbon capture and sequestration (CCS) is the only technology available method that can reduce the concentration of CO_2 in the atmosphere due to the burning of fossil fuels (Ayoub *et al*., 2018; Paul and Thomsen, 2012; Rahman *et al*., 2017; Sang Sefidi and Luis, 2019; Thee, 2013; Zhang, 2021). CCS is a technology that can only be used only for stationary sources, which are responsible for 50% of total emissions (de Coninck *et al*., 2018; Edenhofer *et al*., 2011; Hoegh-Guldberg *et al*., 2018; Tschakert *et al*., 2018). But like any new technology, CCS cannot be widely applied due to its direct cost constraints. In addition, there are not enough data on the long-term health effect of this process for humanity. Carbon dioxide capture can be achieved by several methods, some tested on an industrial scale, some on pilot scale, and others under development and evaluation. The choice of a technology for a particular method of capturing carbon dioxide depends on many factors, such as: the partial pressure of CO_2 in the gas flow; the required recovering degree of CO_2; sensitivity to impurities; desired purity of carbon dioxide; process operating costs and the cost of additives needed to overcome dirt and corrosion; environmental impact etc. Chemical absorption systems seem to be the best short-term option for capturing CO_2 from gaseous streams (Zhang *et al*., 2021).

2. Conventional solvents for carbon dioxide capture by absorption

The process of chemical absorption involves the reaction of carbon dioxide with a chemical solvent, resulting an intermediate compound. This intermediate can be decomposed by heating, producing the original solvent and a CO_2 stream that is directed to other technologies (Lawal *et al*., 2011; Zhang *et al*., 2021). In absorption processes, a CO_2-containing waste gas stream enters the bottom of an absorber. The solvent enters the top of the absorber and comes into contact with the gas (counter current flow). Thus, CO_2 is removed and the treated gas exits through the top of the column. CO_2 is absorbed and the CO_2-enriched solvent exits at the bottom of the absorber (Fig. 1).

The selectivity of this separation process is relatively high and there is a possibility of obtaining a relatively pure CO_2 flux. Although feasibility studies show that it will remain competitive in the future, this technology has many problems, depending on the solvent chosen for separation: high cost, toxicity, degradation and solvent evaporation, low absorption rate etc. (Ayoub *et al*., 2018; Lawal *et al*., 2011; Leung *et al*., 2014; Mondal *et al*., 2012; Thee, 2013).

Solvent selection plays an important role in the success of the CO_2 absorption process because the performance of the CO_2 separation process depends on the behaviour of the solvent. A number of factors must be considered when choosing an absorbent solution: the solubility of the gaseous component in the liquid phase, the reactive properties of the gas and the absorbent (Lawal *et al*., 2011; SINTEF, 2009). CO_2 is considered an acid gas because, when it is absorbed by a water-based solvent, an acid solution will form. Thus, an alkaline absorbent is desirable for the absorption of acid gas. Alkaline solutions allow the neutralization of any acid formed by the absorption of gas, thus increasing the amount of gas that can be absorbed for a certain volume of liquid solvent.

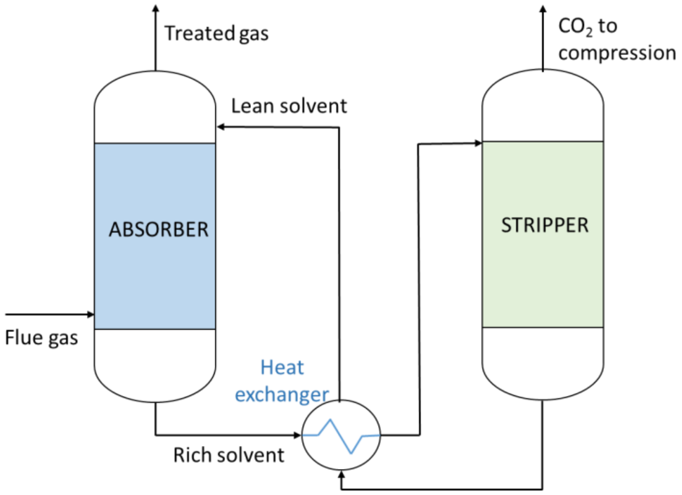


Fig. 1 − Schematic of a basic chemical absorption process for CO_2 capture.

The process of chemical absorption of carbon dioxide is applied in several variants:

- absorption in potassium carbonate solutions;
- absorption in amine solution;
- absorption in aqueous ammonia solutions;
- absorption in ionic liquids.

Potassium carbonate (K_2CO_3) has been chosen as a system to be used in high pressure CO_2 capture applications and is the main method practically used. Aqueous potassium carbonate is a good solvent for capturing carbon dioxide due to its low regenerative energy, low degradation and low corrosivity (Borhani *et* *al*., 2015; Ramazani *et al*., 2016; Shen *et al*., 2013; Wang *et al*., 2017). The main challenge of the CO_2 absorption system in potassium carbonate solutions is the slow reaction rate compared to the amine solution absorption system, especially in post-combustion conditions: low temperature and low CO_2 partial pressure (Grimekis *et al*., 2019; Harja *et al*., 2021; Hu *et al*., 2017b; Li *et al*., 2019; Skurygin and Poroyko, 2020; Wang *et al*., 2017). The activation of K_2CO_3 solutions appears to be an effective way to improve this solvent performance (Devries, 2014; Grimekis *et al*., 2019; Harja *et al*., 2021; Hu *et al*., 2016).

The absorption in amine solutions has been used in natural gas industry in the last decades to remove CO_2 (Godin *et al*., 2021; Tataru -Farmus *et al*., 2007). Monoethanol amine (MEA) is the most widely used solvent, due to both its fast absorption rate and its low cost (Lang *et al*., 2017; Leung *et al*., 2014). However, MEA and others amines have some disadvantages, such as: high energy loss for solvent regeneration (over 80 % of the total energy consumption), high degradation rate and corrosivity in presence of O_2 (Godin *et al*., 2021; Wu *et al*., 2020). Another problem that must be addressed when using amines as a solvent for carbon dioxide is related to their toxicity. A solvent with high toxicity is not the best choice because it involves environmental issues (Koronaki *et al*., 2015; Lang *et al*., 2017).

The process of absorbing carbon dioxide from flue gases into ammonia solutions has been reported to be very effective (Kozak *et al*., 2009; Molina and Bouallou, 2015, 2017; Rivera-Tinoco and Bouallou, 2010; Yeh *et al*., 2005). The advantages of this technology are lower regeneration energy consumption and fertilizer production. However, the problem of ammonia loss is the main obstacle to the widespread application of ammonia absorption technology. Due to the volatile nature of ammonia, it is necessary to cool the flue gases and to reduce its ammonia volatilization. As a result, this process involves high energy consumption and costs (Jilvero *et al*., 2014; Liu *et al*., 2009; Molina and Bouallou, 2015; Valenti and Bonalumi, 2018).

As a result of the requirements for more environmentally friendly, but also more sustainable techniques, a category of solvent has developed: ionic liquids (IL). IL could be classified into two main groups: conventional and functionalized. CO_2 is absorbed in the conventional IL by the means of a physical process, at low capacity and selectivity, while the structure of functionalized IL is modified to permit a chemisorption of CO_2 at high capacity, rate and selectivity (Farsi and Soroush, 2020). The advantages of carbon dioxide capture technology in ionic liquids are due to its remarkable properties, such as: negligible volatility, high thermal stability, non-flammability, solvation and high solubility of CO_2 (Aghaie *et al*., 2018; Bai *et al*., 2017; Hasib-ur-Rahman *et al*., 2010; Ramdin *et al*., 2012; Sánchez, 2008; Wappel *et al*., 2010; Zhang *et al*., 2016). The number of ILs that can be used al absorption solvents is very large; the imidazolium class is most widely investigated and reported in the literature (Janiczek *et al.*, 2012). However, the use of ionic liquids as solvents for carbon dioxide presents 2 major challenges: high viscosity and high production price (Aghaie *et al*., 2018; Brennecke and Gurkan, 2010; Zareie-kordshouli *et al.*, 2016; Wu *et al*., 2020).

A special category of absorbents are solvent blends. These mixtures are usually composed of two individual solvents of different compositions. The advantage of mixed solvents would be that they can extract favourable performance from the initial solutions, eliminating the disadvantages. In the literature there are studies on blends based on potassium carbonate – amines (Behr *et al*., 2011; Cullinane and Rochelle, 2006; Devries, 2014; Harja *et al*., 2021; Ramezani *et al.*, 2018; Tang *et al*., 2011; Thee, 2013), potassium carbonate - ionic liquids (Ochedi *et al*., 2020; Sistla and Khanna, 2015), mixtures of amines (Adeosun and Abu-Zahra, 2013; Ansaloni *et al*., 2019; Chen *et al*., 2016; Du *et al*., 2016; Luo *et al*., 2017; Nwaoha *et al*., 2018; Pacheco *et al*., 2000; Perinu *et al*., 2017), amines and ionic liquids (Haider *et al*., 2016; Xiao *et al*., 2019; Wu *et al.*, 2020).

Recent literature concludes that the solvents developed so far have many disadvantages and there is a need for research and development of new technologies and new solvents for carbon capture.

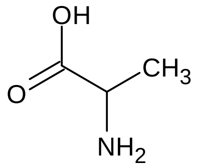
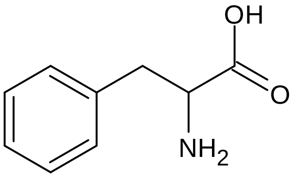
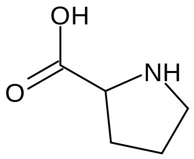
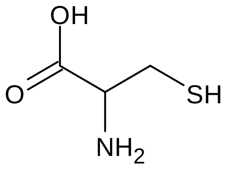
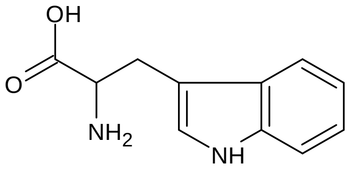
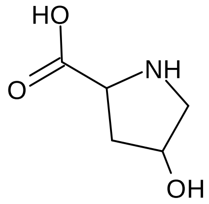
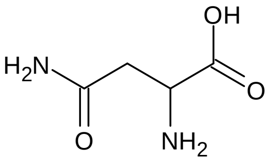
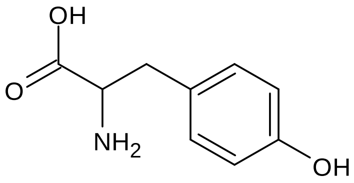
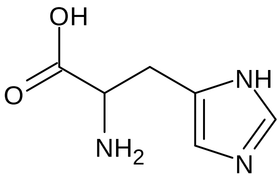
3. Amino acid salt (AAS) as CO_2 absorption solvent

Because the amine group is the main reason for the use of amines as a solvent for CO_2 separation, specialized studies have focused on other types of solvents with this functional group, but have better properties than amines (Holst *et al*., 2009; Sanchez-Fernandez *et al*., 2013; Sang Sefidi and Luis, 2019; Song *et al*., 2012). Amino acids are amphiprotic species, organic compounds that contain one or more amino groups (—NH_2) and one or more carboxyl groups (—COOH). CO_2 absorption using amino acid-based solvents is a bio-mimetic treatment of the CO_2 capture process, due to its resemblance to the binding of CO_2 in blood by proteins, such as hemoglobin (Lerche *et al*., 2009). According to the nature of the radical on which the two functional groups are grafted, amino acids can be aliphatic, aromatic and heterocyclic (Table 1).

In water, amino acids form a zwitterion. Based on the reaction mechanism of CO_2 and amino acids, the basic (alkaline) species is the active form. Therefore, amino acids must be deprotonated to react with CO_2 at neutral pH. Deprotonation of amino acids is achieved by adding an equimolar amount from a strong base (Ayoub *et al*., 2018; Li *et al*., 2019; Paul and Thomsen, 2012; Shen *et al*., 2015; Thee *et al*., 2014; Zarei *et al*., 2020; Zhang *et al*., 2018). Some of the bases considered to neutralize amino acids are KOH, NaOH and LiOH. Research has suggested that deprotonated amino acids with potassium hydroxide are more reactive than those deprotonated with sodium hydroxide (Holst *et al*., 2012; Lim *et al*., 2012; Zarei *et al*., 2020; Zhang *et al*., 2018). Majchrowicz (Majchrowicz, 2014) showed that the reactivity of the potassium salt of L-prolinate was higher than that of its sodium salt in reaction with CO_2.

Deprotonation of amino acids can also be achieved with amine solutions. Literature reports the use of monoethanolamine MEA and methylamino-propylamine MAPA solutions for this stage of neutralization. In the case of neutralization with MEA, the amine group of the amino acid is the active agent for CO_2 capture, and the amine group of the MEA acts as a counterion to deprotonate the amino acids. The results are better for CO_2 absorption in the amine-amino acid salts than absorption in acid salts of an inorganic base, like KOH (Ciftja *et al*., 2014; Knuutila *et al*., 2011).

Table 1
Classification and structure of some amino acids

aliphatic	aromatic	heterocyclic
Alanine $C_3H_7NO_2$ 	Phenylalanine $C_9H_{11}NO_2$ 	Proline $C_5H_9NO_2$ 
Cysteine $C_3H_7NO_2S$ 	Tryptophan $C_{11}H_{12}N_2O_2$ 	Hydroxyproline $C_5H_9NO_3$ 
Asparagine $C_4H_8N_2O_3$ 	Tyrosine $C_9H_{11}NO_3$ 	Histidine $C_6H_9N_3O_2$ 

Regarding the absorption of CO_2 in different amino acid salt solutions, Table 2 lists some of the experimental studies. Thus, the most commonly used amino acids are taurine, alanine, sarcosine and proline as potassium or sodium salts.

Table 2
Amino Acid Salts used in experimental studies

Compound	AAS solutions	Reference
Arginine	Potassium salt	(Holst *et al*., 2009; Song *et al*., 2012)
Glycine	Potassium salt	(Bao *et al*., 2022; Hallenbeck *et al*., 2015; Kumar *et al*., 2003; Li and Rochelle, 2011; Song *et al*., 2012)
	MEA / AMP	(Ciftja *et al*., 2014)
Taurine	Potassium salt	(Hallenbeck *et al*., 2015; Kumar *et al*., 2003; Li and Rochelle, 2011; Majchrowicz *et al*., 2006; Song *et al*., 2012)
	Sodium salt	(Majchrowicz *et al*., 2006)
	Lithium salt	(Majchrowicz *et al*., 2006)
	MEA/AMP	(Ciftja *et al*., 2014)
Alanine	Potassium salt	(Holst *et al*., 2009; Li and Rochelle, 2011; Lim *et al*., 2012; Majchrowicz *et al*., 2006; Song *et al*., 2012)
	Sodium salt	(Majchrowicz *et al*., 2006)
	Lithium salt	(Majchrowicz *et al*., 2006)
	MEA/ AMP	(Ciftja *et al*., 2014)
Aminobutyric acid	Potassium salt	(Majchrowicz *et al*., 2006; Song *et al*., 2012)
	Sodium salt	(Majchrowicz *et al*., 2006)
	Lithium salt	(Majchrowicz *et al*., 2006)
Methyl alanine	Potassium salt	(Song *et al*., 2012)
Serine	Potassium salt	(Song *et al*., 2012)
	MEA/AMP	(Ciftja *et al*., 2014)
Cysteine	Potassium salt	(Song *et al*., 2012)
Proline	Potassium salt	(Hallenbeck *et al*., 2015; Lim *et al*., 2012; Paul and Thomsen, 2012; Shen *et al*., 2015; Song *et al*., 2012)
	MEA / AMP	(Ciftja *et al*., 2014)
Pyroglutamic acid	Potassium salt	(Song *et al*., 2012)
Asparagine	Potassium salt	(Song *et al*., 2012)
Glutamine	Potassium salt	(Song *et al*., 2012; Zarei *et al*., 2020)
Diglycine	Potassium salt	(Song *et al*., 2012)
Lysine	Potassium salt	(Hallenbeck *et al*., 2015; Shen *et al*., 2015)
6-aminohexanoic acid	Potassium salt	(Holst *et al*., 2009; Majchrowicz *et al*., 2006)
	Sodium salt	(Majchrowicz *et al*., 2006)
	Lithium salt	(Majchrowicz *et al*., 2006)
Glutamic acid	Potassium salt	(Holst *et al*., 2009)
Methionine	Potassium salt	(Holst *et al*., 2009)
Sarcosine	Potassium salt	(Holst *et al*., 2009; Knuutila *et al*., 2011; Li and Rochelle, 2011)
	Sodium salt	(Li and Rochelle, 2011)

Additionally, in the experimental studies, the absorption performance of amino acid salts is compared to MEA as the most important conventional amine and the authors claimed that the solutions of the amino acid salt can be considered as an attractive solvent. Lysine, proline, glycine, alanine and sarcosine are reported as having a good reaction constant with carbon dioxide, larger than that of amines. Some of the AAs properties like ionic strength, large CO_2 solubility, high surface tension and the cation used are shown to be important factors influencing the reactivity of amino acids (Ramezani *et al*., 2020).

Although amino acids are more expensive than amines, they have advantages due to their physical and chemical properties: low volatility, low toxicity and resistance to degradation. For these reasons and because they are not harmful for the human health, amino acids can be selected for carbon dioxide absorption. Nevertheless, the main disadvantages of amino acid salts are: solvent corrosive nature and high energy needed for regeneration. Amino acid salts precipitate at high concentrations or high CO_2 loading during CO_2 absorption which causes damage to absorption plant and also reduces mass transfer (Ramezani *et al*., 2020).

In the latest years, different blends of AASs and other liquids to form a new absorbent mixture has gained attentions (Ahmed and Wiheeb, 2020; Chen *et al*., 2018; Hu *et al*., 2018; Kasturi *et al*., 2021; Li *et al*., 2019; Thee *et al*., 2014). The role of AAS in a blended solution is to enhance the absorption or the desorption performance. The results showed a great potential to apply the blended solution in industry. Proline and sarcosine have better promoting effects compared to MEA in alkaline solution solvents (Aronu *et al*., 2011; Hu *et al*., 2017a).

3. Conclusions

The recent research avenues has investigated various new solvents, in order to identify the absorbent with the lowest energy consumption for regeneration and which would eliminate the difficulties encountered for amine-based CO_2 removal systems. Among them, aqueous solutions of alkaline salt of amino acids can offer a promising alternative. In general, amino acid salt solutions can be characterized by greater stability to oxidative degradation, a good level of biodegradability, low toxicity, water-like viscosities. Although amino acids (salts) are found in a number of industrial applications, information about these systems is quite limited. Based on the study, potassium salts of Sarcosine, Lysine, and Proline can to be considered as attractive candidates for CO_2 absorption. On the other hand, it is possible that one of the amino acids already researched has the potential to absorb carbon dioxide in a new solvent configuration. For future work, the performance of an amino acid potassium salt as promoters in potassium carbonate solvents, for industrial CO_2 capture will be evaluated.

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EVALUAREA SOLVENȚILOR PE BAZĂ DE AMINOACIZI PENTRU ABSORBȚIA DIOXIDULUI DE CARBON

(Rezumat)

Separarea post-combustie şi captarea CO_2 din gazele de ardere rezultate din arderea combustibililor fosili este o tehnică dificilă din cauza presiunii scăzute şi a concentrației scăzute în dioxid de carbon, a impurităților din gazele de ardere și a costului. Cercetătorii au dezvoltat diverse tehnologii de captare a CO_2, cum ar fi criogenia, adsorbție, absorbția și separare prin membrană. Dintre acestea, absorbția este cea mai utilizată. O mare varietate de substanțe organice și anorganice au fost propuse ca solvenți pentru absorbția CO_2. Însă, acești solvenți au unele dezavantaje majore, cum ar fi: viteza scăzută de absorbție pentru soluțiile de K_2CO_3, consum mare de energie pentru soluțiile de amine, volatilitatea și toxicitatea ridicate în cazul soluțiilor de amoniac. Acestea pot fi rezolvate folosind un alt solvent selectiv pentru dioxid de carbon. Această lucrare oferă informații disponibile în literatura de specialitate despre solvenții actuali și sărurile de aminoacizi, potențial a fi utilizate ca absorbant pentru captarea CO_2, ca solvent individual sau în amestec.