Review

Enhancing CO_2 Capture Utilizing Deep Eutectic Solvents

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Enhancing CO_2 Capture Utilizing Deep Eutectic Solvents

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Abstract: Both natural gas production and fossil fuels production are the main sources to most of the energy consumption, this gas presented a series of impurities, i.e. CO_2, which needed to be eliminated in order to prevent several concerns as the corrosion of equipments, greenhouse gas emissions and others. It is thus clear, that the development of efficient CO_2 capture and storage processes are important to reduce both CO_2 production and its contribution to global warming. CO_2 can be capture from gas streams by three technologies: absorption, adsorption and membranes, however, they have some challenges in its utilization to be resolved, and some groups of scientist try to resolve it by the inclusion of deep eutectic solvents in them. In the present work, the most recent developments (2024 year) in CO_2 capture using deep eutectic solvents (DESs) jointly to absorption, adsorption or membrane-based technologies have been reviewed.

Keywords: CO_2 capture; deep eutectic solvents; absorption; adsorption; membranes; environment; global warming

1. Introduction

Carbon dioxide is the main green house gas that is leading to changes in climate and in the global warming, thus, its elimination, or at least decreasing its concentration, from gas streams is of the utmost importance. In relation to CO_2 capture, chemical absorption processes using aqueous-amines based systems are the most used and effective [1]. Despite the above, this technology is not without its drawbacks which are often related to water content in the solvent, hindering the own solvent regeneration [2,3]. Thus, authors used deep eutectic solvents (DESs) to improve the performance in the capture of CO_2 utilizing the absorption procedure.

Since the introduction of DESs to Chemistry [4], the properties of these remarkable chemicals had opened their utilization in many fields of interest [5]. With many variations in their formulations, DESs are often composed by a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA). DESs are classified into five groups [6], with further DESs developments: natural deep eutectic solvents (NDESs) [7], formulation base on mixtures of dimethylthetin (DMT), oxalic acid dihydrate and water [8], mixtures of imidazole and monoethanolamine [9], etc. Its properties: non-toxicity, low cost production, easy to prepare, biodegradable, environmental adaptability, etc., has led them to be considered as green solvents. However, and this must also be considered, several studies argued against the environmental friendship and non-toxicity of these DESs [10–12], though this toxicity can be eliminated or reduced by tunning the composition of these DESs (both the hydrogen bond donor or acceptor and its concentration ratios). Of these compounds, the former have the greatest influence on cytotoxicity due to the various strength of hydrogen bonds present in these [13].

DESs are used in a wide field of interest, such as the pharmaceutical industry [14,15], membrane preparation for environmental separation technologies [16], next-generation lithium batteries [17], recovery of valuable and critical metals [18,19], biocatalysis [20], synthesis of nanomaterials [21], and many more [22,23]. Including in these utilizations, DESs also known a wide use in the capture of CO_2.

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It was reviewed the physical and chemical properties of DESs and their effects on the capture of this gas [24,25], whereas these chemicals had been use as carbon precursors facilitating CO_2 capture and improving energy storage [26]. By the use of machine learning procedures it was predicted the solubility of CO_2 into DESs, i.e. choline-based DESs among others [27–30], being DESs performance for enhancing the electrocatalytic CO_2 conversion also reviewed [31].

The present manuscript reviewed the most recent publications (2024 year) related to CO_2 capture using absorption, adsorption and mixed matrix membrane processes together with the use of deep eutectic solvents. The review utilized Scopus "www.scopus.com" and Web of Science https://webofscience.clarivate.cn/wos/woscc/basic-search databases (both accessed on October to December 2024). The search terms include: CO_2 capture and deep eutectic solvents, absorption, adsorption, and membranes.

2. DESs and Absorption

In absorption technology, CO_2 is removed from the syngas previously to the H_2 purification step. DESs are a type of chemicals suitable for CO_2 removal because they allow to have competitive alternatives to amine solutions [32,33].

Alanine or lysine-based ionic liquids tetraethylammonium alaninate ([N_{2222}][Ala]) and tetraethylammonium lysinate ([N_{2222}][Lys]) have been prepared and used to fabricate two DESs [34], in which the ionic liquid and ethylene glycol acted as hydrogen bond acceptor and donor, respectively. Lysine-based DES presented a higher CO_2 removal capacity than the analogous DES-based on alanine. Both DESs have also been used to investigate its performance on the direct CO_2 capture from open air. It is found that in this condition, the lysine-based DES remove a great concentration than its alanine counterpart, 1.06 mol/mol against 0.47 mol/mol, respectively. Those DESs are reversible toward the absorption and desorption of CO_2 up to several cycles.

Based in the concept of carbon capture, utilization and storage (CCUS), the next reference [35] worked with functionalized deep eutectic solvents to reached the CO_2 capture and integrating CO_2 desorption and biofixation in microalgal culture. Used DESs contained choline chloride, ethylene glycol, and monoethanolamine, showing effective CO_2 desorption from the solvent (90% efficiency). These DESs were also used in the promotion of microalgal cultives based on a *Chlorella sp.* strain.

1,3-bis(isopropyl)imidazolium 1,2,4-triazolide ([IiPim][Triz]) and ethylene glycol (EG) formed a DES which was utilized on CO_2 capture [36]. The as formed [IiPim][Triz]-EG deep eutectic solvent presented a CO_2 uptake capacity of near 1 mol/mol for experiments carried out at 25º C and 1 atm. In the CO_2 removal process, the gas molecule reacted with ethylene glycol but not with the C-2 site of the ionic liquid, being this behavior attributable to the steric hidrance in this C-2 site.

Super-nucleophilic deep eutectic solvents were formulated using carbanion-based ionic liquids as the key components for the construction of these DESs [37]. These formulations improved the otherwise slow CO_2 capture when only the ionic liquid, due to its high viscosity, is used. Synergism between the carbanion siting and hydrogen bonding is the responsible for the fast absorption of the new formulated DESs.

An investigation about the removal of CO_2 from natural gas, biogas and coal as fuels that originate the CO_2 flux was performed using a DES formed by choline chloride and urea (1:2) [38]. The results from this investigation revealed that the use of coal as fuel in the combustion stage presented the best sustainability indicators. Moreover, comparison of the results with those derived with the use of monoethanolamine as solvent, indicated that the utilization of the DES is preferred if costs and environmental terms were considered.

Various DESs formulations (1.3 to 1:10 molar ratios) based in [MEACl][EDA] and different water content (30-60 wt%) were used to investigate their performance on CO_2 capture [39]. Experimental results showed that 40 wt% of [MEACl][EDA] (1:5 molar ratio) presented the highest CO_2 uptake (22.09 wt%), improving the capacity (15.74%) of a 40 wt% MEA solution. Furthermore, recycling studies reveal about 88 % regeneration of the aqueous DES solution operating at 100 °C.

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This investigation utilized a mixture of DESs and sulfolane as medium to capture CO_2 [40]. The DESs were formulated on a choline chloride and MDEA at a 1:6 molar ratio, being the above mixture blended with sulfolane in weight percentages of 5, 10, and 15. Whereas the presence of sulfolane harmed CO_2 removal, results indicated that increasing the pressure and reducing the temperature (from 70 to 50º C) led to increase CO_2 removal. Comparison of these results with those obtained using the same DES but adding piperazine to the system [41], allowed to conclude that against the performance of sulfolane, the presence of piperazine is beneficial towards CO_2 removal.

A number of ternary superbase/betaine-based deep eutectic solvents (DESs) (Table 1) were synthesized to capture CO_2 [42]. All the DESs presented an acceptable affinity towards CO_2 (best in the case of the DES synthesized from betaine, 1,2-propanediol and 1,8-diazabicyclo(5.4.0)undec-7-ene). This affinity increased with the increase of the pressure from 0 to 1 MPa, and the decrease of the temperature from 50 to 30º C. CO_2 was uptook onto the DES by simultaneous chemical and physical processes. CO_2 was desorbed under vacuum at 90º C and operating during two hours.

Table 1. Composition of the ternary DESs used in the experimentation.

Betaine:1,2-propanediol:1,8-diazabiocyclo(5.4.0)undec-7-ene
Betaine:1,2-propanediol: 1,5-diazabicyclo(4.3.0.)non-5-ene
Betaine:1,2-propanediol:1,1,3,3-tetramethylguanidine
Betaine:diethylene glycol:1,8-diazabicyclo(5.4.0)undec-7-ene
Betaine:diethylene glycol:1,5-diazabicyclo(4.3.0)non-5-ene
Betaine:diethylene glycol:1,1,3,3-tetramethylguanidine

All the DESs in 1:6:1 molar ratio basis. Adapted from [42]. The chemical structures of the above DESs are shown in Figure S1 (Supplementary Information).

In the next reference [43], quasi-deep eutectic solvents (QDESs) based on proton donors and organic superbase were synthesized and used to adsorb CO_2. The results showed that 1,5-diazabicyclo[4.3.0]non-5-ene:ethylene glycol (1:1) presented the best removal uptake (0.214 g/g at 40º C under 20 kPa). This capture was attributed to the dual sites of ethylene glycol, which formed $[OOCOCH_2CH_2OCOO]^{2-}$ species with CO_2. Against the above, the substitution of ethylene glycol by imidazole produced a decrease in the capture of CO_2 (around 0.15 g/g).

Tetraethylenepentamine (TEPA) was transformed to its quaternay ammonium salt derivative (HBA) by reaction with hydrochloric acid [44]. The combination of this HBA with different donors produced a series of DESs, which were used on CO_2 capture. TEPA-MEA and TEPA-EDA DESs presented the best CO_2 capture efficiencies, which were attributed to the lower viscosities of these chemicals.

A deep eutectic solvent formulated as a mixture of 2(methylamino)ethanol ad choline chloride hydroxide in 1:1 molar ratio was used in CO_2 absorption [45]. In the 50-150 PSI range of pressures, the results indicated that 2M DES aqueous solutions presented better CO_2 removal rates than the values presented by the pure DES. CO_2 was captured via carbamate formation.

The presence of water in DESs systems was evaluated using four choline chloride:3-amino-1-propanol (MPA 15-85 wt%) and an aqueous choline chloride: 3-(methylamino)propylamine (MAPA 30 wt%) adsorbent [46]. Experimental results concluded that from an aqueous amine solution, the substitution of the amine by choline chloride has little effect on the CO_2 uptake (expressed as mol CO_2/mol amine), but has a negative effect on the overall uptake (expressed as mol CO_2/kg solvent). The substitution of water by choline chloride does not affect the CO_2 absorption by the DESs.

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Tunability of DESs is one of their properties used to reach specific uses, such as the removal of CO_2 from gas streams. In the present work and taking choline chloride as basis [47], different solvents are formed with four hydrogen bond donors, namely: urea (U), formamide (F), monoethanolamine (MEA) and 1-aminopropan-2-ol (Apr). Experimental results showed that no one of the above formulations remove CO_2 better than the single monoethanolamine system (Table 2).

Table 2. Carbon dioxide removal using various absorbents.

Absorbent	g CO_2/g absorbent
MEA	0.32
ChCl-MEA	0.22
ChCl-Apr	0.18
ChCl-U	nil
ChCl-F	nil

Pressure: 0.1 MPa. Temperature: 30º C. Time: 2 hours. Adapted from [47]. The structures of the above chemicals are shown in Figure S2 (Supplementary Information).

Another absorption systems of interest used liquid surfactants to improve CO_2 solubility on DESs. In the next reference [48], one of such surfactants Triton X-100 was added to aqueous solutions of ethaline (choline chloride and ethylene glycol) to investigate the performance of this ternary solvent mixture on the gas solubility. Since Triton X-100 adsorbed CO_2, the presence of this compound in the ternary system is beneficial with respect the removal of CO_2, and also with respect to the results derived from ethaline-water binary system.

The concepts of circular economy and green chemistry call for the use of processes and involving chemicals that reduce the need to use toxic or harmful products. Thus, deep eutectic solvents are being using to create new processes or to modify known ones. In the next work, choline chloride:urea (1:2) is used to capture CO_2 from post-combustion gas [49]. Utilizing this DES and comparing when an amine (MEA) was used, the environmental impact of the DES-process was reduced by a 13.97%, whereas 25.38% less energy was required if based on amine absorption. The above contributed to a reduction in global cost by 32.11% (equipment), 19.64% (service), and 21.13% lower cost of operation.

Tetrapropylammonium chloride and acetic acid with ethanolamine-based DESs was used in the removal of CO_2 [50]. The use of both Density Functional Theory quantum and Molecular Dynamics allowed to establish available voids by CO_2 molecules. The mechanism responsible for this capture were the interactions with ethanolamine and weak hydrogen bonding. Whereas physical absorption was responsible for the gas removal, the presence of O_2, N_2 and H_2O produced a negative effect on CO_2 absorption.

In the next reference [51], a number of trioctylphosphine oxide (TOPO)-based azole deep eutectic solvents (Table 3) are synthesized to investigate their performance on CO_2 capture. Results indicated that the carbon dioxide uptake capacity was influenced by the type of hydrogen bond donors, mole fraction of HBDs (increasing this fraction resulted in a decrease of CO_2 removal), temperature (decreasing gas loading when the temperature varied from 35º C to 50º C), and humidity (the presence of water decreased gas uptake). CO_2-captured in the DES was desorbed by bubbling nitrogen at 50º C.

Table 3. The various HBDs and HBAs used to capture CO_2.

HBA	HBD	[a]CO_2 uptake, mol/mol
TOPO	Im	0.062
	4-MIm	0.063
	Py	0.026
	3-AP	0.033
	123-Tz	0.051
	124-Tz	0.031

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Im: imidazole. 4-MIm: 4-methylimidazole. Py: pyrazole. 3-AP: 3-aminopyrazole. 123-Tz: 1,2,3-triazole. 124-Tz. 1,2,4-triazole. [a]Using HBA:HBD in 1:1 basis. Temperature: 35º C. Pressure: 0.1 MPa. Time. 2 hours. Adapted from [51]. The structures of the various DESs are shown in Figure S3 (Supplementary Information).

Amino-based functionalized deep eutectic solvents are extremely popular in the investigations on CO_2 capture. However, the increase in the viscosity of such systems after CO_2 uptake is a potential constrain in the use of these systems. To resolve this problem, the next investigation and using aromatic amines and ethylene glycol the proton transfer site was transferred from -NH_2 to -N═ preventing the formation of -NH-COO- and -NH_3^+ [52]. In the presence of ethylene glycol, the absorption capacity of 4-aminopyridine increased from 0.10 mol of CO_2/mol of DES to 0.60 mol of CO_2/mol of DES at 30º C and 101.3 kPa. This DES can be regenerated after 20 min at 80º C. In the gas removal process, the formation of carbamate and the protonation of pyridine nitrogen, played a key role. In any case, the viscosity of the system increased from 29.17 to 68.40 mPa·s after absorption.

One step-hydrolisis reaction is widely used to prepare DESs, and in the next investigation a series of γ−aminobutiric acid (Gaba), ε−aminocaproic acid (Eaca) and n-methyl-4-aminobutyric acid (Maba) based amino acid salts (AASs) were prepared through this type of reaction using lactams as hydrogen bond acceptor [53]. Ethylene glycol (EG), diethylene glycol (DEG) and triethylene glycol (TEG) were used as hydrogen bond donors. The absorption capacity of these DESs increased with the increase of the pressure from 0.1 to 1 bar. CO_2 loadings at 40º C varied from 0.77 mol/mol to 0.63 mol/mol: 0.77 mol/mol (K[Gaba]-EG), 0.76 mol/mol (K[Maba]-EG), 0.76 mol/mol (K[Maba]-TEG), 0.73 mol/mol (K[Eaca]-EG), 0.69 mol/mol (K[Maba]-DEG) and 0.63 mol/mol in the case of Na[Eaca]-EG. The absorption mechanism involved both the formation of 1:1 and 1:2 CO_2:AAS species. After heating at 80º C during two hours the CO_2-loaded DESs, the released CO_2 was removed from the system by multiple evacuations (1 min each) to prevent the solvent evaporation.

By the use of the one-step reaction, a series of DESs based on protic ionic liquid and amine was formed (Table 4) and the results of CO_2 removal were compared with that obtained with the use of industrial MDEA solution [54]. In all the cases, the removal of CO_2 was pressure-dependent, increasing with the increase of this variable from 0.1 to 1 bar, exceeding (0.69 mol/mol in the case of [DMAPAH][Ac]-Dmee) the uptake presented by the MDEA solution (about 0.28 mol/mol). Conditions for desorption were 100 min, 0.002 bar and 60º C. In the presence of nitrogen, these DESs presented CO_2/N_2 convenient selectivity values (Table 4).

Table 4. CO_2 uptakes and CO_2/N_2 selectivities values of several DESs.

DES	[a]CO_2 uptake, mol/mol	[a]CO_2/N_2 selectivity
[DMAPAH][Ac]-Im	0.50	29.3
[DMAPAH][Ac]-EIm	0.53	28.9
[DMAPAH][Ac]-1,2,3-Tri	0.48	34.6
[DMAPAH][Ac]-Dmee	0.69	38.0

DMAPAH: protonated 3-(dimethylamino-1-propylamine). Ac: acetate. Im: imidazole. EIm: 1-ethylimidazole. 1,2,3-Tri: 1,2,3-triazole. Dmee: dimethyaminoethoxyethanol. [a]Pressure: 1 bar. Temperature: 25º C. Adapted from [54]. The structures of these various DESs are shown in Figure S4 (Supplementary Information). .

A DES formed by choline chloride and monoethanolamine (ChCl:MEA, 1:5 molar mass ratio) was used in the capture of CO_2 from mimic biogas (40% vol CO_2 and 60% CH_4) [55]. The removal of CO_2 was dependent on temperature (increasing with the increase of this variable from 20 to 60º C), pressure (increasing with the increase of the pressure from 0.5 to 1.5 bar), and water content (increasing from zero to 75% vol. water). With respect to the CO_2/CH_4 selectivity, this value was also affected by the temperature (increasing with the increase of the temperature from 20 to 60º C), pressure (increasing with the change of pressure from 0 to 1.5 bar), though the water content in the DES phase had a minor influence on these CO_2/CH_4 selectivity values.

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It is know that the use of microchannel reactors favored chemical processes due to capacity to improve the limited gas-liquid mass transfer. These reactors in conjuction with the DES formed by mixing the ionic liquid 1-ethyl-3-methylimidazolium chloride (EMIMCl) and ethanolamine (MEA) in a molar ratio of 1:1 were used for CO_2 capture [56]. As in many systems, the presence of water in the DES decreased the viscosity of the phase (54.10 mPa·s in pure DES, 2.59 mPa·s with 60% vol water content), and increased the removal of CO (up to 50% vol water, decreasing at 60% vol). This gas was loaded onto the DES phase by a chemical process between CO_2 and MEA. Desorption was performed at 110º C.

Mixtures of 2-amino-2-methyl-1-propanol (AMP) and 2-(ethylamino)ethanol (EAE) amines were used on CO_2 capture [57]. Different mixtures were prepared on a molar ratio basis, namely: 3AMP:1EAE, 1AMP:1EAE, and 1AMP:3EAE. In these systems, the increase of the temperature (20-50º C) produced a decrease in the absorption of CO_2. Desorption experiments carried out at 90º C showed that these mixtures performed better than a 30% MEA aqueous solution in terms of desorption rate and energy consumption in this step.

A 7:3 mixture of 1,8-diazabicyclo [5.4.0] undec-7-ene imidazole and ethylene glycol was used in the capture of CO_2 from mimic flue gas stream [58]. With a CO_2 uptake of 1.89 mol/L, vacuum flashing desorption was used as energy-saving solvent regeneration. Simulation of the process in an industrial scale originated a regeneration energy consumption of 1.54 GJ/tCO_2. With respect to the costs associated to this capture, the methodology offered a viable alternative with respect to the use of other established solvents.

The commonly used one-step process was used to synthesize DESs formed by tetrabutylammonium bromide or tetrabutylphosphonium bromide, and two hydrogen bond donors as ethanolamine and N-methyldiethanolamine [59]. After, water was added to the respective mixture to formulate various 50 wt% (HBA+HBD) DESs compositions. Under the different formulations, the decrease of the temperature (30-50º C) and the increase of the pressure (100-900 kPa) produced and increase in the CO_2 uptake, thus maximum loadings were obtained working at 30º C and 900 kPa. Also, the variation in the HBAs:HBDs mass ratio from 1:8 to 1:10 resulted in an increase of the gas loading. It was concluded that the influence of the donor on the CO_2 capture is more influential than that of the acceptor, which is attributable to the reactions involved in the removal of the gas, with the amines reacting with CO_2 molecules. At 40º C and pressures exceeding 400 kPa, the formulation 50 wt% (1 HBA+10 ethanolamine) presented higher CO_2 uptakes than the values resulted from the use of a 30 wt% aqueous ethanolamine solution. No DESs regeneration data were included in the work.

In the next investigation [60], acetamide and diethylene glycol, 1,2-propanediol, or 1,3-propanediol were used to formulate various DESs compositions. It was experimentally demonstrated that in all DESs, CO_2 was physically removed, having the diethylene glycol-based compounds the highest CO_2 loading. As in the previous reference, the decrease of the temperature (30-50º C) and the increase of the pressure (200-800 kPa) produced the highest CO_2 loadings or solubilities in the DESs. CO_2 desorption was performed under vacuum at 90º C during two hours.

A series of ternary deep eutectic solvents containing choline chloride, glycerol and ethanolamine at different molar ratios were prepared [61]. Among all the formulations, ChCl-Gly-MEA (1:1:10) presented the highest loading (0.18 g CO_2/g at 25º C and 101.3 kPa). This loading increased with the decreased in water content and with the increase of the gas flow rate. Gas uptake was attributed to both physical and chemical processes, whereas the chemical reaction was favored by the use of greater amine concentrations in the DESs formulation.

Cyclodextrins (CDs) and monoethanolamine (MEA) were the basis for the formulation of different DESs designed for CO_2 capture [62]. From all of these compositions, MEA:CD with a 3:1 mass molar ratio presented the best capture results in the 30–110 °C range (i.e. 28.7% efficiency at one hour and 80° C). The temperature of 110º C was selected to carry out desorption from the CO_2-loaded DES.

In the next reference [63], tetrapropylammonium bromide (TPAB) and formic acid (Fa) were used to form the DESs with compositions TPAB-Fa (1:1) and TPAB-Fa (1:2). Similarly to other

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previous cases, the capture of CO_2 increases at low temperatures (i.e. 25º c) and high pressures (i.e. 35 bar). Of the above DESs, the formulation 1:2 presented the highest physical-based CO_2 capture efficiency at 25º C and 35 bar. Both formulations presented better regeneration characteristics than that presented by the widely used 30 wt% aqueous MEA solution.

Since water plays a key role in DESs properties, the next investigation studied the influence of the presence of this diluent on a series of amine-based DESs (amines: monoethanolamine, diethanolamine or methyldiethanolamine versus choline chloride) [64]. To these DESs (1:6 amine:choline chloride mass ratio), water (2.5-15 wt%) was added to investigate its influence on CO_2 capture. As it was somewhat expected, the presence of water decreased the viscosity of all the systems, being this property also influenced by the increase of the temperature up to 80º C. With respect to the CO_2 uptake (40º C and 100 kPa), no one of the DESs formulations presented better results than that derived from the use of 30 wt% aqueous MEA solution. Not desorption data included in the work.

In the next work, choline chloride-monoethanolamine-piperazine DESs were fabricated and used on CO_2 capture [65]. It was demonstrated that the presence of piperazine increased the removal of CO_2 with respect to the use of choline chloride-monoetahnolamine DES. Also, the presence of 50 wt% water in the DES formulation helps to yield better CO_2 removal efficiencies and to decrease the viscosity to a minimum value (11.19 mPa·s) after CO_2 capture. The removal of this gas was attributed to a chemical reaction between the CO_2 molecule and the DES via the zwitterion-formation mechanism. Whereas desorption was performed at 90º C, the energy consumption in this step was lower than that of the well known 30 wt% aqueous MEA solution.

The apparent drawbacks generated in the use of aqueous amine absorbents to remove CO_2 are the basis for the next investigation. Thus to resolve these problems, 1:6 choline chloride:monoethanolamine mixture was used to capture carbon dioxide [66]. Gas absorption increased with the increase of the amine concentration in the DES, this absorption was also favored in water-bearing DES, low temperatures and high pressures.

The next study used an amine-based deep eutectic solvent formed by ethanolamine hydrochloride (EAHC) and diethylenetriamine (DETA) in 1:9 molar ratio. Further, 30 % water was added to the DES [67], the presence of water reduced the viscosity and improved the fluidity and mass transfer properties of the aqueous DES solution. As it was being the rule, CO_2 capture was favored using low temperatures (30º C) and high pressures (1.6 MPa). Not desorption data included in the work.

Though it is difficult to compare the results obtained by the use of these DESs, basically due to the different experimental conditions used in each work, the next Table 5 summarized CO_2 loadings of some of these reviewed systems in order to gain knowledge about their performance on CO_2 capture.

Table 5. Summary of CO_2 uptakes using various DESs formulations.

DES	CO_2, uptake, mol/mol	Reference
$[N_{2222}][Lys]$:EG	1.2	[34]
$[N_{2222}][Ala]$.EG	0.81	[34]
ChCl:EG:MEA (1:2:1)	0.29	[35]
ChCl:MEA (1:8)	0.65	[35]
[IiPim][Triz]:EG (1.5)	0.99	[36]
DBN:EG	2.0	[43]
$[TEPA][Cl_3]$:EDA	4.2	[44]
ChCl:MAPA (4.92%:25.05%)	1.4	[46]
ChCl:MEA (1:5)	0.61	[55]
AMP:EAE (1:3)	[a]0.96	[57]
ChCl:MEA (1:6)	0.38	[66]
EAHC:DETA (1:9)	3.8	[67]

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[a]Under the same experimental conditions, 30% MEA has a CO_2 uptake of 0.61 mol/mol. See the corresponding reference to gain knowledge about the structure of the different chemicals shown in the Table. .

3. DESs and Adsorption

When this process on CO_2 removal is used, DESs are confined onto the solid material or they are part of its intimate structure. CO_2 removal can be attributed both to DESs and to the properties of the material.

Montmorillonite was functionalized with a DES composed by choline chloride:urea to investigate its performance on CO_2, O_2, and N_2 adsorption [68]. It was determined that CO_2 removal is due to a heterogeneous multilayer and chemophysical adsorption process, fitting the Hill isotherm and Ellovich kinetic model. Against the above, oxygen and nitrogen on a monolayer basis were captured, which fits the Langmuir isotherm and first-order kinetic equation. Whereas in single gases investigations the highest adsorption occurred at 25º C, in binary mixtures CO_2/O_2 and CO_2/N_2 the optimal adsorption were found to be at 35º C. The impregnation of the DES onto the montmorillonite increased the gas uptake from 106.3 mg/g (montmorillonite) to 134.1 (montmorillonite+ DES). Gas desorption was best performed at 100º C.

By using the same adsorbent (montmorillonite+choline chloride:urea) that in the previous reference, in the next work the adsorption of CO_2 in a continuous fixed-bed adsorption system was investigated [69]. Experimental data indicated that reduction in both breakthrough and exhaustion time as the flow rate increased from 20 to 40 mL/min were observed, which was attributable to a faster saturation due to the superior velocity gradient. The increase of the inlet CO_2 concentration (5-15%) resulted in a decrease in the breakthrough time as consequence of the acceleration in the adsorption rate, which resulted in a faster saturation.

The utilization of co-solvents in a DES can be of utility to improve the CO_2 removal by a given DES-adsorbent system. Thus, a DES ([MEACl][DEA] (1:5)) containing a co-solvent (water) was immobilized on silica to investigate its performance on CO_2 capture [70]. From all the formulations used, the sample containing 3 wt% of (5 wt% DES@silica) in 40 wt% aqueous DES) showed the highest capacity of 24.93 wt% at 22º C and viscosities of 7.32 and 21.82 mPa·s before and after CO_2 loading, respectively. These results compared well with those derived from the use of pure silica (22.77 wt%) and aqueous DES (22.09 wt%). It was worth to consider that the use of low temperatures enhanced CO_2 capture (Table 6), but also increased the viscosity after this capture (23 mPa·s at 20º C versus 12.3 mPa·s at 50º C). By heating the CO_2-containing slurry at 100º C, the gas were desorbed.

Table 6. CO_2 capture at various temperatures.

Temperature, ºC	CO_2 uptake, wt%
5	25.3
22	25
40	22
60	20
80	12.5

Pressure: 1 bar. Adapted from [70].

The next work [71] incorporated DES to high photothermal conversion materials. Choline chloride and 2-ethanolamine (1:7) formed the DES, which was mixed with reduced graphene oxide (RGO). Different formulations (RGO addition to the DES) were investigated, being that formed by 500 ppm on DES (DES-500) the one presenting the highest CO_2 uptake (about 0.4 g/g), which represented a notable improvement with respect to the capacity found in pure DES (about 0.28 g/g). As in other systems, the increase of the temperature (in this case until 75º C) improves CO_2 desorption.

The same DES that in the previous reference was used in this next investigation [72], but here impregnating it in porous silica gel. In the work, different weight percentages (10-60 %) of the DES

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(ChCl:MEA, 1:8 mass molar ratio) were utilized, and from these, the modified silica gel containing 50 wt% DES (Silica-CM50) presented the highest CO_2 removal capacity of 89.32 mg/g at 25º C. The increase of the temperature (25 to 60º C) produced a decrease in the gas removal efficiency, and against this behavior, the increase of the pressure (0.5 to 9 bar) produced an increase in the CO_2 uptake capacity for any of the temperatures mentioned above. It was mentioned in the published manuscript that nitrogen gas was used for desorption studies.

Taking as a basis the low partial pressures (0.1-0.2 bar) encountered in flue gas, the next reference investigates the removal of CO_2 under low pressure conditions by the functional metal-organic framework (NH_2-MIL101(Cr)) impregnated with choline chloride:urea (1:2) [73]. Using the pristine framework or the impregnated one (DES/NH_2-MIL101(Cr)), the CO_2 uptake increased with the increase of the pressure (0.15-1 bar) and the decrease of the temperature (15-30º C). Moreover, at the limiting conditions (1.0 bar and 15º C), the pristine MOF presented a lower capacity (2.62 mmol/g) than DES-impregnated MOF (4.38 mmol/g). In the case of investigating the CO_2/N_2 selectivity at 15º C and 0.1 bar, the results indicated that the pristine MOF presented a higher value (around 9) than the impregnated MOF (3.5). CO_2 desorption was investigated at 25º C under vacuum conditions.

Though with very limited data to compare and considering the different experimental conditions used, the utilization of DESs in adsorption processes produced also great discrepancies between the CO_2 uptakes onto the different systems, as results in references [68] (134 mg/g) and [72] (89.32 mg/g) attest to this.

4. DESs and Mixed Matrix Membranes

Being the less developed technology, DESs-based membranes can be proclaimed as a promising development for CO_2 capture and/or separation due to their permeation and selectivity properties.

It can be of interest the use of rare earth elements derivatives to form specialized membranes used to CO_2 capture. Thus, CeO_2-DES, acting as functionalized filler, to form a mixed matrix membrane together with polysulfone as polymer support were used [74]. The DES used in the investigation was formed by choline chloride:urea (1:2) mixture, whereas the membrane was prepared by the casting method. As one can expected, the composition of the membrane influenced the removal of CO_2 from the gas stream and also the selectivity CO_2/CH_4 and CO_2/N_2 (Table 7).

Table 7. CO_2 permeation and CO_2/CH_4 and CO_2/N_2 selectivities using CeO_2-DES-polysulfone mixed matrix membranes.

Membrane	CO_2 permeation, barrer	CO_2/CH_4	CO_2/N_2
Polysulfone	27.2	46	53
Polysulfone-CeO_2	30.2	47	50
Polysulfone-CeO_2-DES (2%)	33.1	49	58
Polysulfone-CeO_2-DES (5%)	41.4	60	79
Polysulfone-CeO_2-DES (10%)	45.6	61	78
Polysulfone-CeO_2-DES (15%)	48.2	62	86

Temperature: 25º C. Adapted from [74]. The structure of the used DES (choline chloride:urea is shown in Figure S2 (Supplementary Information).

5. Conclusions

The joint use of DESs and the various separation technologies have shown progress in carbon dioxide capture. However, further research is needed to develop cost-effective and sustainable materials for large-scale applications. Further research involving techno-economic analysis and taking into consideration, among others, the large inventory of absorbents that are necessary in an industrial plant is required.

Future research must examine the absorption selectivity of CO_2 (i.e. CO_2 capture from biogas is a promising approach to facilitate the utilization of biogas by improving its CH_4 purity) and

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adsorption-desorption performance in order to fulfil the requirements to be used on real industrial applications. Since most, if not all, of the data reported in the literature have been tested under laboratory (mild) experimentation, the real usefulness of these DESs, jointly with the separation technologies, are mostly unknown when they work under extreme harsh conditions as normally found in the industry, thus, it is crucial carry out these investigations UNDER REAL SCENARIOS. Moreover, the in situ conversion of CO_2 in DESs after absorption may be another of the future hot spots to be developed.

More research is needed in the following areas (with no particular order): molecular structures and the impact of water on CO_2 solubility in DESs; understanding of the properties of new DESs; modeling of the properties of DESs. Investigations about the viscosity, and its reduction especially after CO_2 loading, associated to these chemicals appeared to be one of the points of future interest and necessity.

One interesting promising use of DESs is their utilization in the synthesis of advanced porous carbon-based materials useful to carbon dioxide capture.

Development of DESs-bearing nanofluid systems that with their characteristics (i.e. increasing the system temperature) help in the desorption process and, thus, favored the continuous use of these adsorbents.

In any case, future developments must be in accordance with real industries necessities, at least with reference with present or near future time, and in accordance with the general policy of the reduction or mitigation of global warming.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org.

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