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Sustainable advancements in hazardous gases capture: Harnessing the potential of deep eutectic solvents

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ABSTRACT

Deep eutectic solvents (DESs) represent a promising solution for gas capture within existing industrial frameworks, requiring minimal retrofitting. This review explores the gas absorption and solubility capabilities of diverse DESs, emphasising their tunability through variations in hydrogen bond donors (HBDs), hydrogen bond acceptors (HBAs), and molar mixing ratios. The introduction of water marginally enhances gas absorption, while increased HBD ratios and elevated temperatures lead to decreased gas solubility. Conversely, pressure elevation improves gas solubility in DES. The alkyl chain length of the DES HBA correlates with gas solubility, attributed to an increase in DES free volume. Other factors like ionicity and alkalinity of DESs were reported to enhance gas capture. This review also discusses the selectivity of DESs in capturing distinct gases, unveiling a robust hydrogen-bond supramolecular network as the key determinant for high gas selectivity. Examples include effective NH_3 capture within NH_3/CO_2 mixtures and targeted SO_2 capture in CO_2/SO_2 mixtures. The absorption mechanism of gaseous pollutants using DES is both physical and chemical, with physical absorption playing a dominant role. During the regeneration of DESs, the stability and volatility of DESs are significant factors to consider. These insights underscore the potential of DESs in tailoring selective gas capture strategies, providing avenues for future research and practical applications in environmental and industrial contexts.

1. Introduction

Controlling the discharge of some gases into the atmosphere is necessary owing to their detrimental effects on human health and the environment when released through anthropogenic activities and other sources [1]. In this case, gases like CO_2, NH_3, NOx, SOx, and H_2S are involved. Reducing these emissions is among the global problems because of their role in climate change and global warming [2]. There

Abbreviations: ILs, Ionic liquids; DESs, Deep eutectic solvents; NADES, Natural deep eutectic solvents; HBAs, Hydrogen bond acceptors,; HBDs, Hydrogen bond acceptors; ChCl, Choline chloride; U, Urea; EG, Ethylene glycol; BTMAC, Benzyltrimethylammonium chloride; TBAC, Tetrabutylammonium chloride; TEAC, Tetraethylammonium chloride; TEMA, Triethylmethylammonium chloride; TBAB, Tetrabutylammonium bromide; BTEAC, Benzyltriethylammonium chloride; BHDE, N-Benzyl-2-hydroxy-N,N-dimethylethanaminium chloride; GUA, Guanidinium hydrochloride; MEA, Monoethanolamine; AA, Acetic acid; LA, Lactic acid; OA, Octanoic acid; LV, Levulinic acid; MA, Malic acid; DA, Decanoic acid; Gly, Glycerol; MTPPB, Methyltriphenyl phosphonium bromide; 1,2-PD, 1,2 propanediol; ACC, Acetyl choline chloride; TEAB, Tetraethylammonium bromide; DEA, Diethanolamine; MDEA, Methyldiethanolamine; TEA, Triethanolamine; TMAC, Tetramethylammonium chloride; TPAC, Tetrapropylammonium chloride; GC, Guaiacol; Imi, Imidazole; DEH, Diethylamine hydrochloride; DBU, 1,8-diazabicyclo[5.4.0]undec-7-ene; TBD, 1,5,7-triazabicyclo[4.4.0]-dec-5-ene; DBN, 1,5-diazabicyclo[4.3.0]-non-5-ene; EU, 2-Imidazolidone; [BMIM][$MeSO_3$], 1-Butyl-3-methyl imidazolium methanesulfonate; MTOAB, Methyltrioctylammonium bromide; MTOAC, Methyltrioctylammonium chloride; TOAB, Tetraoctylammonium bromide; TOAC, Tetraoctylammonium chloride; BA, Benzyl alcohol; DMU, Dimethylolurea; DMLU, 1,3-Dimethylurea; EMMIC, 1-Ethyl-3-methylimidazolium chloride,; SNT, Succinonitrile; TEG, Triethylene glycol; FMP, N-Formylmorpholine; PPZB, 1-Hydroxyethyl-1,4-dimethyl-piperazinium bromide; TU, Thiourea; MLA, Malonic acid; CA, Citric acid; BMIMC, 1-Butyl-3-methyl-imidazolium chloride; AcM, Acetamide; PA, Phenylacetic acid; Mimi, 4-Methylimidazole; 2-NH_2Py, 2-Aminopyridine; PrA, Propionic acid; FA, Formic acid; TEAHC, Triethylamine hydrochloride; $PM_{2.5}$, Particulate matter 2.5; EHCl, ethylamine hydrogen chloride; 1,3-DMTU, 1,3 Dimethylthiourea; TBPC, Tetrabutylphosphonium chloride; TBPB, Tetrabutylphosphonium bromide; Res, Resorcinol; [APH]NO_3, Amino-2-propanol nitrate; Phenol, PhOH; MOFs, Metal-organic frameworks; COFs, Covalent organic frameworks; $[Bmim]_2$ $[CuCl_4]$, 1-Butyl-3methylimidazolium tetrachlorocuprate II; $[Emim]_2[CoNCS_4]$, 1-Ethyl-3-methylimidazolium tetraisothiocyanatocobaltate II; [EtOHim][SCN], 1–2−Hydroxyethyl-3-methylimidazolium thiocyanate; [EtA][SCN], Ethanolamine thiocyanate; [Py][NTf_2], Pyridinium bistrifluoromethylsulfonylimide; [2-mPy][NTf_2], 2-Methylpyridinium bistrifluoromethylsulfonylimide; EaCl, Ethylamine hydrochloride; EmimCl, 1-Ethyl-3-methylimidazolium chloride; TeEG, Tetraethylene glycol; Pyr, Pyrrolidine; Oxazolidinone, Oxa.

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are several drawbacks to the existing methods for capturing, adsorbing, and separating these gases, which include the following: (i) they require a significant amount of energy (including high regeneration energy); (ii) they release potentially harmful byproducts; and (iii) they utilise volatile organic solvents [3].

Advancement in sustainable and green chemistry has become a critical environmental benefit in recent years [3]. In this context, the eco-design of chemical processes grounded in the principles of green chemistry is currently of utmost importance [4,5]. When it comes to chemical processes, the gas sweetening process is the one that is most frequently utilised in chemical processing plants or oil refineries for extracting gases from natural gas streams [6,7]. A good alternative to the gas capture process is believed to be methanol, dimethyl carbonate, propylene carbonate, selexol, and ionic liquids (ILs), which all physically absorb gases [3]. By definition, salts which are liquid at temperatures below 373.2 K are most commonly described as “ILs” in the literature [8]. ILs are a kind of salt composed of cations and anions. They have several beneficial properties, such as strong chemical and thermal stability, low flammability, and low volatility [8,9]. However, the use of ILs and other previously mentioned solvents is limited by their difficult preparation method, toxicity, high cost, high viscosity, and poor biodegradability [10,11]. For the gas capture process, it would seem necessary to look for and discover more affordable, effective, and environmentally friendly sorbents. It is interesting to note that deep eutectic solvents (DESs) have been found as alternatives recently. Some of the gases that have been successfully captured using DESs are depicted in Fig. 1 (**A and B**).

A class of "designer" extraction solvents known as DESs are easily prepared by combining and heating two or more constituents that function as hydrogen bond acceptors (HBAs) and hydrogen donors (HBDs) at a particular molar ratio [14,15]. For more details about the properties and available methods for preparing DESs, readers should consult a recent review article published by Al-Bodour et al. [16]. The melting temperature of DESs is significantly lowered compared to their components as a result of the hydrogen bond formation. This results in the majority of DESs being liquid at room temperature. The high dissolving power, ease of preparation, high thermal, chemical, and electrochemical stability, low vapour pressure, and inexpensive precursors have made DESs increasingly useful in a variety of scientific and technological endeavours [17]. They have been utilised for the separation,

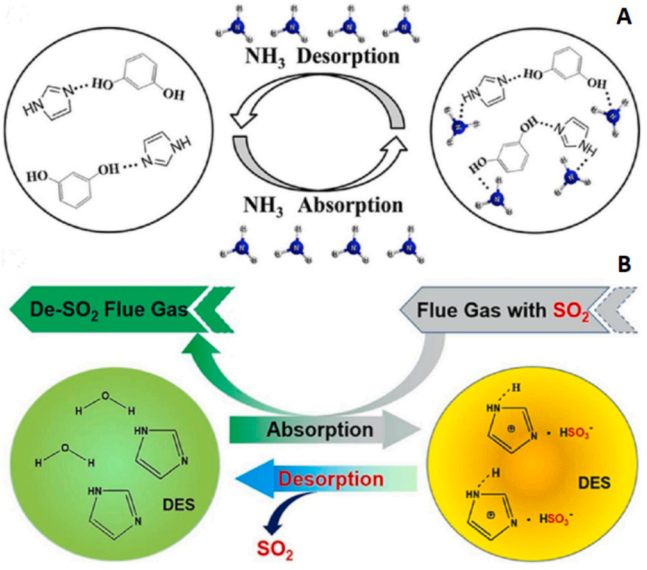

Fig. 1. (A) DESs utilised for the capture of NH_3. Reproduced from [12], Copyright 2021, with permission from Elsevier; (B) DESs used for absorption of SO_2. Reproduced from [13], Copyright 2020 with permission from the American Chemical Society.

capture, and adsorption of the previously mentioned hazardous gases from gas mixtures of various sources due to their uniqueness and environmental friendliness [18]. In addition, while the synthesis of DESs is far simpler, less expensive, safer, and greener than that of ILs, the chemical and physical properties of these solvents are fairly similar to those of ILs [19]. Furthermore, temperature, molar ratio of DES components, and HBA or HBD molecules can all be modified to tune the properties of DESs [20]. The first set of DESs to be proposed relied on donors and acceptors that were soluble in water [21]. These DESs fit the definition of hydrophilic solvents. The most common HBA is choline chloride (ChCl) which is highly soluble in water [22]. The HBDs are water-soluble substances like urea (U), ethylene glycol (EG) and carboxylic acids among others [23].

Gas capture using DESs has been extensively reviewed, with a predominant focus on CO_2 capture in recent literature. However, a comprehensive examination of gas selectivity, absorption mechanisms, DES regeneration, and comparisons of capture capacities with other materials has been notably absent [24–28]. Addressing this gap, this paper provides an exhaustive overview encompassing CO_2, SO_2, NH_3, H_2S, NO_2, and NO capture by DESs. The perspective outlined here includes various critical sections: (a) capture of various gases by DESs and influencing factors: This section explores the capture of CO_2, SO_2, NH_3, $H2_S$, NO_2, and NO by DESs, shedding light on both structural and external factors influencing gas capture; (b) selectivity of capturing gas mixtures by DESs: An in-depth discussion ensues regarding the selectivity of DESs in capturing gas mixtures, showcasing their ability to discern and capture specific gases effectively; (c) mechanisms underlying gas capture by DESs: This segment provides insight into the fundamental mechanisms governing gas capture by DESs, offering a nuanced understanding of the processes involved; (d) comparison of gas capture by DESs with other materials: A comparative analysis is presented, evaluating the gas capture capacity of DESs in contrast to other materials. This broader context aids in understanding the relative effectiveness of DESs; (e) regeneration of DESs: The regeneration process for DESs is explored, examining the existing method for recovering these solvents for prolonged use; (f) future directions: A forward-looking exploration highlights potential research directions and advancements in the gas capture field using DESs and (g) concluding remarks: it summarizes the key findings and insights, this section provides concluding remarks on the present state and prospects of gas capture with DESs. By consolidating these sections, this paper seeks to address existing gaps and offer a holistic understanding of gas capture using DESs, providing a foundation for informed future research and applications in this evolving field. The chemical structures of HBAs and HBDs of DESs commonly utilised for gas capture are presented in Fig. 2 **(A and B)**.

2. Capturing of CO_2

The management of CO_2 emissions is essential to the sustainability of the environment. Chemical adsorption, or amine-based processes, is one of the most widely explored technologies for CO_2 capture. However, this technology has been limited by issues such as solvent deterioration, cost, and high corrosion rate [29]. However, scientists must act quickly to develop practical carbon capture technologies to address the present anthropogenic sources of atmospheric CO_2 levels [30]. Unless otherwise specified, the SI unit used to compare CO_2 capture is typically $mol.kg^{-1}$. Because of their tunability, ILs have been seen as a promising solvent for CO_2 capture [31]. However, some problems have been identified through research to date that have impeded the commercial use of ILs for capturing CO_2 gas as previously highlighted [32]. So, in this section, the utilisation of DESs for CO2 is comprehensively elucidated. Owing to their outstanding characteristics previously mentioned, DESs have been investigated recently for CO_2 capture, as illustrated in Fig. 3.

Since DESs are also described as the analogue of ILs, many studies conducted in the last few years have concentrated on employing DESs to

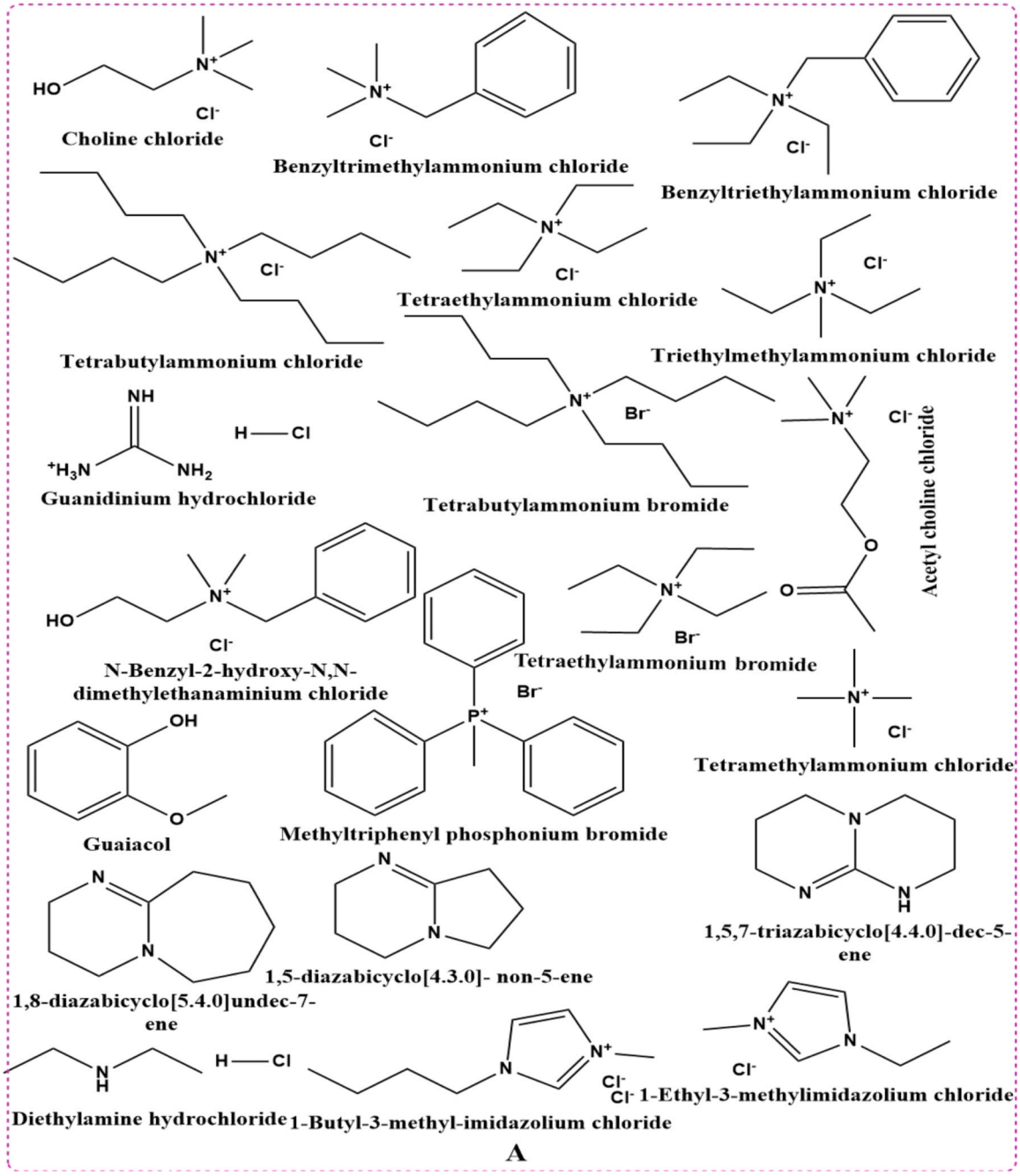

Fig. 2. The structures of Common DESs (A) HBAs; (B) HBDs used for capturing gases.

capture CO_2 through various HBA/HBD combinations. It is interesting to note that most studies involving the use of DES for CO_2 capture employed ChCl/U (1:2) combination. The pioneering work involving the measurements of CO_2 solubility in ChCl/U DES at a 1:2 molar ratio was performed at pressures as high as 13 MPa and temperatures of 313.15, 323.15, and 333.15 K by Liu et al. in 2008 [33]. The adsorption of CO_2 in DESs was discussed by investigating the effects of several parameters, including temperature, pressure, the nature of HBA or HBD, the HBA: HBD molar ratio, and the water composition of DESs. These factors were found to play a significant role in CO_2 absorption as demonstrated in the subsequent discussion.

Sarmad et al. investigated CO_2 capture using different DESs in which acetic acid (AA) was used as HBD [34]. At a 1:2 molar ratio of DES composition, 298.15 K, and pressure of about 2 mPa, the authors discovered that the effect of HBA in the DESs during the capture of CO_2 followed the trend benzyltrimethylammonium chloride (BTMAC) > tetrabutylammonium chloride (TBAC) > tetraethylammonium chloride (TEAC) ≈ triethylmethylammonim chloride (TEMA) > tetrabutylammonium bromide (TBAB) > benzyltriethylammonium chloride (BTEAC) > N-Benzyl-2-hydroxy-N,N-dimethylethanaminium chloride (BHDE). Fig. 4**A** provides a clear illustration of this trend. The same authors also looked at how HBD affect CO_2 capture [34]. For instance, DESs based on TEMA were synthesised by combining TEMA (HBA) in a 1:2 molar ratio with five distinct HBDs such as lactic acid (LA), EG, glycerol (Gly), and levulinic acid (LV). At 298.15 K and a pressure of about 1 mPa, the TEMA-based systems demonstrated the sequence TEMA/AA > TEMA/EG > TEMA/LV > TEMA/LA > TEMA/Gly as depicted in Fig. 4**B**. The strong interactions between CO_2 and the

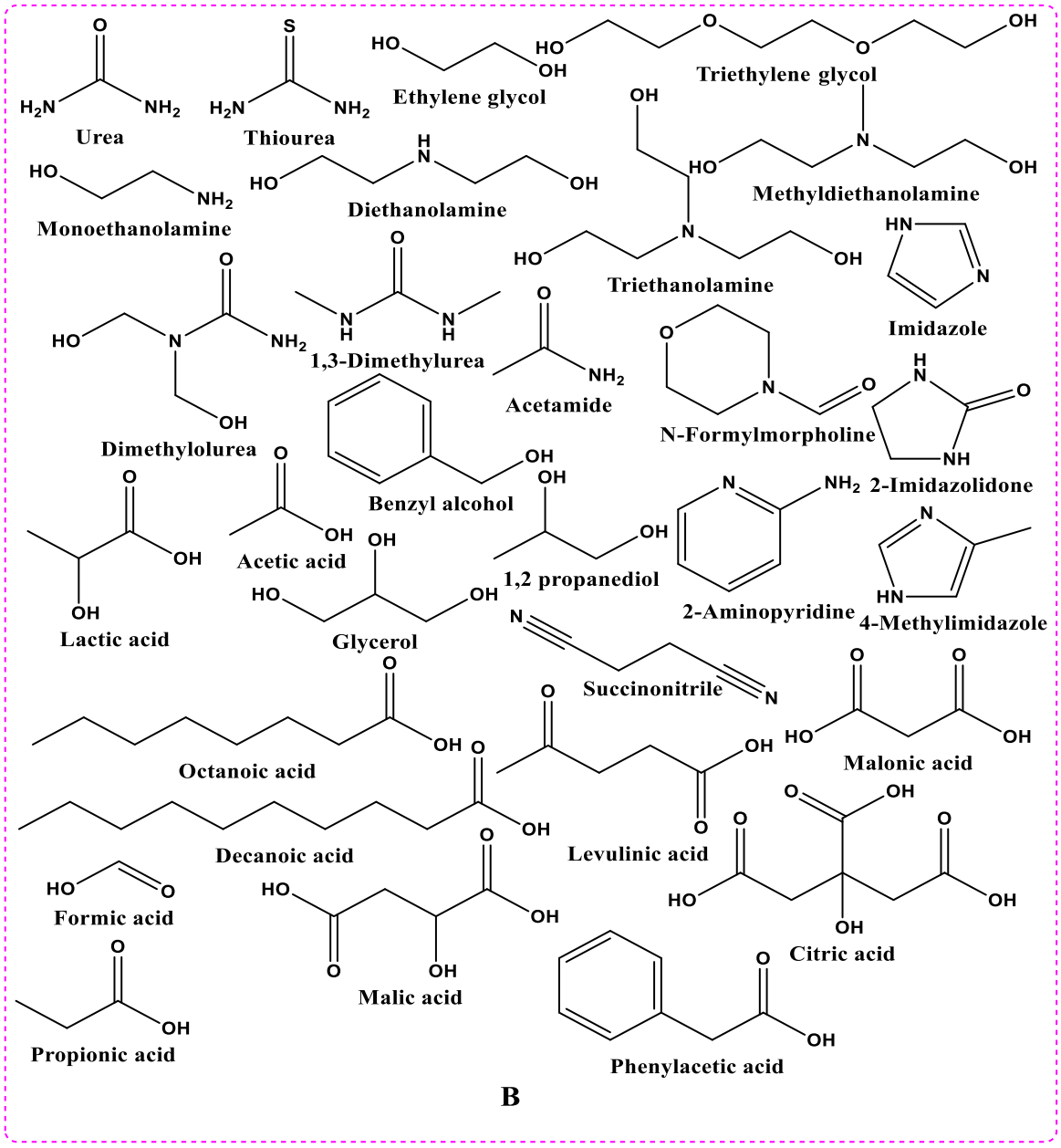

Fig. 2. (*continued*).

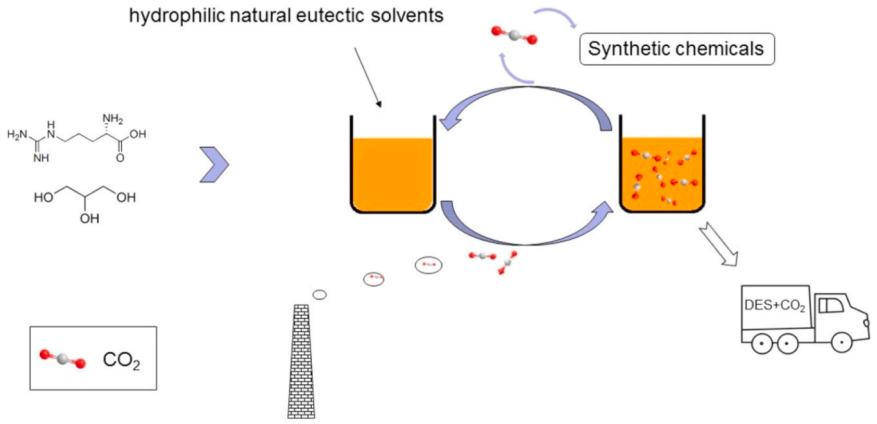

Fig. 3. DES constituted by hydrophilic and natural bases used for sustainable CO_2 absorption. Reproduced from [11], Copyright 2018 with permission from Elsevier.

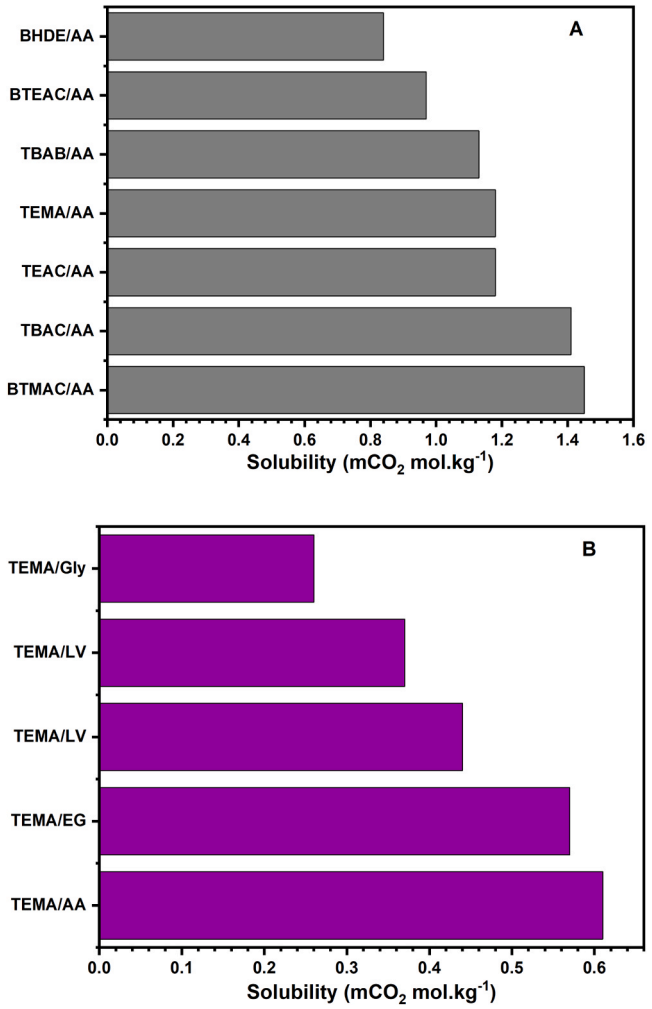

Fig. 4. (A) The effect of various HBAs of DES on the solubility of CO_2; (B) The effect of various HBDs on the solubility of CO_2 involving TEMA-based DESs. The data used for plotting these graphs were extracted from [34].

functional groups in the HBD were responsible for the differences in the results obtained. Moreover, LA has stronger intermolecular hydrogen bonds than AA or LV due to the closeness of the OH^- to the −COOH group. This explains why it is challenging to break the hydrogen bonds to make adequate contact with CO_2. Furthermore, compared to other DES systems, AA has higher CO_2 solubility because its molecules can interact with CO_2 more readily owing to the presence of weak intermolecular hydrogen interactions. It was noticed that for DESs based on AA, the solubility of CO_2 increased with the alkyl chain length of the HBA. For instance, the solubility of CO_2 rose from 1.177 to 1.411 mol. kg^{-1} when the alkyl chain length was increased from ethyl to butyl. Additionally, Sarmad et al. found that as temperature drops and pressure rises, CO_2 solubility in DES increases [34]. In other words, as temperature rises, CO_2 solubility in DES decreases. The kinetic energy of the gas molecules concept can be used to explain this phenomenon; as the temperature rises, the intermolecular bonds between the gas molecules formed within the solute break, increasing the tendency for the gas to escape from the solution. Thus, at high temperatures, the solubility of CO_2 in the DES decreases.

Furthermore, Zubeir et al. observed that as the alkyl chain length increases, CO_2 becomes more soluble in DES [35]. For example, it was noticed that CO_2 becomes more soluble in methyltrioctyl than in tetraoctylammonium. That is to say, more carbon atoms in the HBD alkyl chain mean that CO_2 is more soluble. The higher CO_2 solubility is caused by an increase in free volume with alkyl chain length, which explains this behaviour [36]. Deng et al. also investigated the impact of HBA of DES on CO_2 solubility in five LV-based DESs. All the DESs employed were prepared at a molar ratio of 1:3 (i.e., HBA to HBD molar ratio) [37]. The greatest CO_2 absorption of nearly 0.3 was displayed by the acetylcholine chloride (ACC)/LV and TBAC/LV DESs at a constant temperature of 303.15 K and pressure of approximately 0.55 mPa. Conversely, tetraethylammonium bromide (TEAB)/LV demonstrated the smallest solubility value of 0.24. According to Deng et al., the type of salt used to prepare the DES plays a significant role in absorbing CO_2. Several ammonium salts (ACC, TEAB, TEAC, TBAB, and TBAC) were used in this case to synthesise DESs that were utilised to capture CO_2. DESs with greater cations demonstrated increased CO_2 solubility. As a result, the cations of the salts dominated the capacity to absorb CO_2. The authors also used five LV-based DESs to demonstrate how pressure affects CO_2 solubility. It was observed that the solubility of CO_2 in DESs is inversely proportional to temperature and directly proportional to the equilibrium pressure of the gas phase. The impact of pressure and temperature on CO_2 solubility in LA:TBAB DES is illustrated in Fig. 5.

Li et al. also prepared a variety of DESs for CO_2 absorption by employing several ammonium-based salts as HBAs and MEA, diethanolamine (DEA), methyldiethanolamine (MDEA), and triethanolamine (TEA) as HBDs [38]. For the solubility of CO_2, the authors observed the order ChCl ≈ tetramethylammonium chloride (TMAC) > TEAC > TEAB > TBAC > TBAB, whereas for HBDs, they noticed the sequence MEA > DEA > MDEA > TEA. However, because there is no hydrogen on the nitrogen atoms, MDEA and TEA displayed low CO_2 absorption. The ability of ChCl and TMAC salts to absorb CO_2 was found to be similar because of their similar chemical structures. In addition, at 303.15 K, the effect of ACC-based DESs HBD was investigated. LV, guaiacol (GC), and imidazole (Imi) were used in the DES synthesis. The trend for CO_2 solubility obtained by the authors was ACC/GC (0.18) < ACC/Imi (0.29) < ACC/LV (0.3). Furthermore, it was demonstrated in the research by Li et al. that the capacity of DES to absorb CO_2 can be altered by the addition of inorganic salts [38]. The CO_2 absorption capacity of DESs containing $NiCl_2$, $FeCl_3$, $CoCl_2$, or $CuCl_2$ was observed to be almost the same as that of pure DESs. However, the addition of $ZnCl_2$, LiCl, or NH_4Cl to DESs increases CO_2 absorption. Because Ni^{2+}, Fe^{3+}, Co^{2+}, and Cu^{2+} have unpaired electrons in their electron shells, bonding with carbonyl oxygen is easier.

According to Liu et al., it was also observed that the solubility of CO_2 increased as the molar ratio of ChCl/GC, diethylamine hydrochloride (DEH)/GC, and ACC/GC increased (i.e., 1:3–1:5) at constant pressure and temperature [39]. This demonstrates the significant contribution of

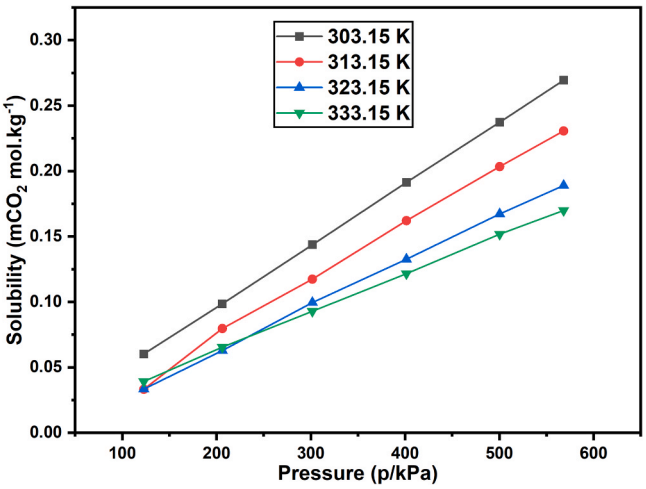

Fig. 5. Solubility of CO_2 in TBAB/LA DES (1:3) at different temperatures and pressures.
The data used for constructing this graph were extracted from [37].

the GC to the CO_2 solubility in the DESs investigated. For instance, when the molar ratio was increased from 1:3–1:5 at 298.15 K, it was discovered that the solubility of ChCl/GC increased from 0.171 to 0.188. When the mole ratio of ACC/GC was increased from 1:3–1:5 (resulting in an increase in CO_2 solubility to 0.196 from 0.187), comparable outcomes were observed. Adeyemi et al. also studied how the solubility of CO_2 can be affected by the molar ratio by utilising various amine-based DESs [40]. It was found that the solubility of CO_2 increased along with the molar ratio of ChCl/MEA and ChCl/DEA DESs, which increased from 1:6–1:10. The obtained results demonstrated that adding MEA or DEA (HBDs) can significantly increase the physical and chemical absorption of CO_2.

The majority of DESs readily absorb moisture due to their high hygroscopic nature [41]. Ren et al. researched the hydrophilic nature of DESs for CO_2 capture based on the hygroscopic nature of many DESs [11]. The authors used DESs with different water contents for CO_2 capture in their study. This was done in order to investigate how the solubility of CO_2 is affected by the amount of water present in the DES. So, they noticed that a small amount of water could boost the CO_2 efficiency in DESs. Also, Ma et al. examined the role of water on CO_2 solubility using DESs based on Gly [42]. The authors found that adding a small amount of water along with enhanced CO_2 solubility, significantly altered the viscosity of some of the DESs they studied. For example, the authors were able to reduce the viscosity of BTMAC/Gly (1:2) DES from 716 to 20 mPa.s by adding a small amount of water. In addition, BTMAC/Gly (1:2) exhibited a 25% rise in CO_2 absorption upon the addition of 0.11 mol fraction of water. However, as the water content was increased further, the solubility of CO_2 began to decrease because it is not very soluble in water.

Moreover, superbases—compounds with a high protonic affinity—have proven to be useful in capturing CO_2. Recently, several new superbase-based DESs have been studied. The capacity of 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) and 1,5,7-triazabicyclo[4.4.0]-dec-5-ene (TBD) based DESs for CO_2 absorption was investigated by García-Argüelles et al. [43]. The TBD/EG (1:4) DESs showed a relatively high absorption capacity of up to 12.9 mol kg^{-1} at 298.15 K and 100 kPa, which the authors found to be superior to the DBU-based DESs in terms of CO_2 absorption capacities. To capture CO_2, Jiang et al. also used four 1,5-diazabicyclo[4.3.0]-non-5-ene(DBN) based DESs [44]. DBN/DMU (2:1), DBN/DMLU (2:1), DBN/EU (2:1), and DBN/EU (3:1) made up the DESs employed. It was found that DBN/2-imidazolidone (EU) at 2:1 and 3:1 molar ratio had a greater capacity for absorbing CO_2 than DBN/DMLU (2:1) and DBN/DMU (2:1). Strong multiple-site interaction between the nitrogen atom of EU and CO_2 was responsible for the excellent results displayed by DBN/EU (2:1 and 3:1). Moreover, it was discovered that a reduction in the DBN ratio improved the CO_2 molarity absorption uptake of DBN/EU, providing compelling evidence that the molar ratio matters. DBN is a possible superbase for DESs synthesis because of the low steric hindrance surrounding the imine structure [45].

A groundbreaking study by Fu et al. has proposed a novel CO_2 absorption mechanism in addition to the role of ionicity and viscosity of DES in CO_2 [46]. The research distinguishes between HBD interactions with and without proton transfer, leading to 2:1 absorption mechanism with amines and 1:1 absorption mechanism with anions, respectively. The research system was further refined to enhance CO_2 absorption by increasing the HBA to HBD ratio, improving ionicity, and reducing DES viscosity. As a result, DBU:Pyrrolidine (Pyr) and DBU: Oxazolidinone (Oxa) DESs were identified as exemplary CO_2 trapping agents, offering superior thermal stability, low viscosity, high absorption capacity and lower optimal absorption temperature. One of the recent studies has revealed that strong relationships do exist between CO_2 capture efficiency and DESs basicity. For instance, A research finding by Qin et al. in 2024 has unveiled a robust linear correlation between the Hammett basicity/acidiity values of DESs and the initial concentrations of CO_2 [47]. This correlation not only highlights the significant influence of DES basicity/acidity, measured by the Hammett scale, on initial CO_2 levels but also suggests that as the basicity/acidity of DES increases, so does its efficiency in CO_2 capture. This discovery provides valuable insights into the deep relationship between DES characteristics and their interactions with CO_2, potentially influencing our understanding and manipulation of processes involving CO_2 capture.

The Kamlet–Taft solvatochromic parameters DESs have also been reported to primarily exhibit correlations with the structures and properties of HBAs and HBDs [48]. Notably, the elongation of the alkyl chain within these substances tends to diminish their capacity to either donate or accept hydrogen. Consequently, both HBA and HBD values exhibit a decrease corresponding to the augmentation of the alkyl chain length. In this context, the π^* (polarizability of the solvents) parameter predominantly reflects the non-specific interaction between DESs and analyte.

The solubility data of CO_2 in several DESs at different temperatures and pressures are presented in Table 1.

3. Acidic gases capturing

Industrial toxic gases include SO_2, H_2S, NH_3, NO_2, and NO. Burning fossil fuels, volcanic eruptions, and industrial waste gas produce acidic SO_2 [1]. Significant amounts of SO_2 in the atmosphere have been linked to human cancer, haze, acid rain, and air pollution [52]. Another acidic gas that can be produced by industrial refineries, natural gas production, decomposition, and volcanic activity is H_2S. H_2S is extremely poisonous and corrosive [53]. NO_x such as NO and NO_2 are produced majorly via the combustion of coal and could lead to acid rain, acid mist, damage to human health and destruction of the ozone layer [52]. NH_3 is usually produced from waste gas during the preparation of urea, which promotes the formation of particulate matter and causes air pollution as well as rhinitis or pharyngitis [54].

Because SO_2, NH_3, H_2S, NO_2, and NO are toxic, it is important to capture them using methods that are sustainable, high-performing, and low-cost. Toxic gas capture would protect the ozone layer, reduce air pollution, improve air quality, and ultimately increase gas use. One of the acceptable decolourisers, bactericides, and preservatives is recovered SO_2 [55]. Fertilizer, nylon, ammonium salt, and refrigeration fluid can all be made with recycled NH_3 [56]. Therefore, the development of sustainable routes with superior selectivity and effectiveness is imperative in order to capture these harmful gases.

3.1. *Capturing of SO_2*

The combustion of fossil fuels and other human manufacturing processes are the main sources of SO_2, which is currently one of the most common air pollutants [57]. Injurious to both humans and the ecosystem, SO_2 emissions cause could cause severe environmental problems (such as acid rain, fog, and haze) [58–60]. With the growth of modern industry, the efficient capture of SO_2 has attracted increased attention. Interestingly, diverse technologies have been investigated for SO_2 absorption to lessen the harm caused by the gas. The ammonia-water absorption, gypsum, and limestone are the three most commonly used traditional flue gas desulphurisation strategies in the modern industry [61–63]. However, the conventional desulphurisation process has many unavoidable drawbacks, such as the absorbent's high volatility and the creation of secondary pollutants. Furthermore, recycling the aforementioned absorbents is not possible [64,65]. For efficient, selective, relevant, and environmentally benign SO_2 removal, it is crucial to look for novel absorbents. Therefore, recently researchers have found DESs to be very effective, green, and selective for capturing SO_2.

A lot of studies have demonstrated the capture of SO_2 gas successfully using DESs. For example, Yang and colleagues used ChCl/Gly DES at different molar ratios to study the solubility of SO_2 [66]. The optimal solubility of SO_2 in ChCl/Gly DES was achieved at 293.15 K and 0.1 mPa

Table 1
Solubility data of CO_2 in several DESs at different temperatures and pressures.

DES	HBA:HBD Ratio	T (K)	P (MPa)	$^{m}CO_2$ (mol. kg^{-1})	References
BTEAC/AA	1:02	298.15	0.551	0.265	[34]
BHDE/AA	1:02	298.15	0.533	0.199	
BHDE/LA	1:02	298.15	0.866	0.122	
BTMAC/AA	1:02	298.15	0.530	0.271	
ChCl/MEA	1:07	298.15	0.651	2.700	
GUAHC/MEA	1:02	298.15	0.563	0.827	
TBAB/MEA	1:07	298.15	0.654	1.036	
TBAB/AA	1:02	298.15	0.715	0.380	
MTPPB/LV	1:03	298.15	0.994	0.161	
MTPPB/Gly	1:03	298.15	0.875	0.111	
MTPPB/EG	1:03	298.15	0.710	0.137	
MTPPB/AA	1:04	298.15	0.652	0.390	
MTPPB/1,2-PD	1:04	298.15	0.861	0.228	
TEAC/OA	1:03	298.15	0.624	0.342	
TEMA/AA	1:02	298.15	0.413	0.192	
TEMA/EG	1:02	298.15	0.314	0.199	
TEMA/Gly	1:02	298.15	0.833	0.126	
TEMA/LA	1:02	298.15	0.418	0.109	
TEMA/LV	1:02	298.15	0.409	0.163	
TMAC/AA	1:04	298.15	0.519	0.296	
TPAC/AA	1:06	298.15	0.554	0.481	
TPAC/MEA	1:04	298.15	0.481	0.338	
	1:07	298.15	0.645	2.051	
MTOAB/DA	1:02	298.15	0.490	0.285	[35]
MTOAC/DA	1:02	298.15	0.490	0.297	
TBAC/DA	1:02	298.15	0.490	0.337	
TOAB/DA	1:02	298.15	0.490	0.337	
TOAC/DA	1:1.5	298.15	0.490	0.305	
	1:02	298.15	0.490	0.307	
ACC/LV	1:03	303.15	0.543	0.301	[37]
TBAB/LV	1:03	303.15	0.568	0.269	
TBAC/LV	1:03	303.15	0.559	0.303	
TEAB/LV	1:03	303.15	0.564	0.240	
TEAC/LV	1:03	303.15	0.562	0.274	
ACC/GC	1:03	303.15	0.432	0.127	[39]
	1:04	303.15	0.432	0.133	
	1:05	303.15	0.428	0.140	
ChCl/GC	1:03	303.15	0.434	0.116	
	1:04	303.15	0.437	0.121	
	1:05	303.15	0.432	0.129	
DEH/GC	1:03	303.15	0.428	0.153	
	1:04	303.15	0.425	0.158	
	1:05	303.15	0.424	0.163	
DBU/BA	1:01	298.15	0.100	1.92–2.15	[43]
	1:04	298.15	0.100	7.70–8.55	
DBU/EG	1:01	298.15	0.100	2.80–3.08	
	1:04	298.15	0.100	12.23–12.48	
DBU/MDEA	1:02	298.15	0.100	3.84	
TBD/BA	1:01	298.15	0.100	3.64–4.04	
	1:04	298.15	0.100	8.40–8.75	
TBD/EG	1:01	298.15	0.100	4.97	
	1:04	298.15	0.100	12.90	
TBD/MDEA	1:02	298.15	0.100	3.97–4.13	
DBN/DMU	2:01	298.15	0.100	0.97	[44]
DBN/DMLU	2:01	298.15	0.100	3.94	
DBN/EU	2:01	298.15	0.100	5.23	
DBN/EU	3:01	298.15	0.100	4.39	
DBU/Pyr	1:01	303.15	0.100	0.708	[46]
DBU/Oxa	1:01		0.100	0.617	
Alanine/LA	1:01	308.15	0.494	0.279	[49]
Alanine/MA	1:01	308.15	0.493	0.346	
Betaine/LA	1:01	308.15	0.493	0.623	
Betaine/MA	1:01	318.15	0.493	0.287	
ACC/1,2,4-triazole	1:01	303.15	0.497	0.186	[50]
ACC/Imi	2:03	303.15	0.487	0.194	
	1:02	303.15	0.526	0.239	
	1:03	303.15	0.479	0.249	
[BMIM][$MeSO_3$]/U	1:01	303.15	0.423	0.245	[51]

with a mole ratio of 1:1 (0.678 g SO_2/g DES) by the authors. In addition, it was discovered that the solubility of SO_2 decreased when the HBA ratio was raised from 1:1–1:4 at constant pressure and temperature. For instance, at 293.15 K and 0.1 mPa, the solubility of SO_2 dropped from 0.678 to 0.320 (g SO_2/g DES) as presented in Fig. 6**A**. Moreover, it was discovered that the absorption capability of SO_2 reduced with increasing temperature when the DES molar ratio and pressure remained unchanged. For example, it was observed that increasing the temperature from 293.15 K to 353.15 K at 0.1 mPa, ChCl/Gly (1:1) DES caused the absorption capacity of SO_2 to drop to 0.158 from 0.678. This effect is illustrated in Fig. 6**B**. According to NMR analysis, SO_2 was absorbed physically. Under various operating conditions, the same research team investigated the solubility of SO_2 using DES constituted by 1-ethyl-3-methylimidazolium chloride (EMMIC) and EG [67]. As the EMIMC molar ratio changed from 1:2–1:1 and 2:1, it was noticed that the absorption ability of SO_2 increased (i.e., from 0.82 and 1.03–1.15 g SO_2/g DES). In a

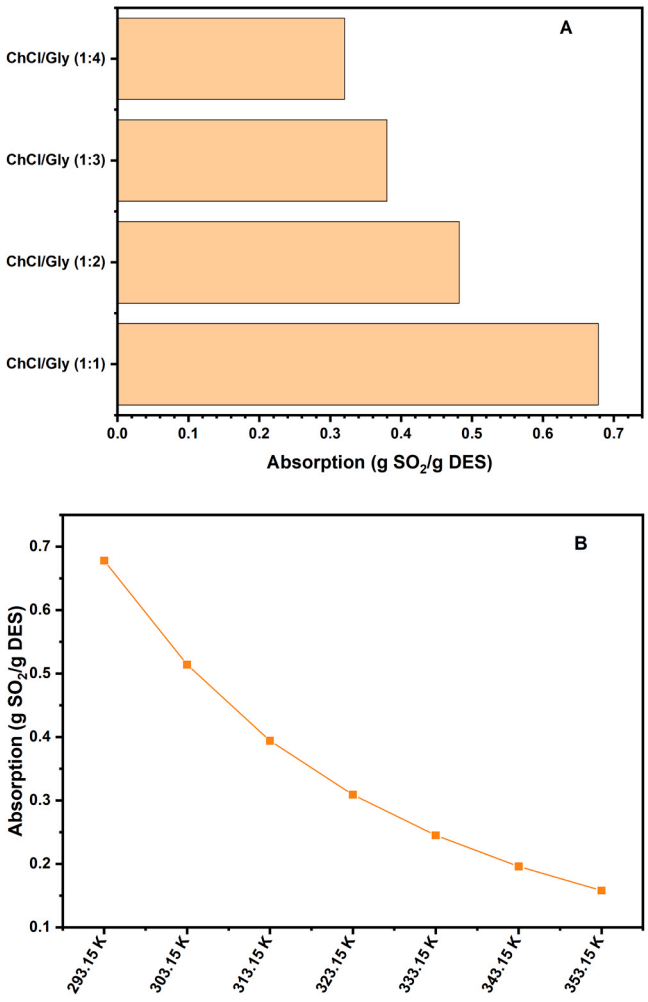

Fig. 6. (A) Effect of increasing molar ratio of HBD of ChCl/Gly on the SO_2 absorption capacity; (B) Effect of temperature on the absorption of SO_2. The data utilised for plotting these graphs were extracted from reference [66].

different investigation, the same research team prepared DESs based on EMIMC by mixing EMIMC with succinonitrile (SNT) or triethylene glycol (TEG) to examine SO_2 absorption capacity [68]. It was found that as the molar ratio of the HBA (EMIMC) increased, the SO_2 capacity for absorption also increased. For example, the maximum absorption in the EMIMC/TEG at a 6:1 molar ratio was 1.25 g SO_2/g DES of SO_2. It was also noted that as the SO_2 partial pressure dropped, the SO_2 absorption capacity decreased. EMIMC/EG (2:1) DES showed a decrease in SO_2 absorption capacity to 0.42 from 1.15 g SO_2/g solvent when there was a reduction in SO_2 pressure from 0.1 to 0.01 mPa.

Furthermore, in some cases, the solubility of SO_2 increases as the molar ratio of DES HBD increases. On the other hand, in certain circumstances, the solubility of SO_2 increases as the molar ratio of HBA increases. An illustration of this is when the solubility of SO_2 increased as the molar ratio of ChCl or EMIMC in DESs like ChCl/PhOH, ChCl/Gly, EMIMC/EG, EMIMC/TEG, and EMIMC/N-formylmorpholine (FMP) increased [69,70]. However, it was found that in ACC/Imi DES, the solubility of SO_2 increased with an increase in the imidazole molar ratio [71]. The strong interaction between the imidazole molecule and SO_2 was attributed to this behaviour. A significant shift in the molar ratio of EMIMC-based DESs and ChCl was observed in the solubility of SO_2. This is not entirely the case for all DESs, though. For example, betaine/EG and caprolactam:EG demonstrated nearly no change in SO_2 absorption at different molar ratios, such as 1:3–1:5 [1,72]. Similar to this, a change in the molar ratio of 1-hydroxyethyl-1,4-dimethyl-piperazinium bromide (PPZB)/Gly DES had little effect on the absorption capacity of SO_2 [73].

The effect of HBD of DESs composed of EMIMC/EG (1:1), EMIMC: TEG (1:1), and EMIMC:SNT (1:1), on the solubility of SO_2 was also examined by Yang et al. [74]. The trend for SO_2 solubility observed was SNT > EG > TEG. The absorption capacity of SO_2 was found to follow the order ChCl/thiourea (TU) (1:1) > ChCl/EG (1:2) > ChCl/malonic acid (MLA) (1:1) > ChC/U (1:2) at 0.1 MPa and 293.2 K using ChCl-based DESs [75]. Additionally, Zhang et al. developed four DESs based on imidazole and its derivatives to evaluate the efficiency of SO_2 capture [76]. To prepare various DESs, glycerol was mixed with Imi, 2-methylimidazole, 2-ethylimidazole, and 2-propylimidazole in a 1:2 molar ratio. The authors utilised Imi/Gly (1:2) DES to attain the highest solubility of 0.253 g SO_2/g DES for SO_2 absorption at 0.002 MPa and 313.2 K. Theoretical calculations on SO_2 capture by 1-butyl-3-methyl-imidazolium chloride (BMIMC):Acetamide (AcM), ChCl:Citric acid (CA), ChCl:EG, ChCl:Gly, ChCl:LV, ChCl:LA, ChCl:MA, ChCl:Phenylacetic acid (PA), EMIMC:AcM, and EMIMC:EG indicate that both cations and anions of HBA are significant components responsible for the good absorption capacity obtained [77].

Another significant parameter that may have an impact on SO_2 capture by DESs is temperature. Because of the high temperature of flue gas, it was discovered that high-temperature SO_2 capture with high capacity was more feasible [78]. Fast and efficient SO_2 desorption is, however, favoured by low SO_2 absorption capacity at high temperatures. In the case of ChCl/Gly (1:1) at 0.1 MPa, the absorption capacity of SO_2 was found to be, respectively, 0.678, 0.309, and 0.158 g SO_2/g DES at 293.15 K, 323.15 K, and 353.15 K [66]. Similarly, using DESs with EMIMC/EG [67], EMIMC/TEG [68], and EMIMC/SNT [74] at higher temperatures resulted in a decrease in the SO_2 absorption capacity. On the other hand, Wu and colleagues observed that when caprolactam:EG (1:3) and betaine:EG DESs were used before 40 minutes of increasing the temperature from 303.15 K to 323.15 K, there was no discernible difference in the SO_2 absorption capacity at 0.002 MPa [72]. After 40 minutes, a higher temperature was found to be associated with a lower capacity for SO_2 absorption.

It is anticipated that SO_2 capture using DESs should be carried out at various SO_2 partial pressures, most notably at low pressure, since SO_2 partial pressure in the flue gas is typically very low. According to Yang et al., at 293.15 K and 0.01 MPa and 0.1 MPa, respectively, 0.153 g SO_2 and 0.678 g SO_2/g DES were absorbed by ChCl/Gly (1:1) [66]. After raising the SO_2 partial pressure from 0.01 to O.1 MPa, the SO_2 absorption capacity by EMIMC/EG (2:1) [67] and EMIMC/TEG [68] also increased by roughly three times. The linear nature of the increased absorption capacity indicates the importance of pressure in DESs' ability to capture SO_2.

According to Chen et al., 2000 ppm of SO_2 is close to the useful concentration of SO_2 in the flue gas, so research on SO_2 capture by DESs at that partial pressure is essential [1]. According to Yang et al., at 293.15 K and 2000 ppm of SO_2, the SO_2 absorption capacities of EMIMC/SNT (1:1), EMIMC/SNT (1:2), and EMIMC/SNT (1:4) were 0.120, 0.085, and 0.051 g SO_2/g DES, respectively [74]. At 313.15 K and 2000 ppm of SO_2 partial pressure, Wu and colleagues prepared environmentally friendly DESs that could achieve a high SO_2 absorption capacity of 0.151 g SO_2/g DES by carnitine/EG (1:3) [72]. In a different study, Wu and colleagues kept other parameters the same but added an alkaline component to DESs to increase the absorption capacity of SO_2 to 0.163 g SO_2/g DES by Imi/Gly (1:2) [76]. Furthermore, at 293.15 K and 2000 ppm SO_2 partial pressure, Deng et al. found that EMIMC/FMP (1:1) absorbed 0.135 g SO_2/g DES [69].

Another important parameter that may affect SO_2 absorption is time. It is expected that in an industrial application, DESs will capture SO_2 quickly due to a short time to reach SO_2-absorbing equilibrium. According to Yang et al., within 10 minutes, SO_2 capture by ChCl/Gly [66], EMIMC/EG, [67], EMIMC/TEG [68], and EMIMC/SNT [74] was nearly constant. The ability to absorb SO_2 increased quickly over the course of 10 minutes. When betaine/EG (1:3) and caprolactam:EG (1:3) DESs are used, it takes 80 minutes to reach a steady state for SO_2 capture at 0.02 MPa [72]. Additionally, imidazole-based DESs required about 150 minutes to reach equilibrium in order to capture SO_2 [76]. While time does not always impact SO_2 absorption, the gas flow rate does have an impact on how long it takes to reach absorption equilibrium. However, it is important to remember that DESs may be more easily volatilised in the event of a high SO_2 flow rate [76].

While using DESs, water may also have an impact on SO_2 absorption. Water is a universal substance and is readily absorbed by DESs due to its high hygroscopicity [41]. According to Deng et al., the presence of 5.0 and 10.0%wt water in ChCl/LV slightly reduced the SO_2 absorption capacity while keeping the absorption rate constant [79]. Similarly, it was found that EMIMC/FMP DES's ability to capture SO_2 was barely affected by the presence of 2.0%wt water [69]. It might be a result of the hydrophilic nature of ChCl/LV DES, which increases the hydrogen bonding interaction between ChCl/LV and water. Hence, there is less of an interaction between SO_2 and ChCl/LV. Furthermore, if hydrophobic DESs were used to capture SO_2, the outcomes might differ. However, it was discovered that adding 10% weight of water improved mass transfer by lowering viscosity. Consequently, even though BMIMC/Imi and BMIMC/4-methylimidazole (MImi) DESs slightly reduced SO_2 capacity, there was a higher rate of reaching saturation point [80]. Additionally, Zhao et al. discovered that increasing the amount of water added to DES constituted by acetamide and EMIMC reduces its ability to absorb SO_2 [81].

Furthermore, Hu and colleagues also investigated the use of acetamide and imidazolium halides in DESs to achieve outstanding SO_2 capture [82]. Strong charge transfer interaction between Cl^- and absorbed sulfide allows for the above-mentioned effective capture of SO_2. Investigation of DESs with higher Cl^- content for efficient SO_2 solubility is encouraged by the earlier studies. In addition, optimising the structure of HBDs would also efficiently raise the SO_2 absorption capacity. As of right now, some studies have demonstrated that DESs with coordination-unsaturated Lewis-alkaline N sites like those based on amides and 1,2,4-triazoles would have a significant capacity to absorb SO_2 [83,84]. As a result, Lewis-alkaline DESs with high Cl^- content may have the best potential and ability to absorb SO_2.

In order to improve the capture of SO_2, research by Wang et al. focuses on preparing DESs with pyridine-based functionalisation that have a lot of Lewis-alkaline and Cl^- sites [85]. With a capture of 1.247 g

SO_2/g DES at 298.15 K and 1.0 bar, the EMIMC:2-aminopyridine (2-NH_2Py) DES at 7:1 molar ratio demonstrated exceptional efficiency in SO_2 capture. The DESs were found to have an absorption equilibrium time of roughly 45 seconds, which is a noteworthy improvement over other absorbents that have equilibrium times varying between 10 and 30 minutes. Additionally, the DES demonstrated outstanding recycling performance, retaining its capacity to absorb SO_2 for at least 40 cycles. Moreover, EMIMC/2-NH_2Py (7: 1) showed stability in the presence of water and over many months, demonstrating its robustness. To verify the emergence of Lewis acid-base and $SO_2\cdots Cl^-$ covalent interactions within the DES, the study employed FT-IR and NMR measurements. Recently, three dihydric alcohol-based DES was applied for the capture of SO_2 gas by Liu et al. [86]. In this study, DES composed of tetraethylene glycol (TeEG) and 1-ethyl-3-methylimidazolium chloride (EmimCl) at 1:2 displayed the best SO_2 adsorption capacity of about 13.25 mol·kg^{-1} and outstanding selectivity of 29.9 for SO_2 to CO_2 at 298.2 K and 1.0 bar.

3.2. Capturing of H2S

In order to meet gas product quality standards, H_2S must be selectively removed from natural gas. H_2S is a colourless, extremely toxic, acidic gas with an unpleasant rotten egg odour that lowers fuel efficiency and market potential [87,88]. The commonly used methods for capturing H_2S include adsorption, biodegradation, and amine scrubbing [89–91]. These technologies still have issues, though, like poor H_2S uptake, expensive regeneration, and easily corroding pipeline apparatus [92]. Since the traditional H_2S absorbers are known to rely heavily on chemical interactions with H_2S, they have a high absorption enthalpy and require a significant amount of energy to regenerate [93]. Therefore, a major advancement in H_2S separation technology requires lowering the energy consumption needed for absorbent regeneration and DESs are the ideal adsorbents that are currently trending.

Few research publications have shown that DESs can effectively capture H_2S. Unless otherwise indicated, the common international unit for comparing H_2S absorption capacity is mole H_2S/kg DES. The best way to remove H_2S is by wet oxidation using iron-based DESs. In this instance, $Fe(NO_3)_3{\cdot}9\ H_2O$ salt is needed and ferric salt is used as the oxidant [94]. The following trend was observed for H_2S solubility in DESs at 298 K and 0.5 MPa, TBAB/propionic acid (PrA) (1:1) > TBAB/AA (1:1) > TBAB/formic acid (FA) (1:1); at 298 K and 0.5 MPa, ChCl/PrA (1:1) > ChCl/AA (1:1) > ChCl/FA (1:1)). It is clear that HBA plays a part in H_2S solubility. Compared to ChCl-based DESs, TBAB-based DESs showed higher H_2S solubility for the same HBD. This behaviour can be explained by the fact that DESs based on TBAB have a weaker hydrogen bond than DESs based on ChCl. Furthermore, the presence of a hydroxyl group in ChCl suggests that the hydrogen bonding interactions in DESs based on this compound are more complex than those in DESs based on TBAB. At lower temperatures and higher pressures, TBAB/PrA and ChCl/PrA DESs may be able to increase their H_2S absorption capacity through a physical process. Moreover, regeneration can be carried out three times at 353.15 K for 240 minutes during an N_2 purge without any discernible loss of NH_3 capacity [94].

The H_2S absorption capacity is also influenced by the mole ratio of HBA like other gases discussed earlier [2]. For example, at a constant temperature and pressure, it was found that as the molar ratio of ChCl/urea DES increased from 1:1.5–1:2.5, the H_2S absorption capacity also increased. It was observed that as temperature increases, the H_2S absorption capacity in ChCl/urea DESs decreases linearly. Moreover, CH_4 can be efficiently and selectively captured using DESs [95]. Mao et al. have improved DESs recently by loading DESs with fumed silica to capture H_2S [96]. Triethylamine hydrochloride (TEAHC) and cupric chloride ($CuCl_2$) were mixed at a 1:1 molar ratio to form the DES that was used. The maximum H_2S capacity that the authors were able to achieve at a 10% DES loading rate and 303.2 K temperature was 9.97 mg/g DES. Compared to TEAHC or $CuCl_2$, TEAHC/$CuCl_2$ (1:1) DES was found to be a more effective loading material.

3.3. Capturing of NH_3

N_2O, nitrate, and toxic ammonium can be generated by reactions between NH_3, which is present in large quantities in the atmosphere and a range of bioprocesses. Strong greenhouse gas N_2O counteracts the reduction in greenhouse gasses from burning NH_3 instead of carbon-based fuels [97,98]. Furthermore, particulate matter 2.5 ($PM_{2.5}$) refers to fine particles that are smaller than 2.5 microns in diameter, and nitrates account for a significant portion of this category. Given that NH_3 plays a significant role in the formation of nitrate, the air may contain more $PM_{2.5}$ due to the rapidly expanding gaseous NH_3 emissions [99]. The mitigation and remediation strategies for atmospheric ammonia must expand per the ammonia industry's growth, given its diverse applications and potential for decarbonisation [97]. To identify, isolate, and collect ammonia from leaks, rogue or fugitive emissions, combustion flue gas, and even straight from the atmosphere, several technologies have been developed [98]. Nevertheless, the use of DESs for remediating NH_3 is trending and is therefore discussed here.

According to a study conducted by Li et al., ChCl-based DESs are not very effective at absorbing NH_3 [100]. For instance, 0.01 g NH_3/g of DES is absorbed by ChCl:U 1:2. Conversely, the same author has reported complex DESs with high NH_3 solubility, including ChCl and two types of HBDs [100]. Using resorcinol/Gly (new components) as HBDs, for example, ChCl-based DES can absorb up to 0.13 g of NH_3 per g of DES at 313 K and 0.1 MPa. Considering the natural origin of the DESs, their biodegradability and affordability lend credence to their potential application in the NH_3 absorption process. Additionally, at 313 K, ChCl: tetrazole:EG DES showed 0.17 g NH_3 per g DES as NH_3 absorption. Therefore, taking into account a wide variety of HBDs, a high NH_3 capturing ability has also been reported for ChCl-based DESs [101]. High solubilities for NH_3 are also produced by DESs based on other compounds, such as those based on amines [102] or azoles [103]. DESs are superior to conventional solvents or ILs as platforms for the development of NH_3 absorption operations, even when taking into account basic ChCl-based DESs [100].

Metal chlorides have been investigated by Cheng et al. as a means of supplying extra weak acidic sites for NH_3 interactions [104]. They used different ratios of glycol, several metal chlorides, and ethylamine hydrogen chloride (EHCl) to prepare DESs. With one of the examined DES, that is $CoCl_2$ based DES, the absorption results were remarkable even at lower pressures, with a 10.24 mmol/g at 6.8 kPa. Also, a new subgroup of DESs called Type V DESs has come under investigation [105]. The fact that neither Type V substance contains any ionic components sets it apart from the other four categories of DESs. Less viscous solutions with significant ideality deviations that are readily regenerable by evaporation result from the absence of ionic components [106]. A Type V DES 3-hydroxypyridine/PhOH at a molar ratio of 1:5 can absorb 10.0 mmol/g of NH_3 at 1 bar as reported by Zhou et al. [107]. When compared to other recent DESs in literature, this is a significant improvement.

According to Wang et al., at 293 K and 1 bar, ternary DESs can achieve NH_3 absorption of 0.245 g·g^{-1} [108]. Luo et al. have prepared a series of DESs recently using resorcinol (Res) as the HBD and imidazole or ethanolamine hydrochloride as the HBA for NH_3 absorption [12,109]. High NH_3 absorption capacity was demonstrated by these DESs with moderate viscosity and good thermal stability. This is explained by the addition of active protons and the creation of hydrogen bonds between –OH or –NH_2 in the DESs and NH_3. These results offer a fresh perspective for upcoming efforts to construct novel NH_3 absorbents. On the other hand, halogen ions found in many DESs used for NH_3 absorption can exacerbate metal equipment corrosion. Shao et al. have therefore lately investigated a wide range of halogen-free DESs for NH_3 capture [110]. According to their research, DES composed of amino-2-propanol nitrate [APH]NO_3 and Res prepared at a 2:1 ratio demonstrated exceptional

NH_3 absorption performance of 0.253 $g{\cdot}g^{-1}$ at 293 K and 1 bar. Recently, at 313.15 K and 0.1 MPa, 2-NH_2Py/Res DES demonstrated excellent NH_3/CO_2 selectivity of 92.2, outstanding thermal stability, and a high NH_3 absorption capacity of 0.185 g NH_3/gDES [111]. The high acidity of Res HBD is responsible for the outstanding results obtained.

3.4. Capturing of NO_2

For decades, there have been significant efforts made to lower the atmospheric NO_2 concentration, making NO_2 control more crucial and urgent [112]. For example, severe limits on NO_x emissions from cars and other combustion sources have prompted the development of various catalytic processes to lower NO_x emissions, such as NOx storage and reduction, and selective catalytic reduction, both of which are only effective at high temperatures [113]. Nevertheless, NO_2 removal at room temperature continues to be difficult. Catalysis-based techniques are unable to effectively reduce NO_2 produced during low-temperature processes such as the cold start of diesel engines, which occurs between 80^0 C and 200^0 C, which results in a significant amount of NO_2 emission into the atmosphere [114]. Techniques involving the use of DESs for removing NO_2 are some of the most current efficient methods in practice.

DESs have also been utilised to absorb NO_2 gas, though the literature on this topic is extremely limited. The absorption capacity of NO_2 for ChCl-based DESs in various HBDs (such as urea, methylurea, and thiourea) was investigated by Waite et al. using theoretical studies [115]. Urea was formed through charge-dipole interactions when used as HBD. Therefore, NO_2 capturing could be done with these DESs. Using ChCl/Gly and ChCl/EG, Chen et al. practically studied the absorption of NO_2 [116]. The values obtained fell within the range of 0.36−0.55 g NO_2/g DES. At a 1:4 molar ratio, the DES composed of ChCl/EG showed the highest absorption capacity. According to the results, there was an increase in the solubility of NO_2 in the DES when the amount of HBD was increased. This makes sense because the NO_2−HBD interaction is stronger than that of ChCl (HBA) [115]. Therefore, by carefully choosing HBDs that can interact strongly with the gas molecules, the solubility of NO_2 in DESs can be enhanced.

3.5. Capturing of NO

It is crucial to develop technologies for reducing NO emissions. Selective catalytic reduction is the most developed method of industrial denitrification. It uses NH_3 and precious metal catalysts at high temperatures to convert NO into N_2 [117–119]. However, the NO_x content of post-selective catalytic reduction exhaust gas is finding it difficult to comply with ever-tougher NOx emission standards. Furthermore, there is an unsustainable conversion of NH_3 and NO to N_2, and the production of NH_3 consumes a significant amount of energy [120,121]. Wet oxidation-absorption technologies have also been applied recently to remove NO. In this process, strong oxidants such as O_3, H_2O_2, and $Na_2S_2O_8$ oxidise NO, which is then removed by basic absorbents [122, 123]. Low-value mixtures of nitrates and bases are frequently the end products of denitrification processes, and the oxidants and absorbents are not readily available or regenerable. The utilisation of DESs for removing NO has proven to be superior to the existing techniques.

Mole NO/mole DES is the unit used to measure NO absorption capacity. Tantai research group studied the NO absorption capacity of DESs using 1,3 dimethylthiourea (1,3-DMTU)-based DESs [124]. At a 1:1 molar ratio of 0.1 MPa and 303.15 K, the adsorption capacity followed the following trend 1,3-DMTU/tetrabutylphosphonium chloride (TBPC) > 1,3-DMTU/TBAC > 1,3-DMTU/ tetrabutylphosphonium bromide (TBPB) > 1,3-DMTU /TBAB. The 1,3-DMTU/TBPC molar ratios went from 1:1–1:3, and the system consisting of 1,3-DMTU/TBPC (3:1) showed the best NO capture performance (4.25 mol. NO per mol DES). This pattern suggests that by deprotonating DESs, 1,3-DMTU/TBPC DESs could effectively absorb NO. Their findings indicate that 1, 3-DMTU-based DESs needed roughly 8 minutes to reach the corresponding saturation of NO absorption [124]. In a similar vein, the same order for NO capture by DESs was obtained when the HBA (1,3-DMTU) was switched to another HBA. Tetrazole/TBPC > Tetrazole/TBAC > Tetrazole/TBPB > Tetrazole/TBAB, for instance, was the same order obtained when "1,3-DMTU" was substituted with "tetrazole" [125]. Furthermore, NO absorption was induced by hetero-nitro atoms. It was discovered that NO absorption decreased at high temperatures and low pressure.

According to Wu et al., amine-based DESs showed good absorption capacity and rate for NO capture (e.g., HBD: EG, glycerol, 1,3-propanediol, and polyethylene glycol 2000; HBA: triethylenetetramine chloride and tetraethylenepentamine chloride) [126]. Water was found to have a negligible effect on NO capture by DESs, even though it lowers viscosity for effective mass transfer. On the other hand, O_2 might marginally reduce NO absorption. Using an N_2 purge, the regeneration of DESs was achieved at least five times at 351.15 K. Theoretically, NO absorption efficiency was investigated using arginine-based natural DESs (NADES), suggesting NADES as a viable substitute for NO absorption [127]. However, the lack of theoretical and experimental data necessitates the development of DESs specifically with features suited for NO absorption.

4. DESs selectivity for gas capturing

SO_2 and CO_2 are the two common acidic gases found in flue gas. Consequently, it is essential to separate SO_2 from mixed gases that include CO_2 and SO_2. According to Ren et al., there is CO_2 in flue gas, so getting DESs to absorb SO_2/CO_2 with more selectivity is crucial [128]. According to Deng et al.'s investigation, SO_2 could be selectively captured in a CO_2/SO_2 mixture by using ammonium-based DESs (selectivity = 134–199) [79] and ChCl/Phenol (PhOH) [70]. This is explained by the fact that SO_2 has a higher acidity than CO_2 [69]. The highest selectivity value of 33.1 for SO_2 to CO_2 could be achieved by PPZB/Gly, according to the work of Cui et al. [73]. Furthermore, EMIMC/ 2-NH_2Py (7:1) showed strong selectivity for SO_2 over CO_2 (312/1), indicating that it could be used in CO_2-rich environments [85].

Selectivity of NH_3 from NH_3, CO_2, and H_2S mixture is also conceivable since industrial waste gas may contain all three simultaneously. Thus, a high degree of selective gas absorption is required. Given that NH_3 is more toxic than CO_2 and that they coexist in industrial streams, there should be a great deal of focus on the more selective NH_3 capture. The strong hydrogen-bond supramolecular network in DESs is responsible for the high selectivity of 142 for NH_3 in the NH_3/CO_2 mixture which was observed in a study by the research team of Ren [100]. Vorotyntsev et al. concluded that there is physical gas absorption in 1-butyl-3-methyl imidazolium methanesulfonate ([BMIM][$MeSO_3$])/U DES [51]. The obtained solubility trend shows the highest selectivity for NH_3 absorption: $NH_3 > H_2S > CO_2$. Zhong et al. achieved an exceptional selectivity of NH_3/CO_2 (284−611) using ChCl/tetrazole/EG ternary DES [129]. Similarly, Deng and colleagues used 1,2,4-triazole/glycerol DES to achieve a very high NH_3/CO_2 selectivity of 216.3 [103].

CO_2 and H_2S are present in natural gases together. H_2S poses a significant risk, whereas CO_2 may reduce the caloric value. Compared to CO_2, H_2S is more detrimental when it comes to natural gas. For this reason, H_2S must have a higher selectivity than CO_2. Using TBAB/PrA and ChCl/PrA DES, there was less H_2S/CO_2 selectivity observation [103]. However, due to the minimal solubility of CH_4, CO, and H_2 in DESs, the selectivity of H_2S via CH_4, CO, and H_2 was extremely high [130].

5. Gas capture mechanism by DESs

The capture mechanism of gas in DESs is a multifaceted interplay of physical and chemical processes. A compelling illustration of this phenomenon is evident in the case of TeG/EmimCl DES during SO_2 capture. So, in SO_2 capture experiments involving TeEG/EmimCl DES, the

absorption mechanism was ascribed to the involvement of chloride ions and ether bonds [86]. To prove this, the same authors conducted ^{1}H NMR, ^{13}C NMR and FT-IR analyses of TeEG/EmimCl DES before and after SO_2 absorption. The ^{1}H NMR spectra revealed noteworthy shifts in the signals of hydrogen atoms on EmimCl after SO_2 absorption, indicative of charge-transfer interactions (Fig. 7**A**). Specifically, the signals of a and b hydrogen atoms shifted from 10.37 and 7.45 ppm to a higher field at 9.27 and 7.426 ppm. This phenomenon results from the quasi-chemical interaction of $SO_2\cdots Cl^-$, thereby reducing the electron density of a and b atoms [131,132]. Conversely, the signals of TeEG exhibited minimal shifts, signifying the involvement of ether bonds in physical absorption. Remarkably, the chemical shifts reverted to their original positions upon SO_2 recovery, underscoring the reversible nature of the process. In the ^{13}C NMR analysis (Fig. 7**B**), the signals of all carbon atoms in TeEG/EmimCl (1:2) barely shifted after SO_2 absorption, corroborating that the capture of SO_2 predominantly relies on physical absorption. This aligns with thermodynamic fitting results, further supporting the distinction between physical and chemical absorption.

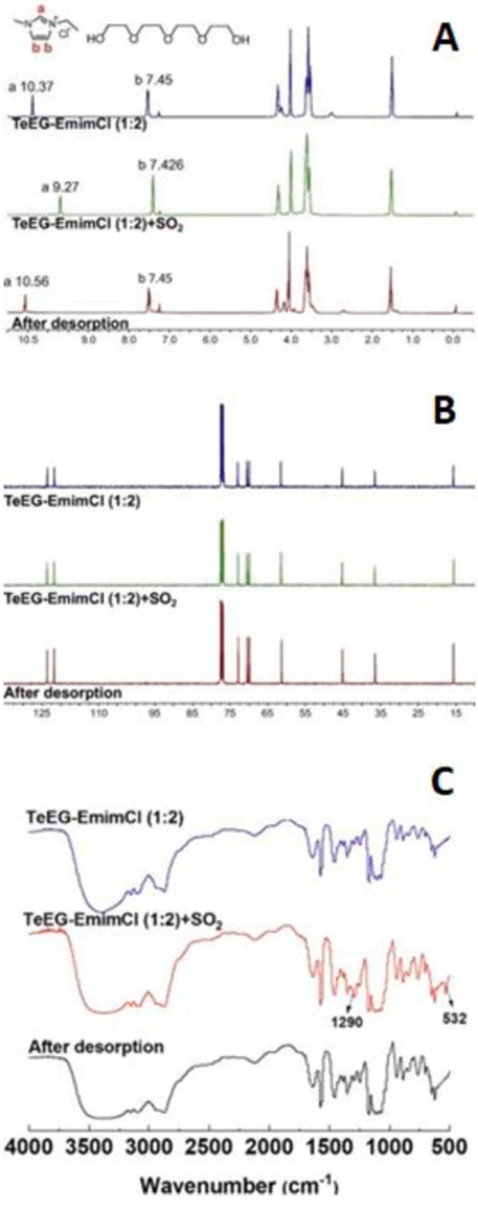

Fig. 7. The NMR and FTIR spectra for TeEG/EmimCl (1:2) DES before and after SO_2 capture. Adapted from [86], Copyright 2023 with permission from Elsevier.

To further understand the absorption mechanism, FTIR spectra of TeEG/EmimCl (1:2) before and after SO_2 absorption were examined by Liu et al. (Fig. 7**C**). Post-SO_2 absorption, the emergence of new absorption peaks at 532 and 1290 cm^{-1} unveiled crucial insights. The peak at 532 cm^{-1} corresponds to the asymmetric stretching of SO_2, confirming its physical absorption [133]. Simultaneously, the peak at 1290 cm^{-1}, attributed to the bending vibration of SO_2, indicates the presence of $SO_2\cdots Cl^-$ interactions [134]. Upon regeneration, the disappearance of characteristic absorption peaks of SO_2 confirmed the complete resolution of SO_2. In conclusion, the absorption mechanism of a typical gas using DES is both physical and chemical, with physical absorption playing a dominant role. This comprehensive understanding contributes to the elucidation of DES applications in gas capture processes.

6. Comparison of gas capture by DESs with other materials

Table 2 compiles and shows the NH_3 absorption capacities of several solid and liquid materials, such as DES, IL, porous carbon, zeolite, metal-organic frameworks (MOFs), covalent organic frameworks (COFs), and the complex of the aforementioned materials. Compared to 2-NH_2Py/Res (1:2), solid materials like MG-2-EM and COF-10 have higher capacities for NH_3 absorption; however, because of their stronger acidity and interaction with NH_3, these materials have a harder time achieving complete regeneration [111]. The NH_3 absorption capacities of metal-based ILs (like $[Bmim]_2[CuCl_4]$ and $[Emim]_2[Co(NCS)_4]$) and protic ILs (like [EtOHim][SCN] and [EtA][SCN]) are higher than those of 2-NH_2Py/Res (1:2). They are, however, constrained by expensive or complex synthesis. In comparison to pyridinium-based ILs, including $[Py][NTf_2]$, $[2\text{-}mPy][NTf_2]$, and phenol-based DESs Im/Res (1:1), ChCl/Res/Gly (1:3:5), EaCl/PhOH (1:2), and ChCl/PhOH/EG(1:7:4), 2-NH_2Py/Res (1:2) has greater NH_3 absorption capacities. Furthermore, pyridine derivatives are less expensive than typical HBAs [111]. Overall, the comparison results point to the 2-NH_2Py/Res DES system's effective NH_3 capture performance, and the new type of HBA known as pyridine derivative has a lot of promise for synthesizing DESs for the storage and capture of NH_3.

7. Regeneration of DES

A crucial aspect to consider in the gas absorption process is the absorbents' ability for recycling and regeneration, pivotal for the industrialization of toxic gas capture. Typically, the separation of toxic gases from DESs is conducted under high-temperature and/or low-pressure conditions. Hence, the stability and volatility of DESs are significant factors to consider. At high temperatures, DESs typically first break

Table 2
1 MPa.

Absorbent/ adsorbent	Temperature (K)	NH_3 Capacity[a] (g/g)	References
UiO-66-OH	293.15	0.096	[135]
MG-2-EM	293.15	0.231	[136]
COF-10	303.15	0.255	[137]
[EtOHim][SCN]	313.15	0.221	[138]
$[Py][NTf_2]$	313.15	0.140	[139]
$[2\text{-}mPy][NTf_2]$	313.15	0.140	[139]
$[Emim]_2[Co(NCS)_4]$	303.15	0.198	[140]
$[Bmim]_2[CuCl_4]$	303.15	0.172	[141]
[EtA][SCN]	313.15	0.240	[142]
ChCl/Res/Gly (1:3:5)	313.15	0.129	[143]
EaCl/PhOH (1:2)	313.15	0.119	[144]
ChCl/PhOH/EG (1:7:4)	313.15	0.130	[145]
Im/Res (1:1)	313.15	0.154	[12]
2-NH_2Py/Res (1:2)	313.15	0.163	[111]
2-NH_2Py/Res (1:2)	303.15	0.185	[111]

Adapted from [111], Copyright 2024, with permission from Elsevier.
[a] g NH_3 per g absorbents or adsorbents

down into HBDs and HBAs as a result of the hydrogen bond interactions becoming weaker. The HBDs that have low boiling points or poor stability then experience volatilisation or decomposition, while the HBAs do the same thing but at a higher temperature. ChCl, the most widely used HBA, for instance, starts to break down at about 250 °C. A key factor in the thermal stability of DESs is the hydrogen bond. Compared to pure HBAs and HBDs, it hinders the "escape" of molecules and requires more energy to break [146,147]. Hence, it is crucial to investigate the thermal stability of DESs before carrying out the regeneration process.

Therefore, in the pursuit of exploring the regeneration capabilities of the Im/Res (1:1) DES, Luo et al. first investigated the thermal stability of the DES without gas absorption [12]. Their findings revealed that, under desorption conditions set at 0.15 kPa and 353.15 K for 10 hours, Im/Res (1:1) exhibited a minimal weight loss of 0.628%. This result underscores the commendable thermal stability of the DES during desorption. The performance of Im/Res (1:1) DES was further assessed through five NH_3 absorption–desorption cycles, as illustrated in Fig. 8**A**. Remarkably, the NH_3 absorption capacity was observed to remain nearly constant across the cycles, indicating the robust reversibility of the binary DES. This finding holds significant implications for the sustained effectiveness of Im/Res (1:1) over multiple absorption-desorption cycles. Complementary analyses using FT-IR and ^{1}H NMR spectra were conducted after the first and fifth cycles as depicted in Fig. 8 (**B and C**), respectively. Notably, these spectra exhibited minimal changes when compared with those of the fresh DES. This consistency confirms that the absorbed NH_3 can be completely released under desorption conditions. The observed lack of significant alterations in the spectra underscores the successful recovery of Im/Res (1:1) DES activity without any noticeable loss after repeated cycles.

8. Future directions

The evolving utilisation of DESs for gas capture presents both challenges and exciting opportunities that warrant focused attention in future research endeavours. Several key areas emerge as focal points for advancement.

Despite the promising results shown by various DESs in gas capture, several common issues persist. Many DESs employed for gas capture contain halogen ions, leading to increased corrosion of metal equipment. Additionally, the strong interaction between DESs and gas molecules results in heightened energy consumption during absorber regeneration. It is imperative to develop halogen-free DESs with a hydrogen bond network for efficient gas capture. Future research should prioritise the design of new, cost-effective, and environmentally friendly DESs with higher gas capacity and low viscosity, enhancing their appeal in gas separation applications. Furthermore, conducting techno-economic research on the use of DESs in gas separation, alongside the development of models for predicting DES properties and gas absorption capacity, is essential for advancing this field.

Most of the published research centres around simulated gas capture in flue gas streams, which may not fully represent real-world gas streams. In actual flue gas streams, besides the targeted gas, there are additional components such as water and other gases. Therefore, it is essential to develop DESs that exhibit true selectivity for the desired gas while demonstrating high resistance to other components. Notably, previous studies on gas capture using DESs have not undertaken a life cycle assessment (LCA) of the process. A comprehensive LCA of DES-based gas capture processes is crucial for understanding environmental impact, energy consumption, and economic implications

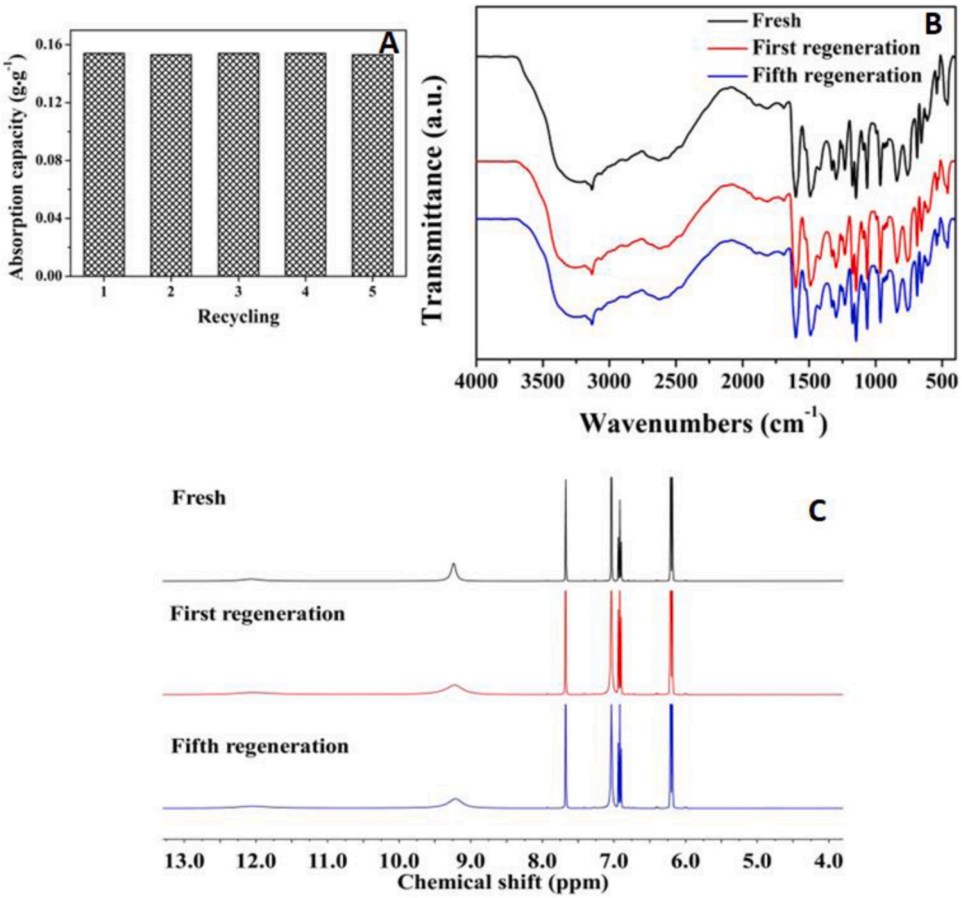

Fig. 8. (A) Five NH_3 absorption–desorption cycles; (B) FT-IR spectra of fresh, first regeneration and fifth regeneration; (C) ^{1}H NMR spectra of fresh, first regeneration and fifth regeneration of Im/Res (1:1) DES.
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throughout the entire life cycle. This knowledge will facilitate more informed and sustainable deployment of DESs in gas capture technologies.

The future of DESs for gas capture holds great promise, but addressing the identified challenges and leveraging emerging opportunities will require sustained research efforts, collaboration, and innovative thinking. By strategically focusing on these future perspectives, the field can advance towards more efficient, sustainable, and scalable solutions for gas capture using DESs.

9. Concluding remarks

In conclusion, this review article discusses the gas absorption and solubility performances of a diverse range of DESs. The findings encompassed crucial structural factors such as HBDs, HBAs, molar mixing ratios, and water content, along with key process parameters including time, temperature, and pressure. This work reveals that elevated pressure enhances gas solubility in DESs, while an increase in HBD ratio and high temperature decreases gas solubility within DESs. The introduction of a small amount of water to DESs marginally enhances gas absorption. Interestingly, a notable correlation emerges, linking the alkyl chain length of the DES HBA with gas solubility, indicating that an increase in alkyl chain length corresponds to an elevation in DES free volume and, subsequently, increased gas solubility. Moreover, this review article establishes a compelling linear correlation between the Hammett basicity values of DESs and the absorption of gas. This correlation highlights the substantial influence of DES basicity on initial gas levels, suggesting that heightened basicity enhances the efficiency of gas capture. This finding provides valuable insights into the deep relationship between DES characteristics and their interactions with gas, influencing our comprehension and manipulation of gas capture processes.

Additionally, deepens our understanding of the selectivity exhibited by DESs in capturing specific gases, revealing the pivotal role of the robust hydrogen-bond supramolecular network within DESs. This selectivity is exemplified in the effective capture of NH3 within NH_3/CO_2 mixtures and the targeted capture of SO_2 in CO_2/SO_2 mixtures using ammonium-based DESs. Furthermore, the gas capture mechanism in DESs involves a multifaceted interplay of physical and chemical processes. The absorption mechanism, predominantly physical in nature, underscores the dominant role of physical absorption. In comparison to DESs, solid materials like MG-2-EM and COF-10 exhibit higher gas absorption capacities. However, their stronger acidity and interactions with certain gases pose challenges in achieving complete regeneration. This nuanced understanding of gas capture mechanisms in DESs and solid materials contributes to the broader landscape of sustainable gas capture strategies. In essence, the insights derived from this comprehensive review contribute to our understanding of the intricate interplay between DES properties and gas absorption, providing a foundation for future research and practical applications in environmental and industrial contexts. The potential for tailored selective gas capture strategies using DESs represents a promising avenue for addressing environmental challenges and advancing sustainable solutions.

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Conflict of Interest

The author declares that none of the work reported in this paper could have been influenced by any known competing financial interests or personal relationships.

Data Availability

No data was used for the research described in the article.

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