

# Carbon dioxide separation performance evaluation of amine-based versus choline-based deep eutectic solvents

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## ABSTRACT

Carbon capture technology can address the fundamental causes of carbon dioxide emissions. Deep eutectic solvents stand out among the several existing, due to their tunability, low viscosity, high carbon dioxide absorption, low cost, and environmental friendliness. In this review, the structural characteristics of amine-based deep eutectic solvents are summarized. Next, the physicochemical properties and influencing factors are analyzed. A comparative analysis of the absorption performance of amine-based deep eutectic solvents is carried out, and the regeneration performance is recorded. It is crucial to note that this is the first time data for thermal conductivity has been compiled. Additionally, outstanding amine-based deep eutectic solvents have been selected. Subsequently, we compare amine-based deep eutectic solvents with choline-based deep eutectic solvents. The findings demonstrate that superior amine-based deep eutectic solvents prefer compounds containing chloride ions as hydrogen bond acceptors and alkanolamines with short alkyl chains as donors. The performance of amine-based deep eutectic solvents is extremely comparable to that of choline-based deep eutectic solvents. Specifically in terms of carbon dioxide absorption, viscosity, and melting point, amine-based deep eutectic solvents outperform choline-based deep eutectic solvents for post combustion CO<sub>2</sub> capture. Finally, it is concluded that the upcoming primary emphasis of amine-based deep eutectic solvents will consist mostly of research into reaction kinetics and energy consumption, which could offer statistics for the design of reaction devices.

## 1. Introduction

Global warming and energy deficiency are the pressing issues of our time [1]. In addition to causing the greenhouse effect [2], global warming directly causes the melting of glaciers and indirectly causes frequent cases of extreme weather, food insecurity, and disease [3]. Due to greenhouse gases, the average global temperature in 2022 was approximately 1.15 °C higher than the baseline between 1850 and 1900 [4]. However, it is primarily attributable to high carbon dioxide emissions [5]. In 2022, the concentration of carbon dioxide gas surpassed 415.7 ppm [6]. In order to mitigate CO<sub>2</sub>, Carbon Capture, Utilization and Storage (CCUS) technologies have emerged.

Carbon capture technology is a method used to address carbon emissions issues at their source. The present carbon capture technologies primarily involve absorption, adsorption and cryogenic separation

technologies, and so on. Excellent adsorbents are essential for the adsorption process.

Regrettably, the presence of impurities in flue gas has been found to impair the absorption capability of solid adsorbents and to raise operating costs [7–9]. Using the effect of water present in flue gas as an example, it can be noted that, typically, the water content in flue gas lies within the range of 5–12 vol% [10]. This is significant as the presence of water has been shown to reduce the CO<sub>2</sub> solubility of the adsorbents in question. Hammache et al. [7] demonstrated that water re-agglomerates the polyethyleneimine (PEI) impregnated in SiO<sub>2</sub>, resulting in a decrease in CO<sub>2</sub> adsorption capacity. Modak et al. [8] demonstrated that water causes pore deformation in metal-organic frameworks (MOFs), which results in reduced adsorption capacity for CO<sub>2</sub>. In addition, Hammache et al. [7] indicated that, in industry, dewatering prior to flue gas treatment increases operating costs. Cryogenic separation has the advantages of avoiding large quantities of chemical additives, preparing

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### Nomenclature

[BTEA]Cl	Benzyltriethylammonium chloride
[MEA] Cl	Monoethanolamine hydrochloride
[TAE]Cl	Thioacetamide hydrochloride
[TEA] Cl	Triethanolamine hydrochloride
[TETA]Cl	Triethylenetetramine Dihydrochloride
[UE] Cl	Urea hydrochloride
AEP	N-Aminoethylpiperazine
AP	3-Amino-1-propanol
Ch [Pro]	Choline hydroxide L-proline
ChCl	Choline chloride
DEA	Diethanolamine
DETA	Diethylenetriamine
EAE	2-(Ethylamino)ethanol
EDA	Ethylenediamine
EG	Ethylene Glycol

Gly	Glycerol
LvAc	Levulinic acid
MDEA	N-Methyldiethanolamine
MEA	Monoethanolamine
MTPB	Methyltriphenylphosphonium bromide
PEG	Poly (ethylene glycol)
PhoAc	Phenylacetic acid
PZ	Piperazine
TBAB	Tetrabutylammonium bromide
TBAC	Tetrabutylammonium chloride
TEA	Triethanolamine
TEAB	Tetraethylammonium Borohydride
TEAC	Tetraethylammonium chloride
TEPA	Tetraethylenepentamine
TETA	Triethylenetetramine
TMAC	Tetramethylammonium chloride

high-pressure, high-purity carbon dioxide products, and storage capacity [11]. Lack of a suitable cryogenic source and a strong reliance on capture efficiency on the operation temperature are the drawbacks of the cryogenic absorption technique [12,13]. The absorption technique has been widely applied in various petrochemical industries with mature technology. As a CO<sub>2</sub> solvent, the aqueous amine solution is widely employed. Among the most widely available amine solvents are monoethanolamine (MEA), diethanolamine (DEA), triethanolamine (TEA), diethanolamine (DGA), and methyl diethanolamine (MDEA) [14]. The concentration of aqueous amine solution is 25–30 wt% [15], and its viscosity is very low, so the mass transfer effect is good. Also, the aqueous solutions have high CO<sub>2</sub> solubility, for example the CO<sub>2</sub> solubility of 30 wt% MEA achieved 0.118 g/g at 303 K and 1 atm [16], whereas the CO<sub>2</sub> absorption of Choline chloride/Ethylene glycol is only 0.013 g CO<sub>2</sub>/g [17].

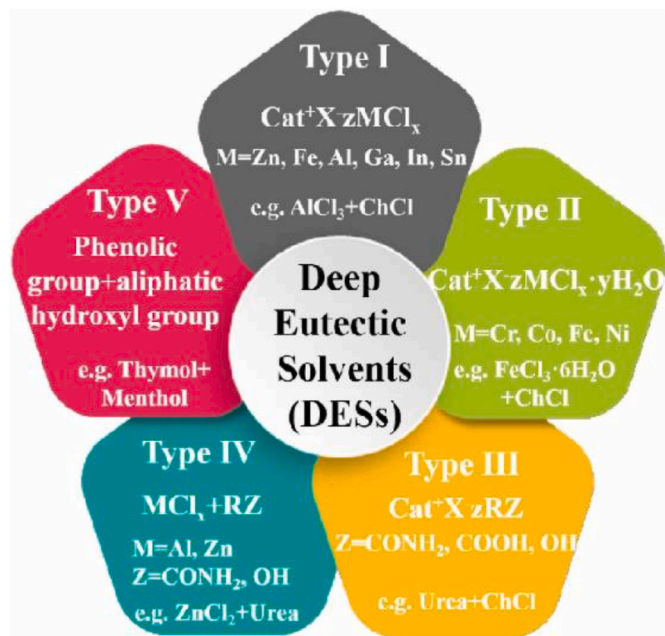
However, the aqueous amine solution suffers from high energy consumption for regeneration [18], corrosiveness [18,19], instability, solvent evaporation loss [20], and degradation of solvent [18,21–24]. In the evaluation of carbon capture, 57% were applied to absorption, 14% to adsorption, 8% to the use of thin films and 21% to mineralization or biofixation [9]. In regard to the current application, the absorption method has a broader scope of applicability. Therefore, it is essential to seek out more efficient absorbents to replace traditional solvents.

Ionic liquids (ILs) have been broadly investigated and utilized as competitive absorbents due to their negligible vapor pressure, non-flammability, thermal stability and modifiable physicochemical properties [25–28], particularly in CO<sub>2</sub> capture. ILs are efficient in capturing CO<sub>2</sub> by adjusting the ionic function of anions and cations [29]. In addition, amine-functionalized ionic liquids are used to capture CO<sub>2</sub> via chemical reaction [30–32], and the molar absorption ratio is normally in the range of 0.5 mol CO<sub>2</sub>/mol ionic liquids [29]. Nevertheless, the molecular weight of the ionic liquids is larger than the molecular weight of the aqueous amine solution. If the unit of absorption is employed to g CO<sub>2</sub>/g ILs, the absorption becomes uncompetitive. Therefore, the high molecular weight of ILs makes it challenging for CO<sub>2</sub> uptake [33]. In addition to this, ionic liquids have some disadvantages including high viscosity, high cost, cumbersome synthesis and purification steps, toxicity, high energy consumption and poor biodegradation [34–41].

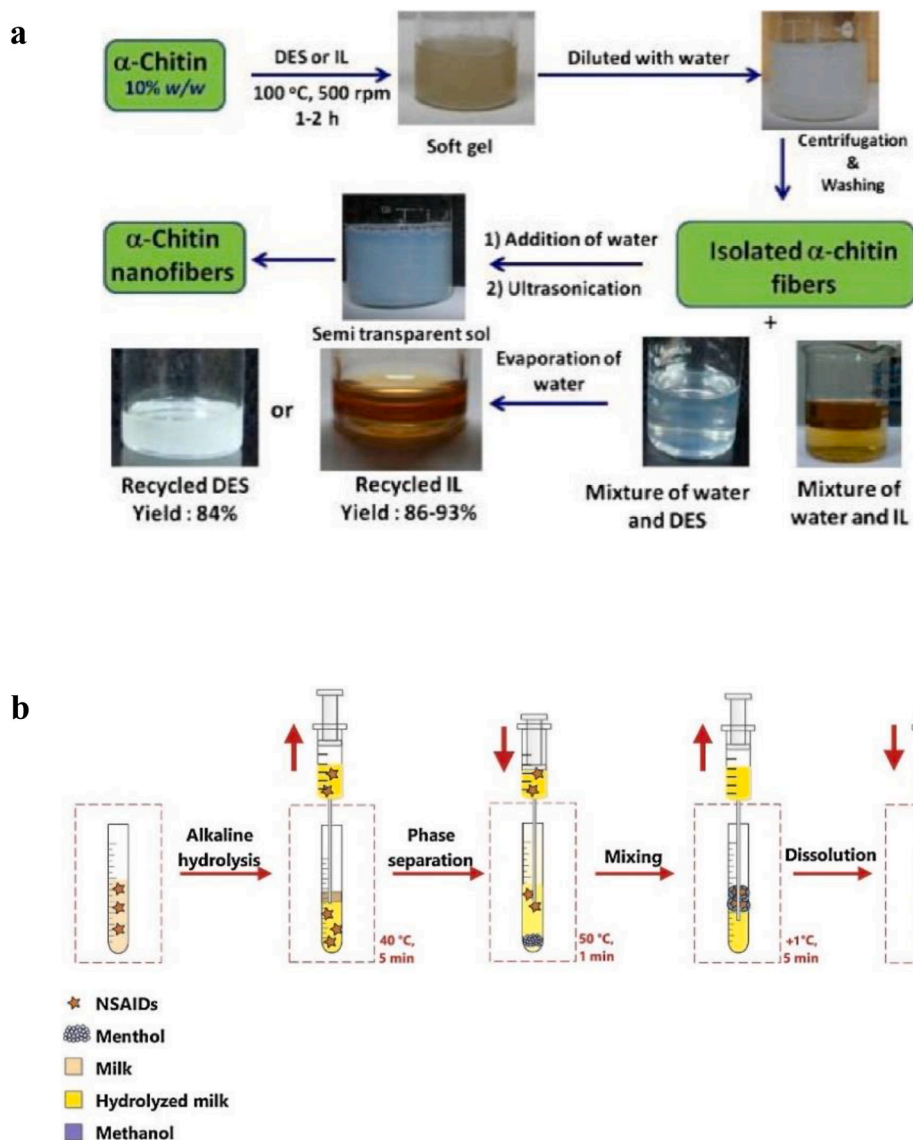
Deep eutectic solvents (DESs), named by Abbott, are a type of solvents similar to ionic liquids, with similar characteristics such as high thermal stability, low-vapor pressure, non-combustibility, and ability to be adjust. In addition, DESs are non-toxic, biodegradable, inexpensive, and are easily of synthesised [43–47]. In general, DESs have lower molecular weights than ILs. DESs are connected by intermolecular hydrogen bonds formed between hydrogen bond donors (HBDs) and

hydrogen bond acceptors (HBAs). Obviously, DESs comprise two or more pure compounds. The melting points of DESs are lower than those of any of the components [35,48]. DESs are divided into groups of five according to different compounds (Fig. 1), which were defined by Abbott and co-workers in 2007 [49].

By 2022, nearly 30 review papers have been produced on DESs. De Jesus et al. [50] mentioned that novel DESs, as environmentally friendly solvents have been applied to lipid extraction in biodiesel production. Mbous et al. [51] investigated the toxicity of DESs and demonstrated that choline chloride can enhance biodegradability. Ge et al. [52] reported that DESs can function as advanced functional materials, including inert reaction media, electrolytes for energy storage devices, and CO<sub>2</sub> solvents. Zdanowicz et al. [53] utilized choline-based DESs for polysaccharide processing. The scheme of the preparation is presented in Fig. 2 (a). Zainal-Abidin et al. [54] studied the potential applications of hydrophobic DESs for COVID-19. An in situ hydrophobic DES system with a simple and environmentally friendly extraction process to remove



**Fig. 1.** General formula for the classification of DESs. R represents an alkyl group; Cat<sup>+</sup> indicates alkali cation. Reproduced with permission [42]. Copyright 2022, Elsevier.



**Fig. 2.** (a) Photographic demonstration for the preparation of  $\alpha$ -chitin nanofibers and recycling of ionic liquid or deep eutectic solvents. Reproduced with permission [55]. Copyright 2014, Elsevier. (b) The schematic diagram of liquid-liquid microextraction based on in situ formation of HDESs from the complexation of menthol with nonsteroidal anti-inflammatory drugs. Reproduced with permission [56]. Copyright 2019, Elsevier.

pharmaceutical contaminants by microextraction techniques was proposed. Fig. 2 (b) describes the process of non-steroidal anti-inflammatory drugs extraction from milk samples. Zhou et al. [42] discussed employing deep eutectic solvents to separate lignin from lignocellulosic biomass.

Several reviews have investigated the properties (e.g., viscosity, density, melting point, ionic conductivity, surface tension, acidity and alkalinity) and application of DESs (e.g., metal deposits, nanomaterials, organic synthesis, gas dissolution, etc.) [43,57–62]. For gas separation, Ul Haq et al. [63] evaluated the application of DESs in oil and gas fields as well as the gas solubility of DESs, with a focus on the solubility of  $\text{CO}_2$  and believed DESs to be effective  $\text{CO}_2$  absorbents. Pelaquim et al. [64] shed light on the differences between the absorption capacities of DESs and ILs, as well as the factors that influence the absorption of DESs. In addition, the solubility was compared to various acid gases. Wang et al. [65] discovered that adding a tertiary component to DESs inhibited high viscosity and enhanced absorption.

The DESs analyzed in these reviews are primarily Type III DESs, especially focusing on choline-based DESs [66]. Literature reviews on other compound types of DESs have been produced [67]. Yan et al. [67]

described deep eutectic solvents for capturing  $\text{SO}_2$  of various types, including inorganic salt-based, azole-based, caprolactam-based, imidazole-based, and polyethylene glycol-based DESs. However, there is a lack of current research to allow comparison of the performance of different types of DESs, and the screening of outstanding gas separation reagents. This especially presents a challenge for carbon capture by absorption.

In this review, the structural characteristics, physical and chemical properties and  $\text{CO}_2$  capture performance of amine-based deep eutectic solvents are investigated and compared with choline-based deep eutectic solvents. The impact of water, HBAs, HBDs, molar ratio and temperature on physicochemical properties are then classified. In addition, the potential amine-based deep eutectic solvents are evaluated and selected.

## 2. Amine-based DESs applied for $\text{CO}_2$ capture

### 2.1. Fundamental

DESs with amine groups as the hydrogen bond donors are named

amine-based DESs. Amine-based DESs belong to Type III and Type IV as shown in Fig. 1. For amine-based DESs, alkanolamines and fatty diamines are included in the hydrogen bond donor (Fig. 3). Hydrogen bond acceptors are broadly classified into three categories: ammonium chloride salts, ammonium bromide salts, hydrochloride salts derived from amides or alcoholic amines and non-ionic organic matter (Fig. 4).

In contrast to amine groups, choline-based DESs are composed of a salt ( $R_1R_2R_3R_4N^+X^-$ ) and a complexing agent (CA) in a particular stoichiometric ratio [34]. CAs consist of both hydrogen bond donors like amine-based DESs and metal salts and oxides. Choline-based DESs belong to Type I, II and III in Fig. 1. It is worth noting that there are DESs that fit the structural features of amine-based DESs and choline-based DESs, such as ChCl-MEA, ChCl-DEA and ChCl-MDEA.

Furthermore, the molecules of amine-based DESs are connected to each other by hydrogen bonding to form a homogeneous liquid phase. Trivedi et al. [29] demonstrated, via Fourier infrared spectroscopy, that [MEA]Cl and EDA form hydrogen bonds.

Choline-based DESs are three of the five categories of DESs: Type I, Type II, and Type III. Choline-based DESs have the following structural pattern: choline + anion + complex = choline group DESs. Organic substances (such as carboxylic acids, amines, and alcohols), metal chlorides, and hydrated metal chlorides are examples of complexes. The structural coverage of choline DESs and amine DESs like ChCl-MEA, ChCl-DEA, and ChCl-MDEA overlap.

## 2.2. Physicochemical properties

The optimal way to develop suitable carbon dioxide solvents is to understand the nature of DESs. The physical and chemical properties of amine-based DESs are described in the following section; factors affecting the performance are investigated, and performance is compared with that of choline-based DESs.

### 2.2.1. Melting points and decomposition temperature

The melting point of amine-based DESs as a mixed solvent is less than that of any of its constituents [35]. For instance, the melting point of choline-based DESs is 578 K, that of monoethanolamine is 283.5 K, and that of ChCl-MEA (1:5) is 276.95 K [68], which is significantly lower than the melting point of its components. A lower melting point can enable a wider temperature range for CO<sub>2</sub> absorption.

Table 1 contains a listing of binary DESs melting points (256.05–293.95 K). A lower melting point of DESs is beneficial to solvent transport and application in the wide temperature range for CO<sub>2</sub> capture. The melting points are influenced by the structures of hydrogen bond acceptor and donor as well as the molar ratio of HBA to HBD.

When the hydrogen bond donor is identical but the hydrogen bond acceptor is different, the melting points of DESs can be tailored. It can be seen in Table 1 that MTPB-MEA (1:6), ChCl-MEA (1:6), and TBAB-MEA (1:6) have melting points of 277.67, 276.84 and 258.19 K, respectively. The hydrogen bond acceptors are ranked in order of the strength of hydrogen bonding with MEA: MTPB-MEA (1:6) < ChCl-MEA (1:6) < TBAB-MEA (1:6). When the hydrogen bond donors are AP, the melting points of TBAB-AP (1:4), TEAC-AP (1:4), and TBAC-AP (1:4) are 277.34,

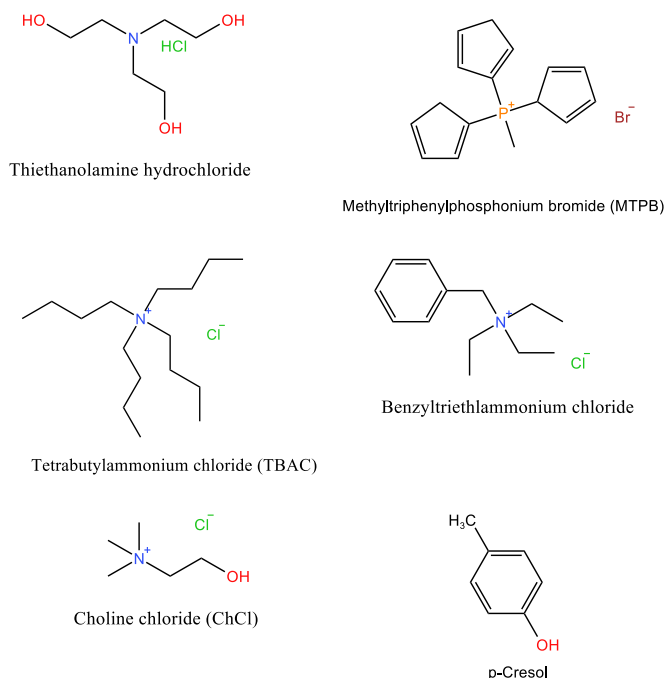


Fig. 4. The hydrogen bond acceptors of amine-based DESs.

272.37, and 276.29 K, respectively, and the order of hydrogen bonding with the hydrogen bond acceptors is as follows: TBAB-AP (1:4) < TBAC-AP (1:4) < TEAC-AP (1:4).

The melting point is also impacted by hydrogen bond donors. MEA has a lower melting point than DEA when ChCl is a hydrogen bond acceptor. The relationship between melting point and molar ratio of HBA to HBD is not purely linear. For example, the melting point of MTPB-MEA, TEAC-AP, and MTPB-MEA may decrease as the molar ratio increases. With an increase in molar ratio, the melting points of [MEA]Cl-EDA, ChCl-MEA, TBAB-MEA, TBAB-AP, and TBAC-AP initially decline and subsequently climb. However, ChCl-DEA has a maximum melting point with a 1: 8 M ratio, the melting point increases first and subsequently falls as the molar ratio increases.

The ternary DESs are as indicated in Table 2; the melting points of ChCl-MEA-DEA (1:7:1), ChCl-MEA-MDEA (1:7:1), ChCl-MEA-AEP (1:7:1), and ChCl-MEA-PZ (1:7:1) are 276.69, 266.32, 268.06, 268.24 and 265.59 K, respectively. It is found that the melting point of ChCl-MEA decreases significantly upon addition of an amine, particularly PZ, which indicates that PZ forms a strong hydrogen bonding interaction with ChCl-MEA.

All in all, it is found that the melting point decreases with a decreasing alkyl chain length. The hydrogen bonding of anions is as follows: Cl<sup>-</sup> > Br<sup>-</sup>. Adding amines to DESs reduces the melting point, especially piperazine. In comparison, the melting points of TBAB-MEA and [MEA]Cl-EDA are determined to be the lowest.

The reported melting point of choline-based DESs is between 235.15 and 432.15 K [34]. In order to reduce the energy loss brought on by melting DESs, which is particularly desirable for industrial applications, DESs should have a melting point of less than 50 °C [59,66]. Amine-based DESs have melting points that are closer to 50 °C than choline-based DESs. Therefore, amine-based DESs are more suited to the carbon capture process, which can slash production costs to some extent.

In conclusion, investigations on the mechanism for decreasing the melting point of ternary DESs, as well as on what model can properly forecast the melting point of DESs are limited, but can be useful for providing technical guidance on the design and synthesis of DESs.

Research on the decomposition temperature ( $T_d$ ) of amine-based DESs is very limited (Tables 1 and 2). The decomposition temperature

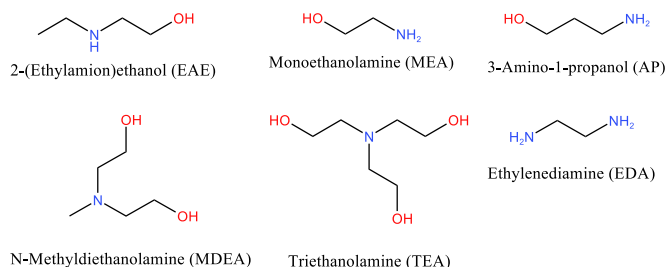


Fig. 3. The hydrogen bond donors of amine-based DESs.



**Table 1**  
Melting points and decomposition temperature of amine-based binary DESs.

Amine-based DESs (mole ratio)	T <sub>m</sub> /HBA (K)	T <sub>m</sub> /HBD (K)	Melting points T <sub>m</sub> (K)	Decomposition temperature T <sub>d</sub> (K)	Refs
[MEA]Cl-EDA (1:2)	355–357	281.5	257.45	–	[29]
[MEA]Cl-EDA (1:3)	355–357	281.5	256.25	436	[29]
[MEA]Cl-EDA (1:4)	355–357	281.5	257.05	–	[29]
ChCl-MEA (1:5)	578	283.5	276.95	410	[69, 74, 75]
ChCl-MEA (1:6)	578	283.5	276.84	–	[74, 76]
ChCl-MEA (1:7)	578	283.5	276.69	413	[69, 74]
ChCl-MEA (1:8)	578	283.5	278.23	–	[74]
ChCl-DEA (1:6)	578	301	291.33	473	[69, 76]
ChCl-DEA (1:8)	578	301	293.98	–	[76]
ChCl-DEA (1:10)	578	301	288.74	–	[76]
MTPB-MEA (1:6)	503–507	283.5	277.67	–	[74]
MTPB-MEA (1:7)	503–507	283.5	272.81	–	[74]
MTPB-MEA (1:8)	503–507	283.5	272.21	–	[74]
TBAB-MEA (1:3)	390	283.5	265.15	–	[74]
TBAB-MEA (1:4)	390	283.5	255.45	–	[74]
TBAB-MEA (1:5)	390	283.5	257.65	–	[74]
TBAB-MEA (1:6)	390	283.5	258.19	–	[74]
TBAB-AP (1:4)	390	283–285	277.34	–	[77]
TBAB-AP (1:6)	390	283–285	274.41	–	[77]
TBAB-AP (1:8)	390	283–285	276.02	–	[77]
TEAC-AP (1:4)	312	283–285	272.37	–	[77]
TEAC-AP (1:6)	312	283–285	272.19	–	[77]
TEAC-AP (1:8)	312	283–285	271.81	–	[77]
TBAC-AP (1:4)	314–317	283–285	276.29	–	[77]
TBAC-AP (1:6)	314–317	283–285	273.19	–	[77]
TBAC-AP (1:8)	314–317	283–285	277.33	–	[77]
MTPB-MDEA (1:7)	503–507	252	172.82	473	[33]
MTPB-MDEA (1:10)	503–507	252	172.61	473	[33]
MTPB-MDEA (1:16)	503–507	252	171.76	473	[33]
ChCl-MDEA (1:7)	578	252	245.8	456	[69]

generally depends on the strength of the hydrogen bonding between the HBAs and HBDs of DESs. The weak intermolecular hydrogen bonding of amine-based DESs results in a lower decomposition temperature.

The type of amine affects the decomposition temperature. Table 2 reveals that the T<sub>d</sub> of the ternary amine-based DESs formed by ChCl-MEA (1:7) is ranked as AEP > DEA > MDEA > PZ. The T<sub>d</sub> of ChCl-

MEA is less than that of ChCl-DEA. Therefore, the T<sub>d</sub> is ranked according to the type of amine: tertiary amine > secondary amine > primary amine.

The T<sub>d</sub> of amine-based DESs is not less than the T<sub>d</sub> of pure amines. For example, the T<sub>d</sub> of pure MEA is 408 K. The T<sub>d</sub> of ChCl-MEA (1:5) is 410 K, which is slightly higher than the T<sub>d</sub> of pure MEA [69]. Overall, the T<sub>d</sub> of amine-based DESs is concentrated in the range of 386–473 K.

The number of publications reporting the decomposition temperature of cholinergic DES is scanty [70–73]. The temperature range of the thermal stability of choline-based DESs can be concluded as 235–474 K. Water alters the decomposition temperature of the choline-based DESs. For instance, according to Dai et al. [70], the T<sub>d</sub> of choline-based DESs in the presence of water is typically lower than 475 K (5.3–11.6 wt%).

Compared to choline-based DESs, studies on the thermal stability of amine-based DESs are relatively limited, especially the T<sub>d</sub>, although the thermal stability temperature range of amine-based DESs is known to be narrower than that of choline-based DESs. The thermal stability of amine-based DESs, in particular the T<sub>d</sub>, needs to be further investigated.

### 2.2.2. Viscosity

Viscosity is a crucial characteristic of DESs, which affects the mass transfer, heat transfer and electrical conductivity of solvents. Surface tension exists at the gas-liquid contact when CO<sub>2</sub> enters DESs through the bubbler. The contact area between CO<sub>2</sub> and DESs decreases with increasing surface tension, making it challenging for CO<sub>2</sub> to transfer. Also, the high viscosity of amine-based DESs present obvious surface tension between the gas phase and liquid phase for CO<sub>2</sub> capture, which distinctly restricts CO<sub>2</sub> diffusion.

At ambient temperature, the majority of DESs are viscous liquids. Hydrogen bonding and other interaction forces can cause high viscosity and small mobility of ions in DESs. Table 4 presents the viscosity range of DESs prior to CO<sub>2</sub> absorption (roughly between 3.6 and 2398 mPa s), which is lower than that of choline-based DESs (from 4 to 75,683 mPa s). As seen in Table 4, the viscosity is mostly at 100 mPa s or less at temperatures close to ambient.

Viscosities of the amine-based DESs are affected by the hydrogen bond acceptors and donors, molar ratio and temperature. When the hydrogen bond acceptor is [MEA]Cl, and does not change, the viscosities of amine-based DESs listed in Table 4 are ranked as follows: [MEA]Cl-TEPA (1:3) > [MEA]Cl-TETA (1:3) > [MEA]Cl-DETA (1:3) > [MEA]Cl-MEA (1:3) > [MEA]Cl-EDA (1:3). In Table 3, it can be concluded that the longer the alkyl chain, the higher the molecular weight and the viscosity. The alkyl chains of MEA and EDA are equal in length, but the hydroxyl group of MEA is heavier than the molecular weight of the amine group of EDA, thus the viscosity of [MEA]Cl-MEA (1:3) is larger than that of [MEA]Cl-EDA (1:3). Similarly, when the hydrogen bond acceptor is ChCl, the viscosities of various hydrogen bond donors are ordered as ChCl-TEA > ChCl-DEA > ChCl-MEA.

For the same hydrogen bond donor of amine-based DESs, when it is EDA, the viscosities of amine-based DESs at 30 °C and 101 kPa are ranked as follows: [TEA]Cl-EDA (1:3) > [TAE]Cl-EDA (1:3) > [UE]Cl-EDA (1:3) > [MEA]Cl-EDA (1:3).

The molar ratio of HBA/HBD influences the viscosities of amine-based DESs. Usually, with the increase in molar ratio, viscosity decreases. In particular, the viscosity of [MEA]Cl-DETA is the highest in this work as the molar ratio is 1: 3.

In general, high temperature can break the hydrogen bond between the hydrogen bond acceptors and the donors, resulting in a decrease in viscosity. Abbott et al. used the hole theory to explain that at higher temperatures, the pore size is comparable to the ion size and the ions tend to migrate into the cavities, leading to a reduction in the viscosity of the ions [57,78–80].

In the literature so far, the viscosities of amine-based DESs before CO<sub>2</sub> absorption have been more widely reported [33,74,77,81–84]. However, the most important viscosity after CO<sub>2</sub> absorption is rarely reported [29,85] as can be seen in Table 4. As shown in Fig. 5 and

**Table 2**

Melting points and decomposition temperature of amine-based ternary DESs.

Amine-based DESs	T <sub>m</sub> /HBA (K)	T <sub>m</sub> /MEA (K)	T <sub>m</sub> /Functionalized amine (K)	Melting points T <sub>m</sub> /DESs (K)	Decomposition temperature T <sub>d</sub> (K)	Refs
ChCl-MEA-DEA (1:7:1)	578	283.5	301	266.32	386	[75]
ChCl-MEA-MDEA (1:7:1)	578	283.5	252	268.06	401	[75]
ChCl-MEA-MDEA (1:7:5)	578	283.5	252	267.69	401	[75]
ChCl-MEA-AEP (1:7:1)	578	283.5	254	268.24	403	[75]
ChCl-MEA-PZ (1:7:1)	589	283.5	382–385	265.59	388	[75]

**Table 3**

The structure and relative molecular mass of [MEA]Cl-based DESs.

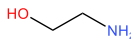
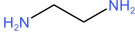
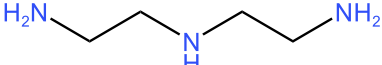
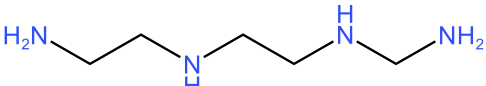
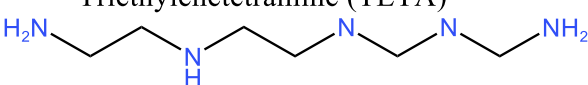
Structure	Relative molecular mass
 Monoethanolamine (MEA)	61.08
 Ethylenediamine (EDA)	60.10
 Diethylenetriamine (DETA)	103.17
 Triethylenetetramine (TETA)	132.21
 Tetraethylenepentamine (TEPA)	161.25

Table 4, the viscosities of [MEA]Cl-EDA (1:3) increase dramatically to thousands of mPa·s after CO<sub>2</sub> absorption, especially at lower temperatures. A high viscosity of DESs after CO<sub>2</sub> absorption may need greater pump power to transport solvents. Moreover, some empirical methods exist to forecast viscosity before absorption. The Vogel-Tamman-Fulcher equation, Arrhenius equation and Redlich-Kister-type equation were used to predict the viscosities of DESs [77,86–88]. Nowosielski et al. [77] indicated that the Vogel-Tamman-Fulcher equation describes the dependence of viscosity on temperature much better than the Arrhenius equation does. Bakhtyari et al. proposed a new model for predicting viscosity by inputting critical pressure, critical temperature and reference viscosity data or by obtaining VFT constants for all DESs, suitable for any DESs [89]. Benguerba et al. presented a novel mathematical model for predicting the viscosities of amine-based DESs using the quantitative structure-property relationship (QSPR) method [90].

This section focuses mostly on the elements that affect viscosity. The lower the viscosity, whether acting as a hydrogen bond donor or acceptor, the shorter the alkyl chain. In general, viscosity before CO<sub>2</sub> absorption may be effectively reduced by raising the temperature and molar ratio. However, there is a dearth of theoretical and experimental studies on viscosity after CO<sub>2</sub> absorption. In order to screen green and low viscosity DESs suited for CO<sub>2</sub> separation, efforts should be made to investigate how to reduce post-absorption viscosity, and a corresponding model should be developed to predict it. The viscosity range of choline-based DESs is approximately 10–75,683 mPa·s [78,79,86, 91–94]. Most solvents have viscosities in the hundreds of mPa·s at near room temperature. The viscosity of amine-based DESs is in the range of

3.6–2398 mPa·s (Table 4). Viscosities are mostly below 100 mPa·s at room temperature. One can see that the viscosity span of choline-based DESs is much wider. Overall, the viscosity of choline-based DESs is higher than that of amine-based DESs. The high viscosities of choline-based DESs compared to amine-based DESs lead to limitation in the speed of mass and heat transfer, solvent conductivity and mobility of dissolved substances through it, impacting the complexity of the application process.

### 2.2.3. Activation energy

During the flow process, viscosity activation energy is the minimum amount of energy needed to overcome the potential barrier of the flow unit and move from its initial position to a nearby “cavity”. Viscosity activation energy characterizes how sensitive viscosity is to changes in temperature and carries important implications for heat transfer rate and heat transfer uniformity. Viscosity is already so low that the effect of temperature change on viscosity is not change significant. To understand the effect of viscosity on CO<sub>2</sub> absorption, the activation energy of viscous flow was calculated using Arrhenius-type equations as shown in Eq. (1) [95].

$$\ln \eta = \ln \eta_{\infty} - \frac{E(\alpha, \eta)}{RT} \quad (1)$$

where  $\eta$  is the viscosity, mPa·s.  $\eta_{\infty}$  is the viscosity at infinite temperature, mPa·s.  $E(\alpha, \eta)$  is the activation energy for viscous flow, J/mol.  $R$  is the universal gas constant, 8.314 J/mol·K.  $T$  is the temperature, K.

Activation energy explains the limiting effect of DESs on CO<sub>2</sub>

**Table 4**Densities and viscosities of pure amine-based DESs and viscosities of amine-based DESs after CO<sub>2</sub> uptakes.

Amine-based DESs (mole ratio)	T(K)	P (kPa)	Densities of pure amine-based DESs( $\rho/\text{g cm}^{-3}$ )	Viscosities of pure amine-based DESs (mPa·s)	Viscosities of amine-based DESs after CO <sub>2</sub> uptake (mPa·s)	Refs
ChCl-MEA (1:5)	298–358	101	1.08–1.04	48.73–5.7	–	[74]
ChCl-MEA (1:6)	298–358	101	1.07–1.03	36.63–5.3	–	[74, 82]
ChCl-MEA (1:7)	298–358	101	1.06–1.02	37.46–5.3	–	[74]
ChCl-MEA (1:8)	298–358	101	1.06–1.02	22.6	–	[74, 82]
ChCl-DEA (1:6)	298	101	1.094	75.1	–	[82]
ChCl-DEA (1:8)	298	101	1.092	70.23	–	[82]
ChCl-TEA (1:6)	298	101	1.125	83.5	–	[82]
ChCl-TEA (1:8)	298	101	1.114	80.46	–	[82]
MTPB-MEA (1:5)	298–358	101	1.20–1.15	129.82–6.95	–	[74]
MTPB-MEA (1:6)	298–358	101	1.11–1.06	33.73–2.54	–	[74]
MTPB-MEA (1:7)	298–358	101	1.10–1.05	26.24–2.54	–	[74]
MTPB-MEA (1:8)	298–358	101	1.09–1.04	26.24–2.54	–	[74]
TBAB-MEA (1:3)	298–358	101	1.06–1.02	106.92–8.45	–	[74]
TBAB-MEA (1:4)	298–358	101	1.06–1.02	79.62–6.15	–	[74]
TBAB-MEA (1:5)	298–358	101	1.06–1.01	51.54–3.08	–	[74]
TBAB-MEA (1:6)	298–358	101	1.05–0.99	20.38–3.08	–	[74]
TBAB-AP (1:4)	273–333	100	1.022–0.993	113.95–16.99	–	[77]
TBAB-AP (1:6)	273–333	100	1.016–0.986	79.29–12.83	–	[77]
TBAB-AP (1:8)	273–333	100	1.011–0.981	65.55–10.91	–	[77]
TEAC-AP (1:4)	273–333	100	0.999–0.971	55.20–10.28	–	[77]
TEAC-AP (1:6)	273–333	100	0.996–0.967	46.64–8.82	–	[77]
TEAC-AP (1:8)	273–333	100	0.995–0.965	43.43–8.30	–	[77]
TBAC-AP (1:4)	273–333	100	0.961–0.935	101.38–16.08	–	[77]
TBAC-AP (1:6)	273–333	100	0.967–0.939	70.66–11.99	–	[77]
TBAC-AP (1:8)	273–333	100	0.97–0.942	60.21–10.73	–	[77]
[MEA]Cl-MEA (1:1)	313	101	1.1503	112	–	[81]
[MEA]Cl-MEA (1:3)	313	101	1.0898	48	–	[81]
[MEA]Cl-MEA (1:6)	313	101	1.0559	29	–	[81]
[MEA]Cl-MEA (1:9)	313	101	1.0424	21	–	[81]
[MEA]Cl-DETA (1:1)	313	101	1.0990	39	–	[81]
[MEA]Cl-DETA (1:3)	313	101	1.0272	52	–	[81]
[MEA]Cl-DETA (1:6)	313	101	0.9863	14	–	[81]
[MEA]Cl-DETA (1:9)	313	101	0.9751	9	–	[81]
[MEA]Cl-EDA (1:1)	313	101	1.0302	113	–	[81]
[MEA]Cl-EDA (1:3)	313	101	1.0218	42	–	[81]
[MEA]Cl-EDA (1:6)	313	101	0.9696	9	–	[81]
[MEA]Cl-EDA (1:9)	313	101	0.9435	4	–	[81]
[MEA]Cl-TETA (1:1)	313	101	1.0974	728	–	[81]
[MEA]Cl-TETA (1:3)	313	101	1.0639	134	–	[81]
[MEA]Cl-TETA (1:6)	313	101	1.0315	52	–	[81]
[MEA]Cl-TETA (1:9)	313	101	0.9981	36	–	[81]
[MEA]Cl-TEPA (1:1)	313	101	1.1151	2398	–	[81]
[MEA]Cl-TEPA (1:3)	313	101	1.0281	257	–	[81]
[MEA]Cl-TEPA (1:6)	313	101	1.0081	103	–	[81]
[MEA]Cl-TEPA (1:9)	313	101	0.9971	70	–	[81]
MTPB-MDEA (1:7)	293–353	101	–	238–11.936	–	[33]
MTPB-MDEA (1:10)	293–353	101	–	192.25–10.639	–	[33]
MTPB-MDEA (1:16)	293–353	101	–	152.67–9.2842	–	[33]
[TEA]Cl-MEA (1:4)	298	101	–	37.6	–	[83]
TBAB-EAE (1:4)	298	101	–	34.3	–	[83]
[BTEA]Cl-EAE (1:4)	298	101	–	39	–	[83]
DETA-AA-EG (1:1:2)	298–313	101	–	85.714–40.06	–	[84]
[MEA]Cl-EDA (1:3)	303–363	101	–	21.6–7.5	3995.0–56.2	[29]
[TEA]Cl-EDA (1:3)	303–363	101	–	52.4–9.7	3343.0–45.1	[29]
[UE]Cl-EDA (1:3)	303–363	101	–	29.5–4.6	3011.0–70.4	[29]
[TAE]Cl-EDA (1:3)	303–363	101	–	30.7–3.6	748.8–41.9	[29]
[TETA]Cl-EG (1:4)	313	101	–	110.4	1326.9	[85]

diffusion. The less the activation energy, the easier CO<sub>2</sub> diffusion. It is influenced by hydrogen bond donors and the molar ratio of HBA/HBD. EDA-based DESs show lower activation energy than that of AP-based DESs, and it decreases as the molar ratio increases (Fig. 6). The potential mechanism still needs to be explored for better synthesizing DESs to capture CO<sub>2</sub>.

#### 2.2.4. Density

Density is the most significant property in the design requirements of the CO<sub>2</sub> capture process. According to Table 4, the densities of amine-based DESs fall roughly within the range of 0.95–1.2 g cm<sup>-3</sup>. Most

amine-based DESs are hydrophilic and denser than water. Firstly, the density of DESs is affected by hydrogen bond acceptors and donors. Under the hydrogen bond acceptor ChCl, the densities of amine-based DESs are ranked as follows: ChCl-TEA (1:6) > ChCl-DEA (1:6) > ChCl-MEA (1:6). When the hydrogen bond donor is AP, the amine-based DESs densities are arranged as follows: TBAB-AP (1:4) > TEAC-AP (1:4) > TBAC-AP (1:4). Density is also affected by molar ratio. The density of DESs decreases as the molar ratio increases, however, the change is not significant.

As the temperature rises, the density decreases due to the motion of ions and the expansion of free volume. In accordance with the hole

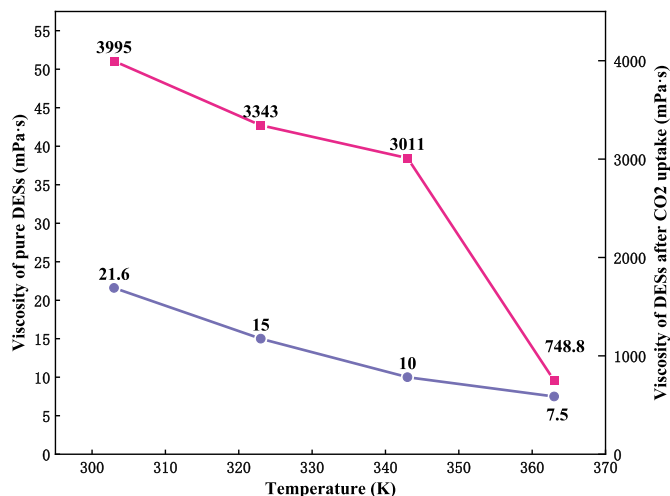


Fig. 5. Viscosities of [MEA]Cl-EDA (1:3) before and after CO<sub>2</sub> absorption [29].

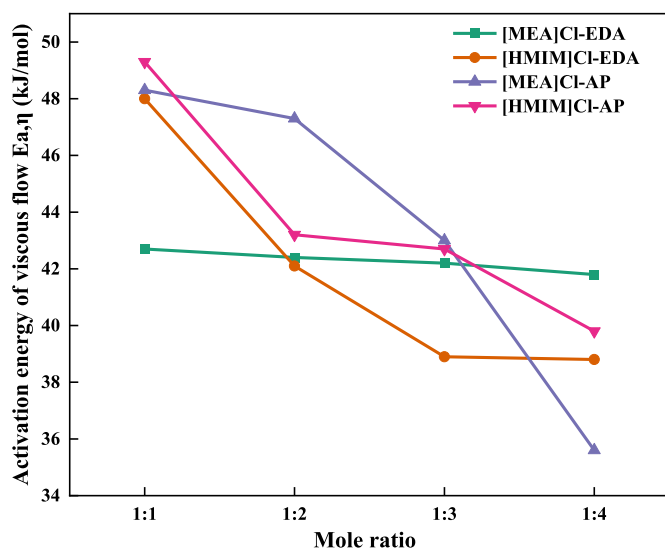


Fig. 6. Relationship between molar ratio and activation energy [96].

theory, the temperature rises and the liquid is locally subjected to thermal fluctuations to produce holes, resulting in liquid space growing larger [57,91].

With an amine-based chloride salt as the hydrogen bond acceptor, the densities range from 0.96 to 1.15 g cm<sup>-3</sup> at 313 K and 101 kPa, which is predominantly close to 1 g cm<sup>-3</sup>. The densities of the choline-based DESs vary between 1.0197 and 1.2420 g cm<sup>-3</sup> at 298 K, 101 kPa [34, 78,79,97–108], which are comparable to the densities of the amine-based DESs. Density can be successfully predicted using empirical and semi-empirical models like the unitary linear equation [104,105, 109], the modified Rackett equation, the neural network NN, the Group Contribution Method GCM [106,110], the tait-type and Redlich-Kister type equation [111].

Density is most obviously influenced by temperature. The corresponding data has been recorded in the literature, especially for fresh solvents, before absorption. However, density after CO<sub>2</sub> absorption has still not been determined, and this is also important for the designing of equipment and the controlling process. Hence, future research should test and forecast density after absorption in order to improve the CO<sub>2</sub> capture process.

## 2.2.5. Thermal conductivity

Understanding the thermal conductivity of solvents is essential in any heat transfer or chemical reaction involving DESs that requires the use of solvents. Moreover, fluid heat transfer data is crucial for the design and operation of CO<sub>2</sub> capture heat exchangers in order to minimize energy consumption and investment costs.

Little research has been conducted on the thermal conductivity of DESs to date [112–116]. The thermal conductivity of choline-based DESs has been shown in most published works (Fig. 7). In contrast, there is basically no study on the thermal conductivity of amine-based DESs. In fact, the thermal conductivities of DESs are affected by water content, temperature, salt content, and HBD, according to the research literature. As the temperature rises, two opposite trends occur in thermal conductivity. The thermal conductivities of ChCl-Urea, N,N-diethyl ethanol ammonium chloride-Urea and ChCl-Thiourea exhibit a decreasing trend with temperature [112]. While those of ChCl-PEG-Glycerol and ChCl-Glycerol increase with temperature [116]. An increase in chloride salt content in DESs causes a decrease in thermal conductivity [117]. This may be due to the fact that electronegativity is positively correlated with the strength of hydrogen bonding, which in turn causes an enhancement and weakening of thermal conductivity.

Celebi et al. [114] investigated the thermal conductivity of aqueous solutions of DESs by the Müller-Plathe (MP) method with the aid of molecular dynamics simulations. Results demonstrated that water enhances the thermal conductivity of DESs, whereas the thermal conductivity of aqueous solutions is independent of temperature.

Water and temperature may visibly alter thermal conductivity while molar ratio has no discernible impact on it. Also, thermal conductivity and viscosity are closely related, with thermal conductivity and viscosity being directly proportional. The relationship between viscosity and thermal conductivity is shown in Eq. (2) [118].

$$\ln\left(\frac{\lambda}{\mu}\right) = A + B \frac{T_b}{T} \left( \frac{T_c - T}{T_c - T_b} \right) \quad (2)$$

where  $\lambda$  is thermal conductivity, W/(m.K).  $\mu$  is viscosity, mPa.s.  $T_b$  is boiling point, K.  $T_c$  is critical temperature, K.  $T$  is operating temperature, K.  $A$  and  $B$  are model parameters. Thermal conductivity and viscosity are proportional to capital cost and energy consumption for CO<sub>2</sub> capture.

An addition of the appropriate amount of water can both reduce viscosity and enhance heat transfer. It is intended that further consideration be given to the experimental and simulation studies on amine-based DESs.

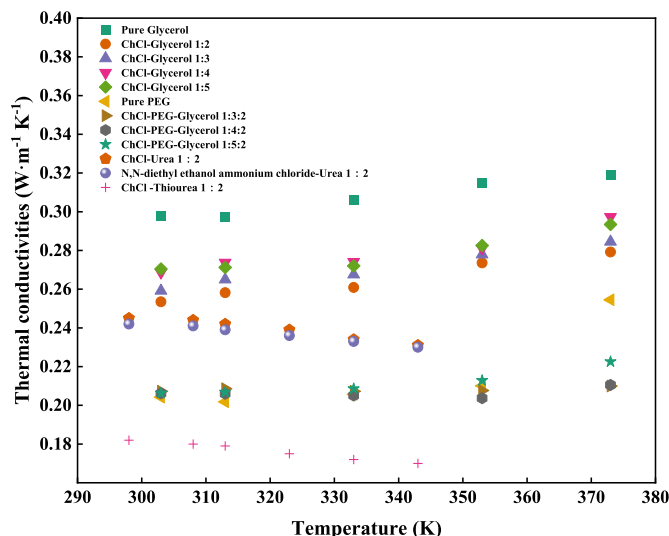


Fig. 7. Thermal conductivity of choline-based DESs at 1 bar [112,116].



### 2.2.6. Surface tension

Surface tension arises at the gas-liquid interface and CO<sub>2</sub> bubbles emerge when CO<sub>2</sub> gas is injected into the amine-based DESs. Reducing the surface tension can be used to increase the surface area available for the CO<sub>2</sub> mass transfer. The surface tension of amine-based DESs ranges from 33.21 to 45.02 mN m<sup>-1</sup> at 298–358 K and 101 kPa, which is generally smaller than those of choline-based DESs (from 41.9 to 75.0 mN m<sup>-1</sup>) [78,79,98,103,119–121].

As illustrated in Fig. 8, the surface tension is affected by the hydrogen bond acceptor and donor, molar ratio and temperature. With MEA as the hydrogen bond donor, the surface tension of amine-based DESs follows: MTPB-MEA (1:5) > ChCl-MEA (1:5) > TBAB-MEA (1:5). Surface tension of TBAB-MEA (1:5) is the minimum due to the weakened hydrogen bond net. Compared with pure MDEA, the surface tension of MTPB-MDEA is stronger. It decreases as the molar ratio increases. Nonetheless, by increasing the molar ratio of the remaining amine-based DESs, the surface molecular cohesion strengthens and the surface tension improves. Owing to the weakening of intermolecular interactions and the increase in kinetic energy, an increase in temperature results in a decrease in surface tension.

In contrast with amine-based DESs, the surface tension of choline-based DESs increases with temperature, except for the ChCl-LaA (1:2) mixture. The surface tension of DESs was described by the hole theory, QSPR and Othme's equation [79,80,98]. Reza Haghighbakhsh [122] initially proposed a generalized model for DESs to estimate surface tension at varying temperatures in 2020. It is mentioned that surface tension increases with an increase in molar ratio and falls linearly with temperature, and TBAB-MEA shows the lowest surface tension. Experiments and simulations of amine-based DESs on surface tension are in short supply, and additional research is required.

### 2.2.7. Heat of reaction

The regeneration energy consumption for CO<sub>2</sub> capture is shown in Eq. (3), where  $Q_{\text{reg}}$  is the heat of reaction and represents heat changing during the CO<sub>2</sub> capture process.

$$Q_{\text{reg}} = Q_{\text{reac}} + Q_{\text{sens}} + Q_{\text{evap}} \quad (3)$$

$$Q_{\text{reac}} = -\Delta H \quad (4)$$

The heat of reaction indicates whether energy consumption and regeneration are feasible. The Van't Hoff equation [29] can be used to calculate the heat of absorption ( $\Delta H$ ) as follows:

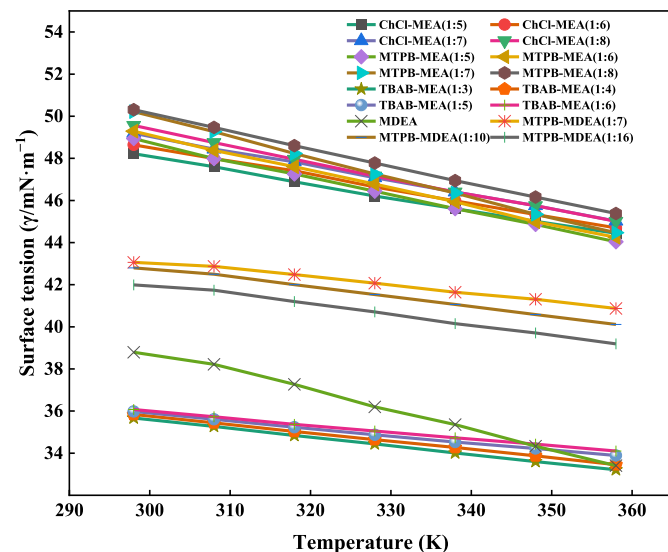


Fig. 8. Temperature dependence of surface tension of amine-based DESs at 1 bar [33,74].

$$\ln K = -\frac{\Delta H}{R} \times \frac{1}{T} + \frac{\Delta S}{R} \quad (5)$$

where  $\Delta H$  is enthalpy of reaction, kJ/kg.  $\Delta S$  is entropy of reaction, kJ/kg.  $R$  is constant of gas.  $T$  is thermodynamic temperature, K.  $K$  is reaction equilibrium constant, 8.314 J/mol·K.

The heat of reaction for the amine-based DESs in Fig. 9 varies from −54.88 to 7.3 kJ/mol. Heat absorption is indicated by a positive  $\Delta H$ , while a negative  $\Delta H$  denotes heat emission. In general, the absorption of CO<sub>2</sub> using DESs is an exothermic reaction, as shown by Shukla et al. [96] and Pishro et al. [123]. The heat of reaction of [MEA]Cl-AP, [HMIM]Cl-AP and [MEA]Cl-TEPA is exothermic. There are a very few cases where the absorption of CO<sub>2</sub> by DESs is a heat absorbing reaction, such as one recorded by Shukla et al. [124], who indicated that the heat of reaction of [HMIM]Cl-EDA is a heat absorbing reaction. Normally, exothermic reactions are easier to perform in the DESs carbon capture process. In addition, the heat released in the absorption process raises the temperature of the reaction system and promotes a reaction. The absolute value form is typically used to evaluate energy usage. A reduced energy need for regeneration is indicated by a smaller absolute value of the reaction heat. Depending on the hydrogen bond acceptors and donors, the heat of reaction falls when the HBA/HBD molar ratio rises. Based on the Gibbs free energy formula,  $\Delta G < 0$ , the reaction is favored in the forward direction. The sequence of the reaction is indicated by the entropy change. When the change in entropy exceeds zero, the degree of disorder increases, which favors the reaction. The entropy change of the EDA-based DESs is larger than 0, which favors the reaction between the solvent and CO<sub>2</sub>. Developing a novel way to use lower energy for breaking the chemical bond between DESs and CO<sub>2</sub> is an ongoing work.

### 2.3. CO<sub>2</sub> absorption and regeneration

#### 2.3.1. Method for the measurement of CO<sub>2</sub> solubility

There are broadly two methods for measuring the absorption of DESs, the pressure drop method and the weighing method.

The first determines the solubility of CO<sub>2</sub> by a pressure drop method via a gas-liquid equilibrium apparatus. The detailed operational steps have been reported in previous literature [125,126]. The pressure drop method works on the principle that the pressure in the system decreases before and after the absorbent absorbs the gas, and the pressure difference is used to calculate the solubility of the gas in the absorbent.

The second one is a weighing method to evaluate the CO<sub>2</sub> solubility.

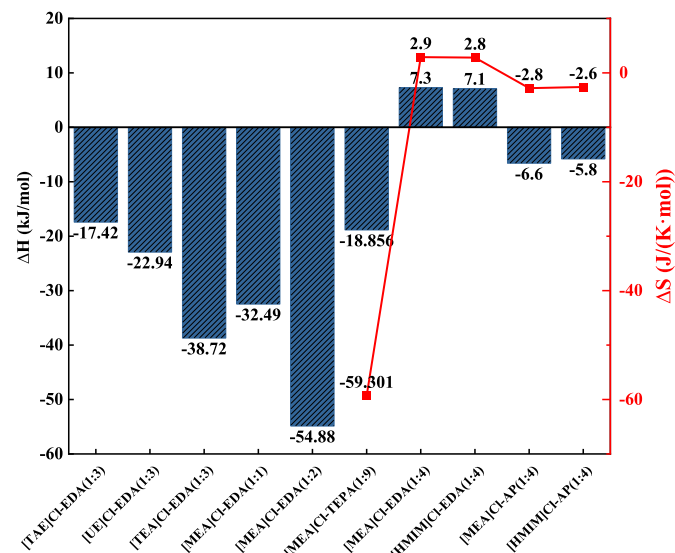


Fig. 9. Enthalpy and entropy of reaction for amine-based DESs [29,96,123].

The method of operation has been previously described in literature [127]. The mass of the absorbent to absorb gas is calculated from the change in weight of the absorbent before and after absorption of the gas.

Table 4 shows that the pressure drop method is appropriate for absorption under high-pressure conditions, whereas the weighing method is more applicable to ambient pressure. Haider et al. [83] measured the CO<sub>2</sub> solubility of TBAB-MEA (1:4), [BTEA]Cl-MEA (1:4), TBAB-EAE (1:4), [BTEA]Cl-EAE (1:4) at 1000 kPa pressure using the pressure drop method. Pishro et al. [81] determined the solubility of [MEA]Cl-MEA, [MEA]Cl-DETA, [MEA]Cl-EDA, [MEA]Cl-TETA and [MEA]Cl-TEPA to CO<sub>2</sub> at 800 kPa following the same measurement method.

### 2.3.2. CO<sub>2</sub> solubility

Carbon dioxide solubility is one of the important properties for CO<sub>2</sub> separation. It is currently the main method used to judge the carbon capture capacity of deep eutectic solvents. Table 5 summarizes the solubility of carbon dioxide in amine-based DESs, showing that hydrogen bond donors have an effect on it. Adeyemi et al. [128] tested the CO<sub>2</sub> uptake of ChCl-MEA, ChCl-DEA and ChCl-MDEA at same molar ratios at 313 K, 100 kPa. By comparison, it was discovered that the absorption was ranked as ChCl-MEA > ChCl-DEA > ChCl-MDEA at the same molar ratio. ChCl-DEA possesses a higher viscosity than ChCl-MEA. The extensive intermolecular hydrogen bonding network of ChCl-DEA makes it difficult to form a hydrogen bonding network with carbon dioxide, which inhibits free matter migration. Hence the solubility of CO<sub>2</sub> is not as high as ChCl-MEA. Pishro et al. [81] selected [MEA]Cl as the hydrogen bond acceptor and the hydrogen bond donor as an amine-based deep eutectic solvent formed by MEA, DETA, EDA, TETA and TEPA. At 313 K, 800 kPa, the same molar ratio of CO<sub>2</sub> uptake was ranked as follows: [MEA]Cl-TETA > [MEA]Cl-DETA > [MEA]Cl-TEPA > [MEA]Cl-EDA > [MEA]Cl-MEA, displayed in Fig. 10. It illustrates that fatty diamines react more readily with CO<sub>2</sub> than alkanolamines with the same carbon skeleton. Also, the lengthier the chain length, the more amine sites, and thus the higher the solubility of CO<sub>2</sub>.

CO<sub>2</sub> solubility is influenced by hydrogen bond acceptors as shown in Fig. 11. Li et al. [15] studied the CO<sub>2</sub> solubility for MEA-based DESs prepared with five double-chain quaternary ammonium salts, TMAC, TEAC, TEAB, TBAC and TBAB, as hydrogen bond acceptors at 303 K, 101 kPa, in a molar ratio of 1:5, with the following CO<sub>2</sub> solubility magnitudes: TMAC-MEA > TEAC-MEA > TEAB-MEA > TBAC-MEA > TBAB-MEA. CO<sub>2</sub> is less soluble in quaternary ammonium salts with lengthier alkyl chains. This is probably because the longer the alkyl chain, the larger the molecular weight, the higher the viscosity of DESs and the smaller the free volume. The effect of anions on the solubility of quaternary ammonium salts is Cl<sup>-</sup> > B<sup>-</sup>/Br<sup>-</sup>.

The molar ratio also has an effect on solubility. In general, the CO<sub>2</sub> solubility is improved with the molar ratio of amine-based DESs increasing because of an increase in amine groups. However, the CO<sub>2</sub> solubility of [MEA]Cl-EDA decreases when the molar ratio increases from 1:3 to 1:4, which suggests that other factors need to be considered. Raising the absorption temperature would appropriately increase the CO<sub>2</sub> solubility of amine-based DESs, due to a decrease in viscosity of DESs at high temperature, but too high a temperature would cause CO<sub>2</sub> desorption from DESs in turn decreasing CO<sub>2</sub> solubility.

Fig. 12 indicated that water impacts on CO<sub>2</sub> solubility. The addition of the proper amount of water would facilitate CO<sub>2</sub> dissolution because water would weaken the hydrogen bonding between the molecules of DESs, thereby facilitating the interaction with CO<sub>2</sub>. However, too much water (generally more than 20%) would cause the amine concentration to drop, resulting in a decrease in CO<sub>2</sub> solubility. The addition of other materials to amine-based DESs for functionalization can also promote CO<sub>2</sub> solubility. Li et al. [15] employed TEA, MDEA and LiCl to TEAC-MEA to enhance CO<sub>2</sub> solubility and found that the addition of MDEA and LiCl significantly increases CO<sub>2</sub> solubility.

The 29 choline-based DESs investigated in the literature show CO<sub>2</sub> solubilities of 0.002–0.399 mol CO<sub>2</sub>/mol DESs at 298 K, 101 kPa [34].

CO<sub>2</sub> separation by most choline-based deep eutectic solvents is physical absorption [17,101,103,129–131]. However, CO<sub>2</sub> capture by amine-based deep eutectic solvents is a combination of chemical and physical absorption (see Table 6). Physical absorption is strongly dependent on pressure [17]. Adeyemi et al. [128] discovered that the CO<sub>2</sub> capacity of ChCl-Glycerol is significantly lower than that of ChCl-MEA (1:8) at 313 K and 100 kPa, indicating that chemical absorption is more powerful than physical absorption.

Therefore, compared to the CO<sub>2</sub> solubility of amine-based DESs, CO<sub>2</sub> is less soluble in ChCl-Glycerol DESs. In addition, physical absorption is highly dependent on temperature and pressure. Furthermore, chemical absorption surpasses physical absorption. Hence, the CO<sub>2</sub> solubility of amine-based DESs is greater than that of choline-based DESs. Choline-based DESs are more suitable for gas streams with high CO<sub>2</sub> concentration, such as biogas (20–40% CO<sub>2</sub>), biosynthesis gas (28–30% CO<sub>2</sub>), lime kiln gas (30–40% CO<sub>2</sub>). A low concentration of high-temperature flue gas (10–20% CO<sub>2</sub>) makes amine-based DESs more suitable for use.

### 2.3.3. Regeneration and stability

Regenerative properties of the absorbent are essential for practical applications when it is applied to CO<sub>2</sub> capture [85]. An unrecyclable absorbent would increase the operating cost, as would a bad or difficult regeneration process; also, regenerating would lead to an increase in the cost, and this is not conducive to industrial applications. Therefore, regeneration performance is a key criterion for evaluating an absorbent. By measuring the recycled absorption-regeneration of the absorbent, a judgment can be made on the recyclability of the absorbent.

From Tables 5 and it was found that the regeneration performance of amine-based DESs was subject to the influence of hydrogen bond acceptors and hydrogen bond donors. ChCl-MEA had the worst regeneration efficiency, which might be attributed to evaporation, thermal and oxidative degradation of amine during the desorption process. The regeneration efficiency of DESs containing EG/DG was generally higher.

Cycling times also impact the regeneration efficiency of DESs. Xu [127] performed 5 cycles of absorption-desorption experiments on p-cresol-MEA (1:1) and noticed that regeneration efficiency decreased from 96.6% to 92.4% with more cycle factors. In [TETA]Cl-DG (1:2) and [TETA]Cl-EG (1:3) the regeneration efficiency decreased from 100% to 97.5% after six absorption-desorption cycles [85]. The influences of molar ratio, absorption temperature and desorption temperature on regeneration efficiency have not been reported yet, so further studies are needed. Also, no literature has been found on the regeneration efficiency of choline-based DESs.

The regeneration efficiency can further reflect to some extent the stability of amine-based DESs. From the investigated data on the regeneration efficiency of amine-based DESs, the cycled CO<sub>2</sub> capacity is not significantly decayed after five absorption-desorption cycles. However, long-term absorption-desorption experiments are necessary to evaluate the stability of DESs.

In effect, evaporation, thermal and oxidative degradation of amine-based DESs may endanger regeneration performance and stability [134]. This may result in the regenerated solvent not regaining its original solvency capacity, thus affecting regeneration effectiveness and service life. Also, structural alterations of solvent molecules, such as bond breaking and cyclization [135,136], can result from thermal and oxidative degradation reactions. These structural alterations may result in changes in solvent characteristics, such as solubility, polarity, surface tension, etc., which may affect the stability of the solvent.

## 3. Discussion

The reported works on carbon capture by deep eutectic solvents have focused on CO<sub>2</sub> solubility and have been used to evaluate the CO<sub>2</sub> capture performance of deep eutectic solvents. However, besides the CO<sub>2</sub> solubility, the viscosity of the deep eutectic solvents before and after CO<sub>2</sub> loading and the stability of the deep eutectic solvents,

**Table 5**  
CO<sub>2</sub> solubility of amine-based DESs.

Solvents (mole ratio)	Absorption measurement methods	Absorption temperature(K)	P <sub>CO2</sub> (kPa)	Absorption (gCO <sub>2</sub> /g solvent)	Desorption temperature (K)	Regeneration efficiency (cycle times)	Refs
amine-based binary DESs							
ChCl-MEA (1:5)	Weighing method	303	15	0.2523	413	72% (5 times)	[15]
ChCl-MEA (1:6)	–	313	15	0.293	–	–	[128]
ChCl-MEA (1:8)	–	313	15	0.315	–	–	[128]
ChCl-MEA (1:10)	–	313	15	0.322	–	–	[128]
ChCl-DEA (1:6)	–	313	15	0.146	–	–	[128]
ChCl-DEA (1:8)	–	313	15	0.151	–	–	[128]
ChCl-DEA (1:10)	–	313	15	0.157	–	–	[128]
ChCl-MDEA (1:6)	–	313	15	0.013	–	–	[128]
ChCl-MDEA (1:8)	–	313	15	0.015	–	–	[128]
ChCl-MDEA (1:10)	–	313	15	0.017	–	–	[128]
p-Cresol/DEA (1:1)	Weighing method	298	101	0.096	–	91.9% (5 times)	[127]
p-Chlorophenol/DEA (1:1)	Weighing method	298	101	0.073	–	–	[127]
p-Cresol/MEA (1:1)	Weighing method	298	101	0.126	393	92.4% (5 times)	[127]
p-Chlorophenol/MEA (1:1)	Weighing method	298	101	0.122	–	–	[127]
p-Cresol/MDEA (1:1)	Weighing method	298	101	0.025	–	–	[127]
p-Chlorophenol/MDEA (1:1)	Weighing method	298	101	0.02	–	–	[127]
TBAB-MEA (1:4)	Pressure drop method	303	1000	0.106	–	–	[83]
[BTEA]Cl-MEA (1:4)	Pressure drop method	303	1000	0.100	–	–	[83]
TBAB-EAE (1:4)	Pressure drop method	303	1000	0.071	–	–	[83]
[BTEA]Cl-EAE (1:4)	Pressure drop method	303	1000	0.09	–	–	[83]
[TETA]Cl-DG (1:2)	Weighing method	313	101	0.159	373	97.5% (6 times)	[85]
[TETA]Cl-EG (1:3)	Weighing method	313	101	0.175	373	97.5% (6 times)	[85]
[TAE]Cl-EDA (1:3)	Weighing method	303	101	0.102	–	–	[29]
[UE]Cl-EDA (1:3)	Weighing method	303	101	0.118	–	–	[29]
[TEA]Cl-EDA (1:3)	Weighing method	303	101	0.171	–	–	[29]
[MEA]Cl-EDA (1:1)	Weighing method	303	101	0.202	–	–	[29]
[MEA]Cl-EDA (1:2)	Weighing method	303	101	0.250	–	–	[29]
[MEA]Cl-EDA (1:3)	Weighing method	303	101	0.287	–	–	[29]
[MEA]Cl-EDA (1:4)	Weighing method	303	101	0.312	–	–	[29]
[MEA]Cl-MEA (1:1)	Pressure drop method	313	800	0.092	–	–	[81]
[MEA]Cl-MEA (1:3)	Pressure drop method	313	800	0.109	–	–	[81]
[MEA]Cl-MEA (1:6)	Pressure drop method	313	800	0.149	–	–	[81]
[MEA]Cl-MEA (1:9)	Pressure drop method	313	800	0.187	–	–	[81]
[MEA]Cl-DETA (1:1)	Pressure drop method	313	800	0.114	–	–	[81]
[MEA]Cl-DETA (1:3)	Pressure drop method	313	800	0.176	–	–	[81]
[MEA]Cl-DETA (1:6)	Pressure drop method	313	800	0.242	–	–	[81]
[MEA]Cl-DETA (1:9)	Pressure drop method	313	800	0.284	–	–	[81]
[MEA]Cl-EDA (1:1)	Pressure drop method	313	800	0.114	–	–	[81]
[MEA]Cl-EDA (1:3)	Pressure drop method	313	800	0.184	–	–	[81]
[MEA]Cl-EDA (1:6)	Pressure drop method	313	800	0.327	–	–	[81]
[MEA]Cl-EDA (1:9)	Pressure drop method	313	800	0.346	–	–	[81]
[MEA]Cl-TETA (1:1)	Pressure drop method	313	800	0.099	–	–	[81]
[MEA]Cl-TETA (1:3)	Pressure drop method	313	800	0.166	–	–	[81]
[MEA]Cl-TETA (1:6)	Pressure drop method	313	800	0.178	–	–	[81]
[MEA]Cl-TETA (1:9)	Pressure drop method	313	800	0.205	–	–	[81]
[MEA]Cl-TEPA (1:1)	Pressure drop method	313	800	0.084	–	–	[81]
[MEA]Cl-TEPA (1:3)	Pressure drop method	313	800	0.101	–	–	[81]
[MEA]Cl-TEPA (1:6)	Pressure drop method	313	800	0.149	–	–	[81]
[MEA]Cl-TEPA (1:9)	Pressure drop method	313	800	0.172	–	–	[81]
TBAB-MDEA (1:4)	Pressure drop method	303	101	0.077	–	–	[35]
TMAC-MEA (1:5)	Weighing method	303	101	0.2459	–	–	[15]
TEAC-MEA (1:5)	Weighing method	303	101	0.2318	–	–	[15]
TEAB-MEA (1:5)	Weighing method	303	101	0.1926	–	–	[15]
TBAC-MEA (1:5)	Weighing method	303	101	0.1770	–	–	[15]
TBAB-MEA (1:5)	Weighing method	303	101	0.1628	–	–	[15]
amine-based ternary DESs							
TEAC-MEA-TEA (1:5:0.5)	Weighing method	313	101	0.2248	–	–	[15]
ChCl-MEA-TEA (1:5:0.5)	Weighing method	313	101	0.2349	–	–	[15]
TEAC-MEA-MDEA (1:5:0.5)	Weighing method	313	101	0.2360	–	–	[15]
ChCl-MEA-MDEA (1:5:0.5)	Weighing method	313	101	0.2383	–	–	[15]
TMAC-MEA-TEA (1:5:0.5)	Weighing method	313	101	0.2461	–	–	[15]
TMAC-MEA-MDEA (1:5:0.5)	Weighing method	313	101	0.2998	–	–	[15]
TMAC-MEA-LiCl (1:5:0.1)	Weighing method	313	101	0.3024	–	–	[15]
ChCl-MEA (1:7)	Pressure drop method	298	2000	0.157344	–	–	[132]
ChCl-MEA-DEA (1:7:1)	Pressure drop method	298	2000	0.130812	–	–	[132]
ChCl-MEA-MDEA (1:7:1)	Pressure drop method	298	2000	0.14784	–	–	[132]
ChCl-MEA-MDEA (1:7:5)	Pressure drop method	298	2000	0.14036	–	–	[132]
ChCl-MEA-AEP (1:7:1)	Pressure drop method	298	2000	0.150744	–	–	[132]
aqueous solution							

(continued on next page)

Table 5 (continued)

Solvents (mole ratio)	Absorption measurement methods	Absorption temperature(K)	P <sub>CO2</sub> (kPa)	Absorption (gCO <sub>2</sub> /g solvent)	Desorption temperature (K)	Regeneration efficiency (cycle times)	Refs
TEAC-MEA-TEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.2438	–	–	[15]
ChCl-MEA-TEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.2539	–	–	[15]
TEAC-MEA-MDEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.2550	–	–	[15]
ChCl-MEA-MDEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.2606	–	–	[15]
TMAC-MEA-TEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.2729	–	–	[15]
TMAC-MEA-MDEA (1:5:0.5) +10%H <sub>2</sub> O	Weighing method	298	101	0.3266	–	–	[15]
30% [MEA]Cl-EDA (1:3) +70%EG	–	–	–	–	373	97.5% (5 times)	[29]

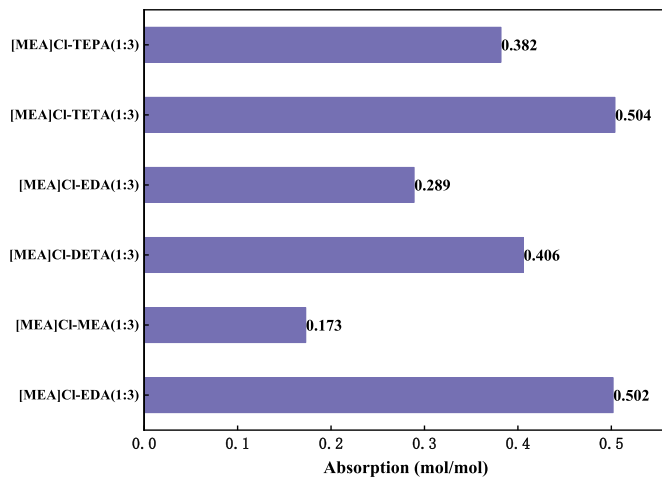


Fig. 10. Solubility of DESs with hydrogen bond acceptors of [MEA]Cl at 313K and 800 kPa [81].

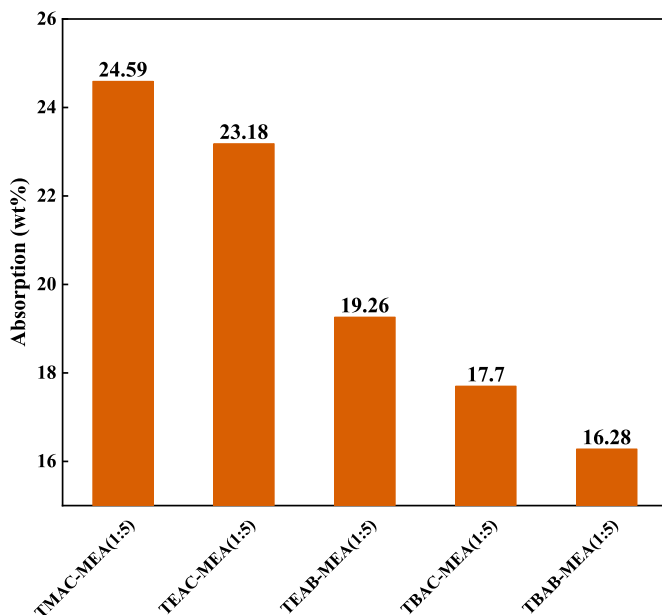


Fig. 11. Solubility of DESs formed from quaternary ammonium salts at 303 K, 101 kPa [15].

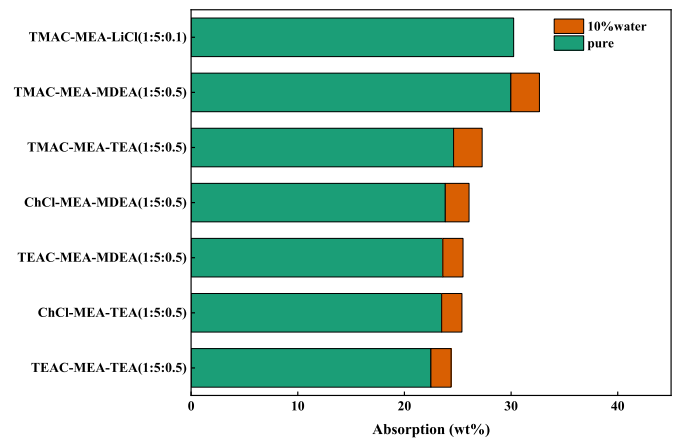


Fig. 12. The effect of water on solubility [15].

Table 6

Solubility of CO<sub>2</sub> in partially choline-based DESs.

Solvents (mole ratio)	Temperature (K)	Pressure (bar)	Absorption (g CO <sub>2</sub> /g solvent)	Methods	Refs
Ch [Pro]/PEG200 (1:1)	323.15	1	0.022	Physics, Chemistry	[133]
ChCl-MEA (1:8)	313.15	1	0.315	Physics, Chemistry	[128]
ChCl:LiAc (1:2)	298	50	0.101	Physics	[36]
ChCl/PhOAc (1:2)	298	10	0.0293	Physics	[17]
ChCl/EG (1:4)	298	10	0.013	Physics	[17]
ChCl/Urea (1:2.5)	298	10	0.112	Physics	[17]
ChCl/TEA (1:3)	298	10	0.198	Physics, Chemistry	[17]
30 wt% MEA	298	10	0.119	Physics, Chemistry	[17]

including thermal and oxidative degradation, also need to be considered.

Firstly, the high viscosity of deep eutectic solvents can seriously affect absorption kinetics, especially after loading CO<sub>2</sub>. The current methods used to solve the high viscosity of deep eutectic solvents mainly involve adding appropriate amounts of water and changing the ratio of hydrogen bond donors and acceptors. However, these methods are not sufficient to completely solve the high viscosity problem. To solve the



problem of high viscosity, it is necessary to systematically explore the molecular structure and carbon capture mechanism of deep eutectic solvents by establishing corresponding models.

Secondly, at this stage, the research on the stability of deep eutectic solvents mainly starts at the decomposition temperature because the degradation problem of deep eutectic solvents could cause the loss of solvents and increase operation costs. Therefore, the thermal and oxidative degradation of deep eutectic solvents also needs to be considered. How to design deep eutectic solvents from the molecular structure dimension to inhibit the degradation of deep eutectic solvents may be the direction of future research.

Finally, how to evaluate and screen deep eutectic solvents is also a question worth thinking about. The question of how to design deep eutectic solvents according to specific user needs is a long-term problem that needs to be studied. It is necessary to develop relevant conformational models and search for a set of methods to design deep eutectic solvents.

#### 4. Conclusions and outlook

Due to eco-friendly and harmonizable qualities, DESs have garnered significant interest. In this review, the structural characteristics of amine-based DESs are summarized. The factors affecting the physicochemical properties of amine-based DESs are analyzed. The conditions, methods, CO<sub>2</sub> solubility and regeneration performance of CO<sub>2</sub> capture by amine-based DESs are summarized.

The results indicate that amine-based DESs are the third type of DESs, consisting of organic salts and hydrogen bond donors. The physicochemical properties are affected by hydrogen bond donors, molar ratio and temperature. The hydrogen bonding donor has a decisive influence on the physicochemical properties. The CO<sub>2</sub> solubility of amine-based DES is dependent on HBAs, HBDs, molar ratio and temperature. The right amount of water will also increase CO<sub>2</sub> solubility. Most amine-based DESs can be regenerated with an efficiency of more than 90% after cyclical absorption and desorption. In addition, the potential solvents were chosen based on their attributes, which include a smaller molecular weight, alkanolamine as a hydrogen bond donor, chloride ion as a hydrogen bond acceptor anion, short alkyl chain, etc. Also, compared with choline-based DESs, amine-based DESs have the following primary advantages.

- a) The viscosities of amine-based DESs are lower than that of choline-based DESs. It helps to resolve problems such as high pumping costs and poor mass and heat transfer performance.
- b) CO<sub>2</sub> is absorbed using the chemical absorption method, which gets rid of the high-pressure and high-temperature steps, makes low concentration CO<sub>2</sub> absorption more efficient, and offers energy saving potential. It is more suitable for capturing CO<sub>2</sub> in flue gas.
- c) The melting point of amine-based DESs approximates the ambient temperature.

Further, amine-based DESs and choline-based DESs share structural components, and amines promise quite excellent potential for absorption in addition to the biodegradability of choline chloride. The most well-known DES is ChCl-MEA.

The absorption kinetics and stability of deep eutectic solvents is an urgent issue which needs to be addressed now. The development of models to design deep eutectic solvents may be a future direction in development.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

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

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