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A review of CO_2 capture for amine-based deep eutectic solvents

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ABSTRACT

Carbon Dioxide can be naturally produced and make up of 0.04 percent in the earth's atmosphere. However, centuries by centuries follows by the modernization of humans, it has escalated to the state of causing inimical impacts towards the human and environment. Additionally, one of the most important fuel sources in the world, natural gas, in its raw state contains high levels of CO_2 that must be removed to avoid issue such heating value reduction and pipeline corrosion concurrently reduces the effect towards environment. To tackle this issue, the development of CO_2 absorbent capable of absorbing large amount of CO_2 hence reducing the effects it has towards the environment has become a topic among researchers this past decades. Conventionally, the use of amine such as alkanolamine have been implemented to capture CO_2, yet it flaws outclass its uses such as limited ability to take up CO_2 and high corrosivity. Following this, ionic liquids (ILs), a new species of solvent, came to be in the limelight since they are tunable according to their function, including carbon capture. Nonetheless, in recent years, a newly discovered green solvent, deep eutectic solvent (DES) stole the limelight. This absorbent was claimed to retain all the advantages and desirable properties of ionic liquids while eliminating some of their greatest limitations. In this review, we examined the potential for carbon capture using multiple amine-based deep eutectic solvents and explored the role of including a third compartment as a performance enhancement mechanism. Last but not least, we are anticipating the publication of future species of amine-based deep eutectic solvents, which will include more information that had previously been considered scarce, such as its level of toxicity, its economic validation, and the evidence of melting point reduction.

1. Introduction

Carbon dioxide (CO_2) is one of the gases which contributes to the everlasting global warming that has become a concern for the last few decades. From past research, it has been found that CO_2 covers about 60 % of the global warming effect (Yamasaki, 2003).The factor that contribute to the increase in CO_2 concentration is possibly due to the human activities which contribute 86 % of the anthropogenic greenhouse gas emissions with China being the top country that emits CO_2 to the atmosphere by fuel combustion in 2019 (Metz et al., 2005; Global Energy and CO2 Status Report, 2019). In that year alone, the concentration of CO_2 in the atmosphere rise to 400 ppm which led to an increase of 0.8 °C in the global temperature (Change, 2014; Tans and Keeling, 2014). There are two main sources for the production of CO_2 which can be categorized as man-made and naturally occurred. For the first category, it can be further divided into four potential sources; (Yamasaki, 2003) industry, Metz et al. (2005) power plants utilizing fossil fuels, Global Energy and CO2 Status Report (2019) hydrogen production from feed stock rich of carbon and (Change, 2014) transportation (Sabouni et al., 2014a).

The increase of CO_2 has led to the results of global warming which affects most living things in the Earth. Climate change, increase in sea level, ocean acidification, droughts and hurricanes are a few of many effects of global warming that the earth has experienced this past few decades (Rubin et al., 2012; Bui et al., 2018; Mukherjee et al., 2019). Besides the effects towards the environment, CO_2 is also lethal to the

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human health. Long-term exposure to low-level CO_2 is not immediately lethal, however it may have adverse health effects for both healthy individuals and vulnerable populations (Rice and Susan Rice, 2004).

Aside from that, CO_2 is a prevalent impurity in the production of natural gas. The concentration of CO_2 in natural gas produced at the wellhead can rose up to 35 vol% (Wu et al., 2021). CO_2 together with H_2S and other acid gases, must be separated from natural gas as these contaminants can form corrosive compounds in the presence of water which lead to pipelines and other equipment to damage. By removing acidic CO_2 from natural gas, we mitigate the environmental impact of its emissions as well as minimize the reduction in heating value. CO_2 to an allowable value must be eliminated prior to distribution to gas users in order to meet gas quality criteria (Rufford et al., 2012). Two to three volume percent of CO_2 in natural gas pipelines are the typical range allowable before it can be distributed (Wu et al., 2021).

Therefore, to both reduce CO_2 emission and separation of CO_2 from natural gas, few technologies had been invented which make use of the postcombustion capture method as it can easily adapted to the existing plant due to its simplicity besides needing less energy than alternative processes compared to precombustion and oxycombustion (Aissaoui et al., 2017; Ochedi et al., 2021). Such technologies are absorption, membranes, adsorption, and cryogenic distillation (Sabouni et al., 2014b). According to reports, chemical absorption is the most suited for this separation process as it is the most conventional and cost-effective post-combustion technology procedure (Borhani et al., 2015). The common material uses for the absorption is by the use of liquid solvents (Rochelle, 2009).

Liquid absorbents exploiting amine-based absorbent for CO_2 capture via absorption process has been applied by the industry ever since as early as 1930s. Currently, this particular method has been considered as the most developed commercially accessible technique for capturing CO_2 (Wu et al., 2020). Their commercial used in the industry is due to their cheap price and high CO_2 capacity which singling them from other absorbents. In addition to all that, the process required only modest condition to operate (Abu-Zahra et al., 2007; Sarmad et al., 2020; Bains et al., 2017; Liang et al., 2016). The idea of the process requires the amine absorbent to be introduced on top of an absorption tower to flow down whereas at the bottom, a CO_2-containing flue gas stream is added with the aid of a blower to pump the gas. Around the temperature of 40 °C, carbamates are form due to the reaction of CO_2 with the amines that occurs via a zwitterion mechanism indicating its capabilities to bind CO_2 due to the specific functional group present in the amine absorbent.

Multiples species of amine have been utilized for the process with alkanolamine being the most popular. These alkanolamines are usually categorize based on the number of alkyl group attached to the nitrogen atom bonded to its amino group resulting in the classification of three different class, primary, secondary, and ternary amines. Commonly used alkanolamines includes monoethanolamine (MEA) for primary, diethanolamine (DEA) for secondary and N-methyldiethanolamine (MDEA) for tertiary (Wu et al., 2020). Despite that, there are a few major setbacks from the use of this type of absorbent such as low capacity to absorb CO_2, significant solvent loss due to the heating necessary for regeneration, high corrosivity and toxicity (Shaikh et al., 2015; Garg et al., 2016a; b). Hence, due to these issues, more absorbents must be studied to uncover future potential absorbents with minimal drawbacks (Aissaoui et al., 2017; Liang et al., 2016; Wibowo et al., 2021).

Recently, to overcome these issues, ionic liquids (ILs) have managed to enticed researchers during the past few decades as an alternative absorbent (Khaidzir et al., 2021). Since its emergence, the number of research published has risen to >5000 in 2016 alone. ILs are a class of molten salt in room temperature which composed of a cation and an anion which its interactions mediated by electrostatic forces with a recorded melting point to be below 100 °C. Compared to other liquids use for absorbent, ILs possess multiple specific dominance. The material has excellent chemical and thermal stability, has extremely low saturated vapor pressure, and has a wide range of electrochemical applications (Lian et al., 2021).

Despite that, in 2003, the emergence of a new class of ILs which has the potential to be a substitute to the ILs called deep eutectic solvents (DES) has once again piqued curiosity among the researchers for its extensive range of applications (Abbott et al., 2003a; Garcia et al., 2015). The DESs consist of two constituents, one of which functions as a hydrogen bond donor (HBD) and the other as a hydrogen bond acceptor (HBA) (Garcia et al., 2015; Francisco et al., 2013). It is easily synthesized via the intermolecular interactions between the two components often through hydrogen bond interaction (Hou et al., 2008; Zhang et al., 2012). They are frequently compared with ILs due to the identical properties that they possess. Nevertheless, though they shared the same properties the DESs doesn't inherit the limitation that the ILs own, given that the DESs are low both cost and toxicity, biodegradable and can be easily prepared without the need of high-cost equipment (Garcia et al., 2015). Notably, these benefits are the result of the careful selection of the chemicals that comprise DESs.

Fig. 1 depicts the number of research articles published in the last 20 years for CO_2 capture utilizing multiple technologies. As early as 2002, CO_2 capture has managed to garner the attention of researchers until modern days. Whereas in Fig. 2, the process of capturing CO_2 is more focused on the use of ILs and DESs. As can be see, the implementation of DESs for CO_2 capture is quite new as it only started to emerged in 2012 compared to the ILs. Nevertheless, there is an increasing trend year by year indicating that more and more researchers are exploring this intriguing topic with the potential of more combination of the DESs for multiple application to be discovered in the upcoming years. Even though the earliest DES recorded to be found is as early as 2003 (Abbott et al., 2003b), however the statistic was create purely based on the article published in ScienceDirect due to continuous reference materials that it offers besides the feasibility for readers to easily switch between books and journals as well as topics and disciplines including the credibility of the website itself.

This article provides an overview of green solvent as well as the understanding for the selection of the component of an amine-based DESs including alkanolamine, polyamine as Hydrogen Bond Donor with a few suitable potential Hydrogen Bond Acceptor. This review also aims to demonstrate the feasibility and advantages of substituting DESs for alkanolamines in the process of capturing CO_2.

2. Green solvent for CO_2 capture

To meet the demand of sustaining the environment development, the use of green solvents has been intensively studied by researchers for the last few couple of years as it can reduce the energy usage and ecological consequences causes by chemical manufacturing (Mulk et al., 2023). Such alternatives that have emerged or even the underrated alleged greener solvent like the task-specific ionic liquid (ILs), amino

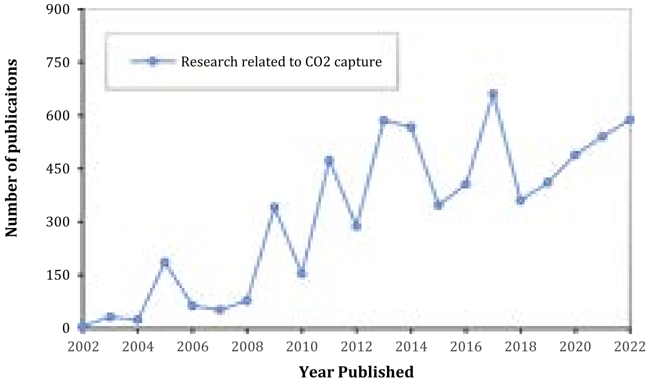


Fig. 1. The number of publications in topics related to carbon dioxide capture. (Data obtained in August 2022 via ScienceDirect).

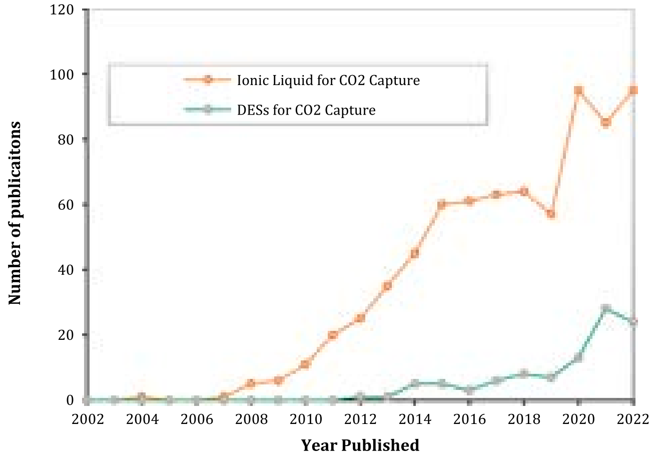


Fig. 2. The number of publications in topics related to ILs and DESs for carbon dioxide capture (Data obtained in August 2022 via ScienceDirect).

acid-functionalized ILs, IL-mixed solvents and the deep eutectic solvent (DESs) are propitious materials with qualities that cannot be achieved with any other material (Ali et al., 2023). Nevertheless, this study mainly focused only on ILs and DESs. Meanwhile, Fig. 3 depicted multiple types of solvent for the purpose of carbon capture, utilization and storage.

2.1. Ionic liquid

As previously mentioned in the introduction, ionic liquid is a new type of industrial solvents and has been a subject of a widespread interest among the researchers and scholars. Contrary from the conventional solvent, ILs is a liquid composed of cations and anions resulting from the fusion of low-melting salts (Ali et al., 2023). The components of ILs can be modified according to its functionality since the use of ionic liquid is not limited only for carbon capture. Notable for having melting point below 100 °C and liquid like state in room temperature (Singh and Savoy, 2020; Wilkes, 2002), it also has a wide range of physicochemical properties which are now recognized, ranging from those that are non-volatile, non-flammable, and both air- and water-stable to those that are volatile, flammable, and unstable.

This property arises as the result of various combination of cations and anions which follows the depiction of ILs (Lei et al., 2017). Its application in carbon capture has been recognized due to its unique characteristics which covers wide liquid range, thermally stable, negligible vapour pressure with also tuneable physicochemical character. These characteristics makes ILs potential green substitute to replace the conventional volatile organic solvents such as monoethanolamine (MEA) (Zeng et al., 2017). It is worth noting, however, that the greenness claim can only be verified through a full life cycle assessment (Mehrkesh and Karunanithi, 2016). One characteristic of the ILs stands out from the others which is its high CO_2 solubility which is fundamental for carbon capture (Hallett and Welton, 2011; Brennecke and Maginn, 2001; Hasib-ur-Rahman et al., 2010). Despite that, a few limitations needed to be overcome for it to be utilize in a large scale for industry use such as costly raw material chemicals, high viscosity, undetermined toxicity, and non-biodegradable (Thuy Pham et al., 2010; Biczak et al., 2014). Tables 1 and 2 display some commonly used cations and anions to produce ILs.

The history of ionic liquid dates back as early as 1914 when Paul Walden who was looking for molten salts in liquid states at the temperature which will allow him to utilise his equipment without any modifications stumble upon the discovery of $[EtNH_3][NO_3]$ which exists in liquid state at the temperature of 12 °C (Welton, 2018). Nevertheless, the discovery goes unnoticed for almost a century until the past two decades when it was rediscovered by Hiroyuki Ohno in 2000 by neutralization of amines to prepare Novel Room-Temperature Molten Salts (Hirao et al., 2000).

As the research on ionic liquids progresses, their tunability, resulting from the various combinations of cations and anions that conform to the definition of ionic liquids, gives rise to a wide range of behaviours. This in facts create a diversity of class for the ILs which has been categorized into various types, such as the room-temperature ionic liquids (RTILs), task-specific ionic liquids (TSILs), polyionic liquids (PILs), and supported ionic liquid membranes (SILMs). The latter category encompasses composites of ionic liquids that are supported on metal-organic frameworks (MOFs). The unique combination of organic and ionic characteristics in ionic liquids (ILs), along with the resultant intermolecular interactions, leads to a diverse array of phenomena, establishing a field of research that is both intriguing and demanding. ILs is notable for its vast range of applications with the most notable one as a highly effective solvents, whilst also demonstrating exceptional capabilities in various fields, which includes organic synthesis, catalysis, electrochemistry, metal separation, gas separation, biomass processing,

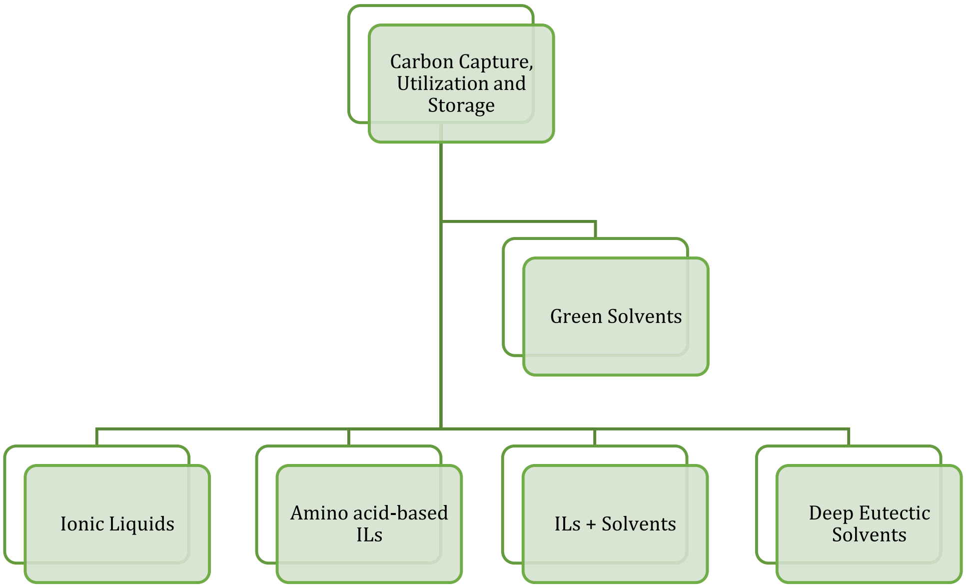


Fig. 3. Schematic representation of the carbon capture, utilization, and storage. IL, ionic liquid.

Table 1
List of commonly used cation for ILs.

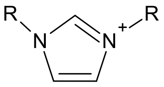
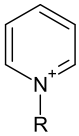
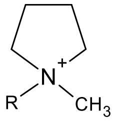
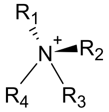
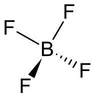
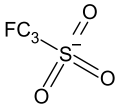
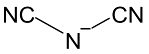
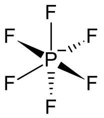
Species of Ions	Cation			
Chemical Structure				
Synonym	Imidazolium ion	Pyridinium ion	Pyrrolidinium ion	Tetraalkyl ammonium
Molecular Formula	$C_3H_5N_2^+$	$C_5H_6N^+$	$C_4H_{10}N^+$	NA
Molecular weight	69.09	80.11	72.13	NA

Table 2
List of commonly used anion for ILs.

Species of Ions	Anion			
Chemical Structure				
Synonym	Tetrafluoroborate	Trifluoromethane sulfonatetriflate	Dicyanamide	Hexafluoro phosphate
Molecular Formula	$[BF_{4]}$	$[OTf]^-$	$[N(CN)_2]^-$	$[F_6P]^-$
Molecular weight	86.81	80.11	67.05	144.96

pharmaceuticals, tribology, and energy storage systems such as batteries, supercapacitors, and fuel cells (Chen and Xu, 2023).

Nonetheless, application of ILs for the purpose of CO_2 capture still gaining the majority of the interest. The nature of the interaction between the CO_2 and the ionic liquids will determine the type of absorption process that takes place, which could either be chemical or physical (D'Alessandro et al., 2010). Usually, mechanism involves for this process is often based on physisorption that involves weak forces between the ILs and CO_2 molecules rather than chemical bonds with the heat of adsorption recorded around −11 kJ/mol. Due to the low heat of reaction, it is evident that minimum energy is sufficient for solvent regeneration (D'Alessandro et al., 2010). Lately, a study by Khan et al., have revealed the application of capturing CO_2 using ILs at room temperature which as a result, will reduce energy usage (Khan et al., 2017). The hybrid solvents are tested for multiples properties which includes surface tension, density, and viscosity by increasing either temperature or concentration of it.

Meanwhile, Indrajit and members focused on the polyionic liquids (PILs), as they explore the synthesis and characteristics of protic ionic liquids (PILs) derived from ethylenediamine, with the objective of improving the absorption efficiency of carbon dioxide (CO_2). The research investigated the influence of alkyl chain length in carboxylate anions on the thermophysical characteristics of polymeric ionic liquids (PILs). The findings indicated that an increase in alkyl chain length leads to enhanced solubility of CO_2, attributed to stronger intermolecular interactions. Among the tested polymeric ionic liquids (PILs), ethylene diammonium heptanoate ([ED][Hep]) demonstrated the greatest capacity for CO_2 absorption, with an uptake of 0.26 mol of CO_2 per mole of ionic liquid at a temperature of 298 K and a pressure of 0.1 MPa (Das et al., 2024).

Moreover, Xingyu and members introduced an innovative approach to improve CO_2 absorption through the utilization of a hybrid solvent that combines metal salts with task-specific ionic liquids (TSILs) within a microchannel reactor. This approach integrates metal salts, including $NaBF_4$, KBF_4, KCl, and $CuCl_2$, with task-specific ionic liquids (TSILs), particularly 1-aminopropyl-3-methylimidazolium tetrafluoroborate [Apmim][BF4], to improve the efficiency of mass transfer in the process of CO_2 absorption. The process of CO_2 absorption is characterized by the interaction of amino groups present in the ionic liquid with CO_2, resulting in the formation of carbamate. This compound can subsequently undergo hydrolysis to yield bicarbonate and ammonium ions. Additionally, the incorporation of metal salts modifies the interaction dynamics within the ionic liquid, thereby improving the efficiency of CO_2 capture. KBF_4 exhibits the most significant improvement in CO_2 capture capabilities when compared to other evaluated metal salts, particularly when incorporated into the [Apmim][BF_4] ionic liquid solution (Yang et al., 2024).

Mariuz and co-workers developed a functionalized water-imidazolium ionic liquid with ternary amines for CO_2 capture. Mixtures of two ionic liquids and five different amines with the addition of water as solvent makes up 10 model system to be studied. It is reported that the addition of water in the functionalized ILs resulted in an increase in absorption rate compared to the model with no water added. For example, when 33 % water was present in [emim][OcSO4], the increase in absorption rate rise to over 300 % compared with 1:1 amine: IL solution. Besides, the types of amines use also influenced the CO_2 solubility as the mixture of 33 wt% of[emim][OcSO4] and 33 wt% of H_2O if pair with 33 wt% TMG will result in better performance compare to 33 wt% DBU (Zalewski et al., 2021).

Cholinium-amino acid ionic liquids (AAILs) based on six different amino acids were synthesized by Noorani et al. to be use as carbon dioxide absorber. The capacity to absorb CO_2 by this system was investigated at temperature between 288.15 K to 318.15 K and pressure of 0 bar to 4 bars. FT-IR and ^{13}C NMR spectroscopy were used to confirmed the chemical absorption in AAILs. The result indicated that among the studied AAILs, highest CO_2 absorption capacity was shown in the AAILs composed of [CHO][Arg] whereas [CHO][Pro] has the lowest CO_2 absorption capacity. Results show that the AAILs with various amine sites have a greater potential to absorb CO_2 (Noorani and Mehrdad, 2022). Table 3 classify the absorption performance by a few species of ILs with its CO_2 solubility.

Table 3
Absorption performance by ionic liquid (ILs) – based solvent in 2016–2022 literature.

Absorbent	T(K)	P (bar)	CO_2 Solubility	Reference
1-butyl-3-methylimidazolium trifluoromethanesulfonate [bmim][OTf] + 30 wt% MDEA+ 3 wt% PZ	313.13	19.75	1.7691 mol $_{CO2}$/ mol$_{solvent}$	(Khan et al., 2017)
1-butyl-3-methylimidazolium acetate [bmim][AC+ 30 wt% MDEA+ 3 wt% PZ	313.13	19.61	1.8434 mol $_{CO2}$/ mol$_{solvent}$	(Khan et al., 2017)
[bmim][OAc] (33%) +DBU (33 %) + H_20 (33 %)	303.15	NA	2.32 ± 0.18 mmol/g	(Zalewski et al., 2021)
[emim][OcSO4] (33 %) TMG (33%) + H_20 (33 %)	303.15	NA	2.93 ± 0.20 mmol/g	(Zalewski et al., 2021)
[CHO][Arg]	288.15	4	0.6046	(Noorani and Mehrdad, 2022)
[CHO][Pro]	288.15	4	0.3886	(Noorani and Mehrdad, 2022)
[ED][Hep]	298.0	1	0.26 mol of CO_2/mole of IL	(Das et al., 2024)
[Apmim] [BF4] + KBF_4	–	–	–	(Yang et al., 2024)

2.2. *Deep eutectic solvent*

Deep eutectic solvent (DES) has emerged as a potential substitute for ILs and volatile organic chemicals (VOCs) for the decade. Contrary to ILs which generated from systems containing essentially a single type of discrete anion and cation, DES is formed from a eutectic mixture of Lewis or Brønsted acids and bases containing a range of either anionic and cationic species or both which act as hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD) (Smith et al., 2014). DES is classified as a new class of ionic liquid (ILs) due to the properties and characteristics that both solvent shares. It was asserted that this absorbent retained all the advantageous and desirable qualities of IL while resolving some of their most significant disadvantages. Both absorbents have low vapour pressure, comparatively broader liquid range, and similar absorption capacity. However, DESs has the upper hand due to the components for manufacturing cost much lower compare to ILs, low viscosity, simple synthesis process and lastly, in comparison to ILs, dual functionality is easily accomplished (Wang et al., 2021a). However, the most recognisable properties that the DESs own is its lower melting point compare to its individual components. The lower melting point is the result of the charge delocalization that occurs through hydrogen bonding, for instance between the halide ion of the hydrogen bond acceptor and the hydrogen-donor moiety. Fig. 4 discuss the comparison and similarity of ILs and DESs.

To prepare DESs, two or three components which have the capability to form either hydrogen bond or weak complexes are simply mixed to form a eutectic mixture (Zhang et al., 2012). Generally, moderate heating is occasionally required for certain mixture to reduce the time taken for the DES to form. Table 4 and 5 shows the list of common HBDs and HBAs use in the production of DES. The first DES synthesized was reported by Abbot and co-workers which constitute a combination of choline chloride as HBA and urea as HBD with a molar ratio of 1:2. The resultant DES was found to have a melting point of 12 °C which is lower than both of its component with choline chloride possessing a melting point around 305 °C, whereas the melting point of urea was recorded to be around 133 °C (Abbott et al., 2003a). Fig. 5 shows the procedure for the formation of ChCl-Urea DESs.

DESs are primarily categorized based on the characteristics of the complexing agent employed, as illustrated in Table 6. They are categorized into four different categories. DESs that are derived from MCl_x and quaternary ammonium salts, classified as type I, can be regarded as analogous to the extensively researched systems involving metal halides and imidazolium salts. Since there's a limitation for type I DESs in terms of low range of non-hydrated metal halides, the potential for deep eutectic solvents can be expanded by incorporating hydrated metal halides alongside choline chloride, thus forming type II deep eutectic solvents. Meanwhile, type III eutectics, composed of choline chloride and hydrogen bond donors, have garnered significant attention for their remarkable capacity to solvate an extensive array of transition metal species, encompassing both chlorides and oxides. Lastly, a selection of transition metals may be seamlessly integrated into eutectics at ambient temperatures, which are now elegantly referred to as type IV deep eutectic solvents. Due to this study aim to incorporate amine-based DES for CO_2 capture which falls under type III, most example will discuss more of the type III synthesized DES for carbon capture (Biczak et al., 2014; Environmental Chemistry for a Sustainable World, 2011; Pena-Pereira and de la Calle, 2018).

The application of DES in CO_2 absorption process have been suggested by Li and members as early as 2008, five years following its discovery using the same HBD and HBA when it was first found (Li et al., 2008). Since then, more studies and research has been conducted to test more HBDs and HBAs including glycerol, ethylene glycol, ammonium based, phosphonium based, and amine based, with the intention of

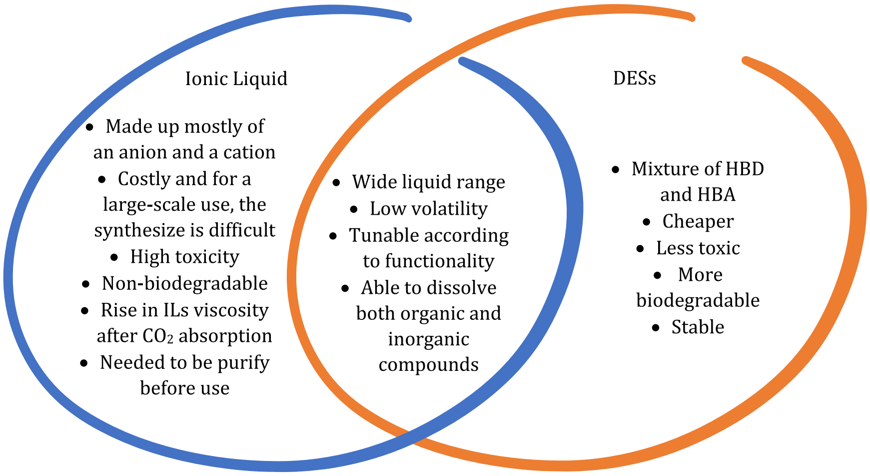


Fig. 4. Comparison between ILs and DESs.

Table 4
List of commonly used HBA.

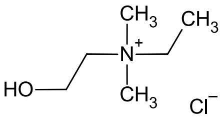
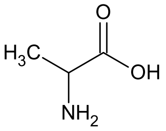
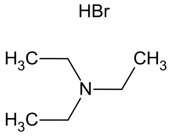
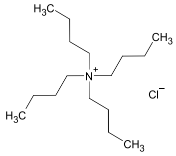
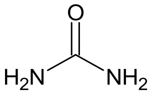
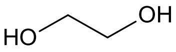
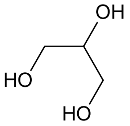
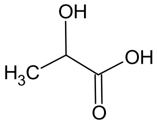
Type	Hydrogen Bond Acceptor			
Chemical Structure				
Synonym	Choline Chloride	Alanine	Triethylammonium bromide	Tetrabutylammonium chloride
Molecular Formula	$C_5H_{14}NO.Cl$	$C_3H_7NO_2$	$C_6H_{16}BrN$	$C_{16}H_{36}ClN$
Molecular weight	139.62	89.09	182.10	277.917
Melting Point	305 °C	272 °C-275 °C	246–248 °C	83–86 °C

Table 5
List of commonly used HBD.

Type	Hydrogen Bond Acceptor			
Chemical Structure				
Synonym	Urea	Ethylene Glycol	Glycerol	Lactic acid
Molecular Formula	C_2H_6	$C_2H_6O_2$	$C_3H_8O_3$	$C_3H_6O_3$
Molecular weight	60.055	62.068	92.094	90.078
Melting Point	133 °C	−13 °C	20 °C	18 °C

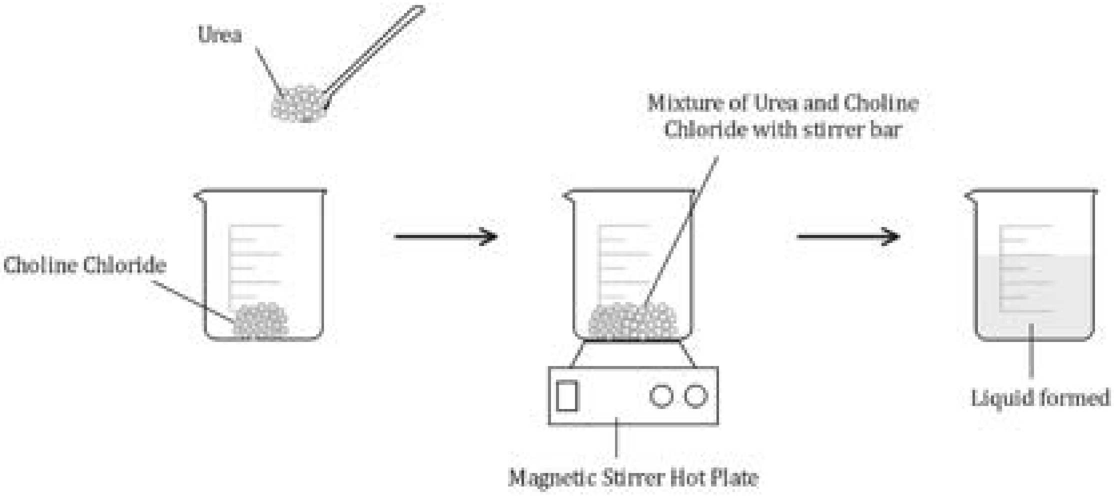


Fig. 5. Procedure for the formation of ChCl-Urea DESs.

Table 6
General Formula for Classifying DESs.

Type	General formula	Terms
type I	$Cat^+X^-zMCl_x$	M = Zn, Sn, Fe, Al, Ga, In
type II	$Cat^+X^-zMCl_x.yH_2O$	M = Cr, Co, Cu, Ni, Fe
type III	Cat^+X^-zRZ	Z = $CONH_2$, COOH, OH
type IV	$MCl_x + RZ = MCl_{x-1}{}^+ . RZ + MCl_{x+1}$	M = Al, Zn and Z = $CONH_2$, OH
Cat^+X^-zY = general formulae of DESs		Cat^+ = any ammonium, phosphonium or sulfonium cation X = Lewis Base Y = Lewis or Bronsted Acid z = number of Y molecules that interact with the anion

increasing CO_2 solubility (Nematollahi and Carvalho, 2019). The molar ratio between HBA and HBD appears to influence the CO_2 solubility of DES. It appears that the absorption pressure influences CO_2 solubilities in DES positively, whereas temperature is negatively impacted.

Despite that, among all types of DES, type III DES has been identified as the most used type of DESs being explored for the purpose of capturing CO_2. M.B. Haider et al. reported the synthesis of three different DES with identical hydrogen bond acceptor, (Methyl-triphenylphosphonium Bromide) paired with different hydrogen bond donor; Diethylene Glycol, Glycerol and Ethylene Glycol. These resulted synthesized DES can be categorized to Type III DESs as mentioned before due to the use of hydrogen bond donors, with either amide, carboxylic acid and alcohols present in it. The prospect of CO_2 absorption from flue gas is investigated experimentally. The experiment was conducted at various temperature (303 K, 313 K and 323 K) and up to 15 bars. FTIR analysis was conducted for the characterization of the DES before and after CO_2 capture process. The result depicted that diethylene glycol-based DESs showed the highest CO_2 solubility at the 303 K and 12 bar (Haider et al., 2021).

To compare, in 2017, Chunyan et al. aim to enhanced the solubility of pure CO_2 in a DESs as it is deemed that the sorption capacity is considered unsatisfactory compared with task-specific ILs despite high CO_2 sorption capacity and low viscosity. Hence, the adding of the DESs with a potential third component are also studied. A third component are added to both DESs synthesized, tetraethylammonium chloride - acetic acid (TEAC/AC) and tetrapropylammonium chloride - ethanolamine (TPAC/EA), both EA and glycerine (GLY), to each DESs, anticipating the CO_2 sorption to increase. The pressure and temperature are maintained before and after the process of functionalization. First species of DES, (TEAC/AC) was found to decrease its CO_2 solubility upon functionalization with either GLY or EA. Whereas, when compared with the second species of DES; (TPAC/EA), upon the functionalization with EA, an increase from 1.4 to 3.2 mol kg^{-1} DES for the CO_2 sorption can be observed compared to its pure DES state with no third component added (Ma et al., 2017).

Recently, Sang and co-workers studied a type III DESs, as they look into the efficiency of pure CO_2 captured utilizing the mixture of three DBN-based ionic liquids; [1,5-diazabicyclo[4.3.0]-non-5-ene + 1,2,4-triaz (Triz)] ([DBNH][Triz]), [1,5-diazabicyclo[4.3.0]-non-5-ene + 2-methylimidazole] ([DBNH][2-Melm]), [1,5-diazabicyclo[4.3.0]-non-5-ene + 2-Oxazolidone] ([DBNH][Oxa]), with each anion having different pK_a value. The DBN-based ionic liquids will be used as hydrogen bond acceptor (HBA) whereas its pair; the hydrogen bond donor (HBD) component were identified to be ethylene glycol (EG). This study was conducted while maintaining the temperature at 313.15 K and varied molar ratio of the HBD component ranging from 0.5 to 2. The result indicated that [DBNH][Oxa]-EG had the optimal absorption performance with the highest absorption capacity of 0.17 g CO_2/g DES. Moreover, it is proved that both IL and EG are able to react simultaneously with CO_2, via ^{13}C NMR spectra analysis to produce carbamate and carbonate. Hence, proving the existence of synergy between imino and hydroxyl group (Sang et al., 2022). Table 7 classify the absorption performance by several species of DESs with its CO_2 solubility.

3. Mechanism of CO_2 capture

3.1. Route

Since the emergence of deep eutectic solvent, many researchers have studied the application of deep eutectic solvent for the purpose of capturing CO_2. The pioneering work of Li and members have managed to grasp the attention of other scholars (Abbott et al., 2003a; Li et al., 2008) which used combination of choline chloride and urea. These DESs, however, are limited to physically absorb CO_2, which makes them impractical for capturing CO_2 from flue gases (Zhang et al., 2018a; Garcia et al., 2015). Besides the DES combination of urea and choline chloride, various other DESs formed by glycerol and lactic acid with choline chloride has also been studied and deemed to absorb CO_2 only physically.

Nonetheless, numerous studies have established that some DESs are capable to chemically capture CO_2. This can be seen as Wu and members studied the functionalized DESs formed by triethylenetetramine hydrochloride ([TETA][Cl]) and both ethylene glycol (EG) and diethylene glycol (DG). FTIR analysis demonstrates the existence of a chemical interaction between carbon dioxide (CO_2) with one of the DES's component; ([TETA][Cl]) (Zhang et al., 2018b). Good reversibility was also detected during the reversible absorption process. Moreover, Iwona and co studied the solubility of CO_2 in deep eutectic solvents based on 3-amino-1-propanol [AP] and tetraalkylammonium salts at low pressures. Besides the alteration in type of salts use, the molar ratio of the HBD components is also varied (1:8,1:6,1:4). The result depicts that DESs [TBAB][AP] at 1:4 molar ratio exhibits the best CO_2 uptake due to the chemisorption that is also confirmed with the use of Fourier Transform Infrared (FTIR) spectroscopy (Cichowska-Kopczyńska et al., 2021).

Several species have been identified in CO_2 absorption products which depends on the functional group of the DESs form. Based on Gurkan et al., who studied a new class of DESs consists of 1-ethyl-3-methylimidazolium 2-cyanopyrrolide ([Emim][2-CNpyr]) and EG at 1:2 molar ratio which are both thermally stable and low viscosity. From the result and discussion of the experiment, it was proposed that four routes could be taken with the basis of NMR and FTIR analysis as explained in Fig. 6 (Lee et al., 2021). The first route is the formation of carbamate group (N–COO) as the pyrrolide anion complexing with CO_2 molecule. Next, in this reaction, the imidazolium cation is protonated by an anion, which forms a carbene zwitterion and then binds CO_2 to form carboxylate (–C–COO). Moving on, Route 3 involves protonating the anion with EG, converting it into deprotonated EG, which is then reacted with CO_2 to form carbonate(−O−COO). Lastly, the last route discussed the formation of bicarbonate molecule (HO–COO) in the presence of water (Wang et al., 2021b).

To further proven this proposed CO_2 capture mechanism, Mingzhe and Xu study the CO_2 capture mechanism by DES formed by Choline Prolinate and Ethylene Glycol. Recently, amino-functionalized deep eutectic solvents (DESs), composed of choline prolinate ([Ch][Pro]) and ethylene glycol (EG), were synthesized for the purpose of CO_2 capture. Mechanistic investigations indicated that CO_2 interacts with the anion [Pro]− to produce carbamate acid, while no reaction occurs with EG. This study further explores the mechanism of CO_2 capture utilizing [Ch][Pro]-EG deep eutectic solvents (DESs), as a result, the findings derived from nuclear magnetic resonance (NMR) and Fourier transform infrared (FTIR) analyses indicate that carbon dioxide (CO_2) can react with both the anion [Pro]− and ethylene glycol (EG). The NMR analysis reveals a

Table 7
Absorption performance by deep eutectic solvent (DESs) – based solvent in 2016–2022 literature.

DES	Molar Ratio	T (K)	P (bar)	CO_2 Solubility	Reference
methyltriphenylphosphonium bromide – ethylene glycol	1:4	303	12	0.074 mol CO_2/mol DES	(Haider et al., 2021)
methyltriphenylphosphonium bromide – glycerol	1:3	303	12	0.075 mol CO_2/mol DES	(Haider et al., 2021)
methyltriphenylphosphonium bromide – diethylene glycol	1:4	303	12	0.1 mol CO_2/mol DES	(Haider et al., 2021)
Tetrapropylammonium chloride - ethanolamine	1:4	298.15	18	1.4 mol kg^{-1}	(Ma et al., 2017)
Tetrapropylammonium chloride - ethanolamine + acetic acid	1:2:2	298.15	18	3.2 mol kg^{-1}	(Ma et al., 2017)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 1,2,4-triaz (Triz)] – ethylene glycol	1:0.5	313.15	1.01325	0.15 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 1,2,4-triaz (Triz)] – ethylene glycol	1:1	313.15	1.01325	0.135 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 1,2,4-triaz (Triz)] – ethylene glycol	1:2	313.15	1.01325	0.12 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-methylimidazole] – ethylene glycol	1:0.5	313.15	1.01325	0.165 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-methylimidazole] – ethylene glycol	1:1	313.15	1.01325	0.13 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-methylimidazole] – ethylene glycol	1:2	313.15	1.01325	0.11 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-Oxazolidone] – ethylene glycol	1:0.5	313.15	1.01325	0.17 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-Oxazolidone] – ethylene glycol	1:1	313.15	1.01325	0.14 g CO_2/g DES	(Sang et al., 2022)
[1,5-diazabicyclo[4.3.0]-non-5-ene + 2-Oxazolidone] – ethylene glycol	1:2	313.15	1.01325	0.12 g CO_2/g DES	(Sang et al., 2022)

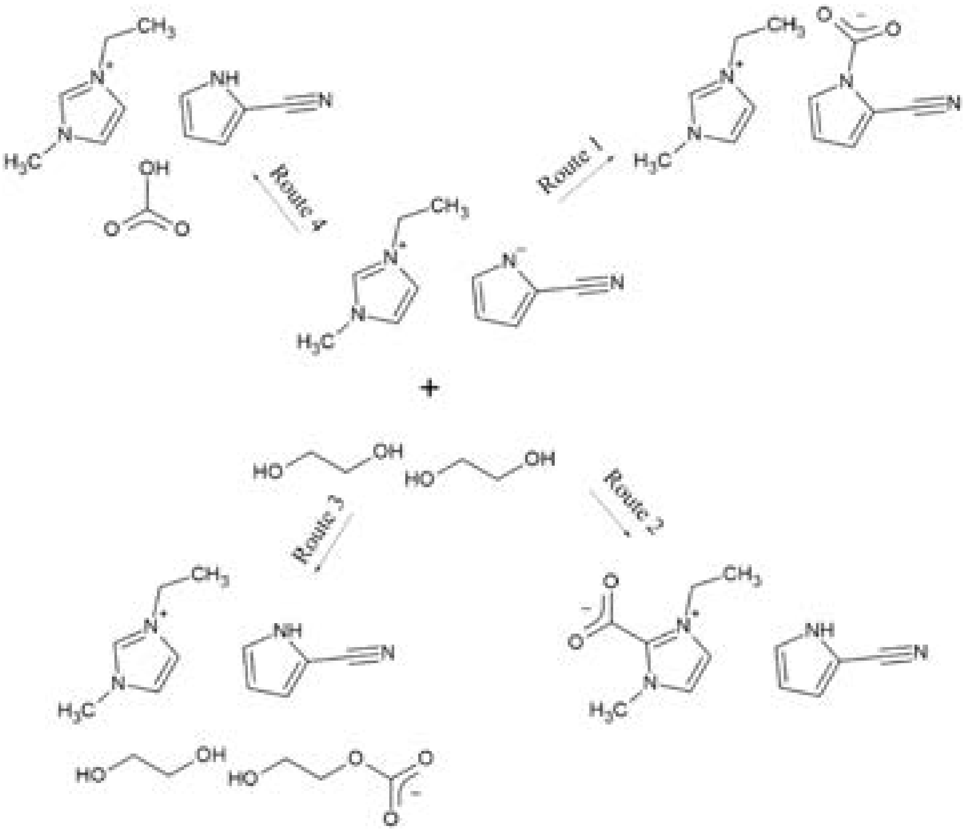
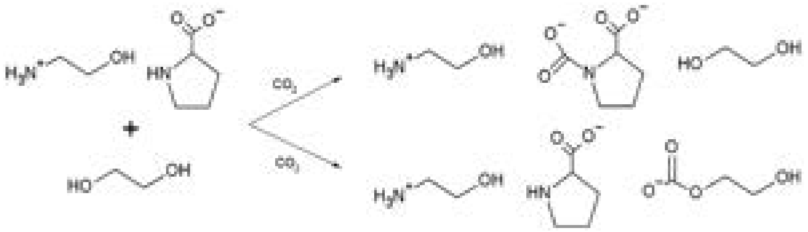
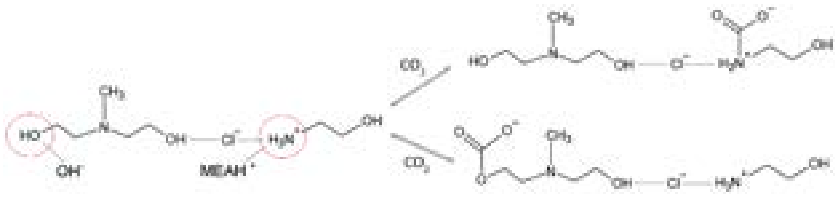


Fig. 6. Proposed reaction network.

new peak at 159.7 ppm, indicating the presence of the carbonyl carbon in carbamate acid, which suggests a reaction between CO_2 and the anion [Pro]-. Whereas, the newly observed peaks at 157.8 ppm are linked to the carbonyl carbon of the carbonate species generated from the reaction between CO_2 and EG. This additionally validates the suggested reaction network, as the carbamate formation corresponds to route one, while the carbonate formation aligns with the proposed route three (Chen and Xu, 2023).

Moreover, further mechanism of CO_2 interaction with DESs has been provided by Ahmad and Lin via the use of nuclear magnetic resonance (NMR) and Fourier transform infrared spectroscopy (FTIR) following DFT calculations and MD simulations. It has been found that MDEA-

Table 8
Mechanism proven by multiple scholars via characterisation.

Species	Functional Group Present	Functional Group Formed	Reference
	–NH –OH	Carbamate (N–COO) Carbonate (–O–COO)	(Chen and Xu, 2023)
	–NH –OH	Carbamate (N–COO) Carbonate (–O–COO)	(Ahmad et al., 2021)
	–NH	Carbamate (N–COO)	(Das et al., 2024)

based DESs, Monoethanolamine hydrochloride pair with methyldiethanolamine ([MEAHCl][MDEA]) which is also labelled as DES 1 exhibit the best performance when compared to the other hydrogen bond donors after 6 h absorption. The FTIR characterization for before and after CO_2 absorption process reveals two new peaks at 1363 cm^{-1} and 1575 cm^{-1}. Consequently, these results indicate carbamate and carbonate formation, indicating that CO_2 reacts with DES 1 via $MEAH^+$ and -OH groups hence validating earlier proposed CO_2 capturing mechanism via DESs (Ahmad et al., 2021).

To boot, the mechanism of CO_2 capture has also been discussed in the study conducted by Zhang and Huang which focusing on Task-Specific Deep Eutectic Solvent. In this study, 1,5-Diazabicyclo[4.3.0] non-5-ene and imidazole ([DBNH][Im]) are synthesized and were later characterized by in-situ IR spectra to analyse it reaction with CO_2. The investigation shows the formation of new peak at 1678 cm^{-1}, which is due to the stretching vibration of C–O in O═C–O as it absorbs CO_2. Whereas the absorption peak at 1044 cm^{-1} confirmed the formation of carbamate as it indicates the newly formed C–N bond (Das et al., 2024). Table 8 list all the mechanism discussed in this chapter.

3.2. *Common functional group discovered in DESs to undergo reaction with CO_2*

From all the above proposed routes, as can be seen, there is no doubt about the role of both hydroxyl and amino group play for the formation of several species as it chemically reacts with the CO_2. These functional group can be detected on either the HBA component nor the HBD component of the DES.

For hydroxyl group, the structure consists of two oxygen atoms bonded to hydrogen atoms with two lone pairs each. Hydroxyl groups are represented by the chemical formula -OH. This functional group is usually attached to an organic compound which is a covalently bonded carbon-hydrogen compound. It is easy for them to form hydrogen bonds, generating either positive or negative charges ion. The term alcohol refers to any organic compound that contains one or more hydroxyl group attached to the carbon atom of an alkyl group (Tisserand and Young, 2014).

Whereas, in an amino functional group, nitrogen is the central component. The nitrogen atom binds to the two hydrogen atoms through a single covalent bond. NH_2 is the molecular formula of the amino group. Due to this electronegative structure and lone pair of electrons, it can either form hydrogen bonds or maintain its net positive charges. The term amine refers to any organic compound that contains amino group (Klecker and Nair, 2017). From the image below, it can be seen the structural formula of amino group is enclosed in the blue box. Meanwhile, the hydroxyl group is enclosed in the red box as shown in Fig. 7.

3.3. *Roles of amino group in CO_2 capture*

The existence of amino group in solvent used for CO_2 capture isn't something new as alkanolamine has been used as conventional solvent decades ago. When anhydrous conditions are met, two equivalents of amine can react with one equivalent of CO_2 to form one equivalent of carbamate. The use of amine-based DESs has been studied by multiples researchers at various parameters which includes molar ratio, pressure, and temperature. However, the literature published were still considered limited unlike any other DES.

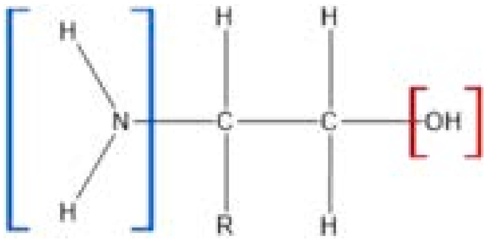


Fig. 7. Example of hydroxyl and amino group attached to an alkyl group.

In recent years, a new amine based DESs; EAHC: TEPA was successfully synthesized at 1:9 molar ratio to determine carbon dioxide solubility. To evaluate the physical properties of the synthesized DESs (EAHC: TEPA) with various ratios, density, refractive index, and viscosity were measured under atmospheric pressure and at temperatures ranging from 298.15 K to 333.15 K. The trend for increasing solubility as the rise in pressure follows the similar trend with previous literature. In addition, the decrease in solubility as the temperature rises also follows the trend of previous literature due to weakened intermolecular interactions, resulting in a higher rate of escape from solutions. Further interaction of the synthesized DES with CO_2 is analysed with both FT-IR and NMR spectroscopy to confirm the formation of hydrogen bonds indicating the successful formation of the DES besides the formation of new groups after CO_2 absorption such as carbamate and bicarbonate (Pishro et al., 2021).

In addition to the above-mentioned literature, the use of amine in DES's functionalization has also been studies by Sarmad et al., who designed five novel three-component DESs by functionalization with different species of amines. Choline chloride-ethanolamine at 1:7 molar ratio was functionalized with various amine; diethanolamine (DEA), methyldiethanolamine (MDE), piperazine (Pz) and 1-(2-aminoethyl) piperazine (AEP). The CO_2 solubilities of [(ChCl-EA) Pz] is found to display the highest compare to the other synthesized DES. The great performance can be attributed due to the fact that Pz was used as an absorption activator, the addition of Pz can improve the absorption capacity of CO_2. In contrast to that, lowest CO_2 absorption was discovered in [(ChCl-EA) DEA] which can be deduced as a result of adding a secondary amine. Formation of carbamate between the reaction of ethanolamine and CO_2 is further confirmed by NMR spectroscopy as new peak form at 162.5 ppm which the formation of carbamate is attributed. However, upon characterization with secondary/tertiary amines, carbamate peak was found to shifted from its original position. A trend can be form to depict the reactivity of amines with CO_2 upon characterization/blending: Primary > Secondary >Tertiary (Sarmad et al., 2020) as shown in Fig. 8.

To study the effect of amine class on the performance of carbon dioxide capture, Liu et al. synthesized three different DESs with choline chloride (ChCl) as the HBA while pairing it with various type of alkanolamine such as ethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) at the optimum molar ratio. The following DES are tested for their physical properties which includes viscosity with ChCl-DEA having the greatest viscosity followed by ChCl-MDEA and ChCl-MEA. Past literature stated that as viscosity increase, the absorption performance reduces. There are several aspects that are affected by a higher viscosity, including a slower diffusion of CO_2 and restriction of the flow of CO_2-loaded solvent into the bulk liquid (Cichowska-Kopczyńska et al., 2021; Shukla and Mikkola, 2018). Despite that, the result for CO_2 capture shows that the performance of ChCl-DEA is slightly better than ChCl-MDEA although having the highest viscosity with ChCl-MEA showing the largest adsorption capacity. The adsorption capacity of the synthesize amine-based DES are following the order:

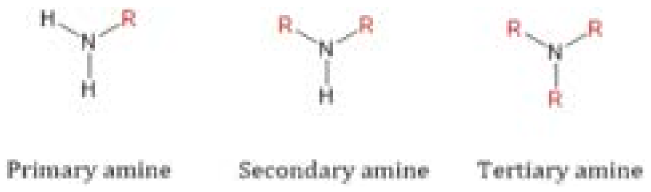


Fig. 8. Class of amine.

[ChCl][MEA] > [ChCl][DEA] > [ChCl][MDEA]

Potential explanation for the result could be ChCl-DEA, a secondary amine undergoes chemisorption with CO_2 while in contrast to ChCl-MDEA, a tertiary amine undergoes physical adsorption. Despite the higher viscosity that it possesses, chemical interactions between the DES and CO_2 obscure the effects of viscosity. Nonetheless, despite both ChCl-DEA and ChCl-MEA undergoes chemisorption, the effects of viscosity play a key role as ChCl-DEA has a higher viscosity than ChCl-MEA. As a result, ChCl-DEA absorbs less CO_2 than ChCl-MEA, which is a reasonable result (Liu et al., 2022). From the result, it correlates with the above studies as similar trend can be found based on the class of amine use for CO_2 capture.

However, a recent discovery by Shukla et al., who studied the effect the effect of HBD on the uptake of CO_2 as shown in Fig. 9, found that several potential HBA when paired with a few amine-based HBD shows that the uptake of CO_2 in both HBA; [MEA.Cl]- and [HMIM.Cl]- decreases as the number of imine group (-NH) in HBD rises. Various amine-based HBD includes ethylenediamine [EDA], diethylenetriamine [DETA], tetraethylenepentamine [TEPA], pentaethylenehexamine [PEHA]. The result indicates that both HBA when paired with EDA shows the highest efficiency for capturing CO_2 after 60 min while pairing it with PEHA ranks it rock bottom. DESs containing [PEHA] have a lower CO_2 uptake capacity than DESs containing [EDA] due to their higher viscosities. As a result, CO_2 molecules are not able to diffuse into the active sites due to high viscosity, thereby preventing rapid CO_2 uptake. The formation of carbamate soon after CO_2 uptake in solvent is confirmed with the assistance of both FT-IR and ^{1}H- and ^{13}C NMR spectra. To conclude, for a chain of alkyl polyamine, as more imine group are added, viscosities is found to reduce hence lowering the efficiency of CO_2 capture (Shukla and Mikkola, 2018). The elongation of the alkyl polyamine can be clearly spot in Table 9.

4. Roles of water in amine-based DES

Even though DESs exhibit numerous benefits compared to other solvent in terms of capturing CO_2, few combinations of species still suffer high viscosity for a huge scale industry. Most synthesized deep eutectic solvents exhibit a comparatively high viscosity, which contributes to sluggish mass transfer and slow sorption kinetics, potentially necessitating larger equipment dimensions. Nonetheless, several approaches to improve the CO_2 absorption rate has been studied in recent years including the use of blends in amine-based DES and adding co-solvent to reduce its viscosity. In this work, we focused on the use of water as a co-solvent to investigate the effect towards the DES's absorption performance.

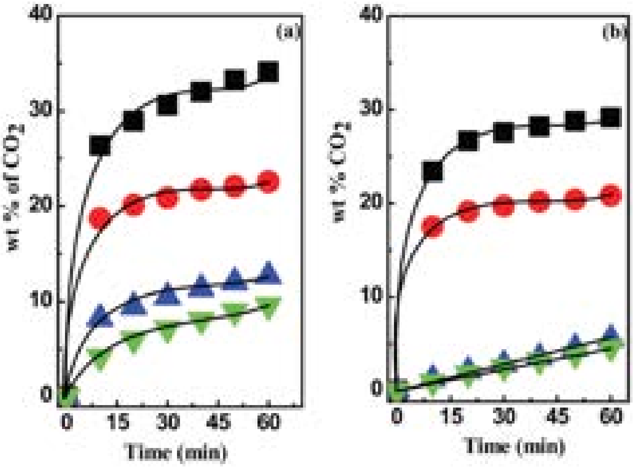


Fig. 9. Intake of CO_2 kinetics in (a) [MEA.Cl]- and (b) [HMIM.Cl]- paired with various HBA – EDA (black), DETA (red), TEPA (blue) and PEHA (green) at molar ratio of 1:4.

Just recently, in 2022, Xiangwei Liu et al. investigated the performance of CO_2 absorption of a few amine-based DES which differs based on their class of amino group; ethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) influenced by the addition of water content (Liu et al., 2022). All these amines are paired with the same HBA; ChCl. According to the author, water addition in both ChCl-MEA and ChCl-DEA decreased the absorption of CO_2 and the inhibition effect become more evident as water content in the DES increases. Hydrogen bond networks are destroyed when water is added to the DES, resulting in a reduction of amino groups concentrations. In summary, DES containing primary and secondary amines are greatly inhibited by water. Since ChCl-MDEA undergoes physical adsorption, hence it is foreseeable for both ChCl-MEA and ChCl-DEA to have better CO_2 absorption. The reason for this is for tertiary amine and CO_2 to undergo reaction, the participation of water is mandatory. The reaction process is graphically explained as follows:

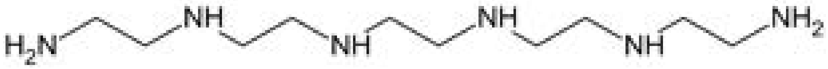
$$R_1R_2R_3 + CO_2 + H_2O \leftrightarrow \ HCO_3^- + \ R_1R_2R_3NH_2^+$$

Result shows that introducing water to the system at 10 wt% after 60 min of reaction shows an absorbtivity of 0.0834 g g^{-1}. This indicated that chemical adsorption occurred as well as physical adsorption. An increase in water content has been observed to result in a decrease in CO_2 absorption; however, when compared to the anhydrous system, which solely adsorbs CO_2 through physical means, the absorption remains higher. This suggests that water could promoted the system to absorb CO_2 as one of the co-solvent candidates (Liu et al., 2022). Xiangwei and teams concluded, in their current work, it is indicated that the water content plays a crucial role in influencing the CO_2 absorption capacity of deep eutectic solvents (DES). It has been observed that deep eutectic solvents (DESs) containing primary and secondary amines show a reduction in CO_2 absorption as the water content rises. In contrast, DESs with tertiary amines exhibit an increase in chemical adsorption with the addition of water, resulting in enhanced CO_2 absorption; however, an excessive amount of water will ultimately lead to a decrease in this absorption capacity (Fig. 10).

A similar trend was noted in the research conducted by Chunyan Ma et al., who examined the influence of water as a co-solvent in glycerol-based deep eutectic solvents (DES), which exhibited notably high viscosity. A typical species of glycerol-based DESs with high viscosity, benzyl trimethylammonium chloride – glycerol (BTMA/GLY) with a molar ratio of 1:2 was chosen out of 15 high performance DESs to be added with water. The viscosity of the water functionalized DES can be seen to reduce drastically from 716 to 20 mPa.s. As a result, the CO_2 performance was found to be improved by 25 % as it increased from 0.26 to 0.33 mol kg^{-1} DES at a time span of one hour. The phenomenon observed may be ascribed to the modification of the three-dimensional network structure of the pure BTMA/GLY (1:2) mixture resulting from the addition of water. This modification results in a decrease in viscosity, thereby improving the mass transfer rate. Identical pattern can be observed from past scholars as it is also reported that the CO_2 solubility first increased then drop as the amount of water is further increased (Ma et al., 2017) (Fig. 11).

Report regarding functionalized DES with water as co-solvent has also been reported by Trivedi and Lee, who test the DES [MEA.Cl][EDA]=1:3 with various water contents ranging from 5 %~20 %. Result obtained in Table 10 and Fig. 13 shows the uptake kinetics of CO_2 improves significantly compare to its original state (no water added) in a period of 60 min. The improvements in kinetics are due to the fact that water weakens the intermolecular hydrogen bonding interactions between the DESs through its own interactions with the DES itself. A decrease in intermolecular interactions leads to a decrease in viscosity and an increase in uptake kinetics. The formation of the water-DES network results in increased free volume of DES, hence resulting in

Table 9
Hydrogen Bond Donor pair with HBA [MEA.Cl]- and [HMIM.Cl]-.

Hydrogen Bond Donor	Name
	Ethylenediamine [EDA] Diethylenetriamine [DETA] Tetraethylenepentamine [TEPA] Pentaethylenehexamine [PEHA]
	
	
	

the kinetics improvement. Despite that, identical trend can be observed as with the increase in water content, the uptake behaviour exhibited instability; specifically, at 20 % water content, a decline in uptake was observed immediately following the initial uptake point. Excess water can attack the carbamate, resulting in carbonic acid as an intermediate compound, which dissociates into CO_2 and water (Trivedi et al., 2016). Fig. 14 illustrate the effect of excess water towards DES performance (Fig. 12).

However, some study records the negative effects for the functionalization of water in DESs as the study on the application was conducted by Shukla and Mikkola. A eutectic mixture of monoethanolammonium chloride (MEA-Cl) and ethylenediamine (EDA) was synthesized at 1:4 molar ratio. Water was added to the eutectic mixture with the intention of simulating CO_2 uptake under more realistic conditions. From Fig. 14.0, it was discovered that CO_2 absorption kinetic revealed a decreasing trend as the weightage of water content in the solvent increases in comparison with no water added. Contrary to the 39.0 wt% of CO_2 captured by pure (MEA-Cl/EDA), the value was found to decline to 35.7 wt%, 35 wt% and 34.4 wt% for 10 %, 20 % and 30 % of water added in the eutectic solvent respectively. This pattern develops as a result of CO_2 and water competing for the active sites in DES, which causes a decrease in CO_2 absorption. With increasing water content, this competition intensifies and reduces aqueous DES's ability to absorb CO_2 (Shukla and Mikkola, 2018).

5. Future work and development for CO_2 absorption

5.1. Evaluation of DES toxicity

DES is known for its "green traits" as it is generally far less in terms of toxicity compare to ILs. Despite the description, this is somewhat inaccurate as in recent years, researchers have recognized that they are not fully green as the earlier report claimed it to be. It is undeniable that it is an alternative for a greener material but this does not represent for it to be non-toxic. In this research, the type of DES mainly focus on is type III which include a diverse range of amides and polyols, including urea, glycerol, ethylene glycol, fructose, and erythritol, etc., having low inherent toxicity. However, low toxicity does not indicate that it is harmless and labelled as "green."

To this date, the research on toxicity level of newly founded DESs are scarce and mostly are not included in published paper. The toxicity level of DES must be measured and included in the research to ascertain either it is harmless to both the environment and people. To studied its effect towards human cell, recently Li and members measure the toxicity of amino acid-based with the help of A459 cells (human non-small cell lung tumour cell) which were seeded and cultured. Following this, the medium was changed to DMEM medium containing different concentrations of amino acid-based DESs for 24 h at 37 °Celsius. From this test, the toxicity level of each amino acid-based DESs were determined (Li et al., 2023).

Besides, in terms of environmental standpoint, future research paper must include a proper way to dispose the newly founded DESs after the toxicity level has been determined in the future. In such way, there will be a few methods on ways to dispose DES based on their toxicity level which will add more value to the definition of "green materials." For a safe, harmless, and biodegradable DESs, they can be safely release to the environment. Whereas, for DES that has been deemed exceed the toxicity level that is permitted to be labelled as harmless, they need to be disposed according to the law of their respective nations to avoid any toxic pollution due to illegal waste disposing.

5.2. Economic validation for DES value

Deep eutectic solvent is well known for its low-cost material as compared other materials that are more costly such as its substitute, ILs. Due to its low cost, it had eased the research and studies for new DESs to be discovered in the future. Nonetheless, from the past research paper, no such data has been discovered to be included regarding the expenses for each material used for the synthesis of DES hence, the validation for its economic value has keep the students in dark. For the fresh undergraduate students who involved in the research of DESs, such information would prepare them in for their research in terms of using the best materials for DESs synthesis based on their research budget given. Future works should include all the expenses use for DESs synthesis in order to further strengthen and proof the claim made by past researchers and scholars.

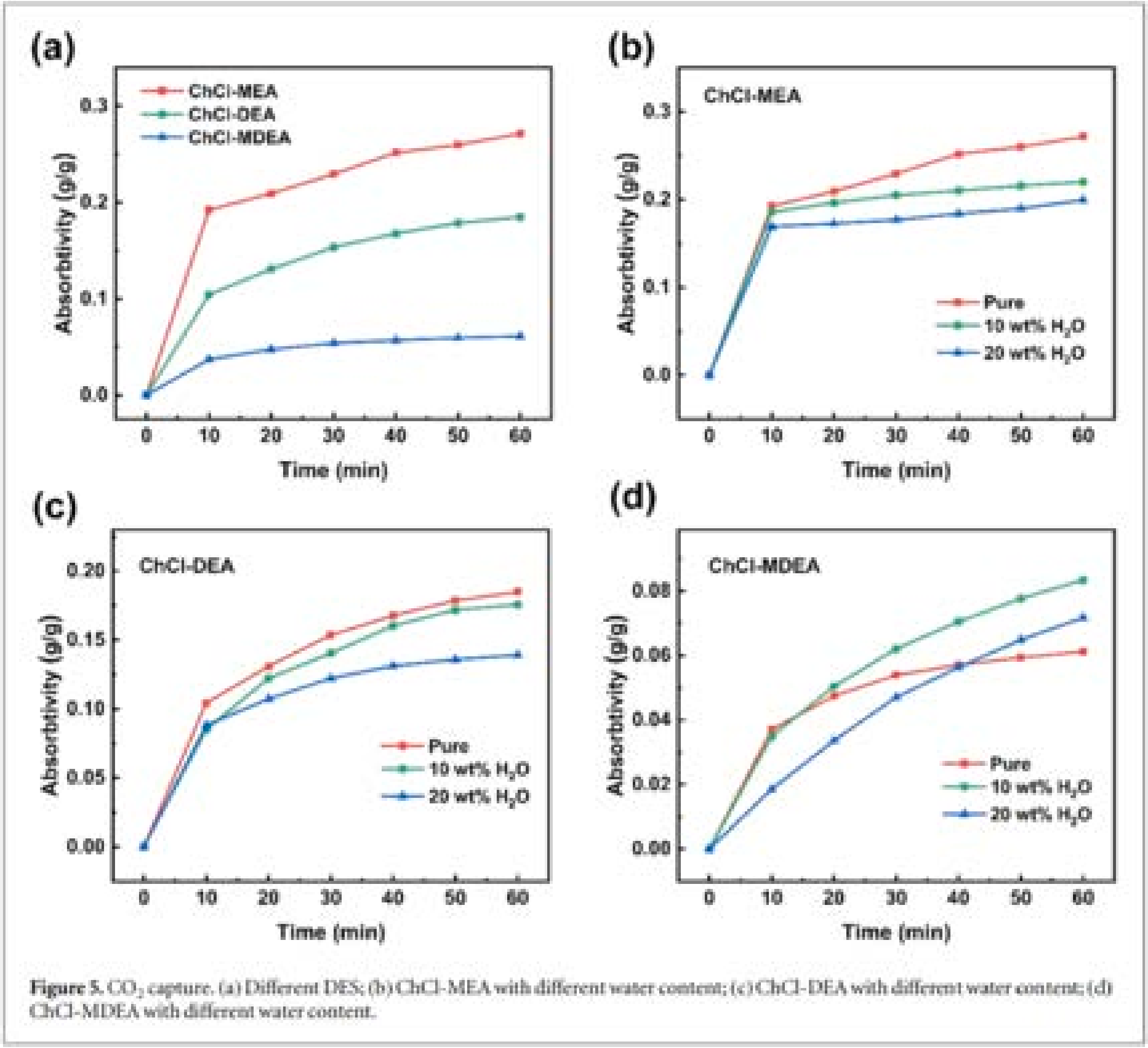


Figure 5. CO_2 capture. (a) Different DES; (b) ChCl-MEA with different water content; (c) ChCl-DEA with different water content; (d) ChCl-MDEA with different water content.

Fig. 10. Carbon Dioxide Capture. (a) DES capture capacity according to their species (b) CO_2 capture for ChCl-MEA with various water content (c) CO_2 capture for ChCl-DEA with various water content (d) CO_2 capture for ChCl-MDEA with various water content.

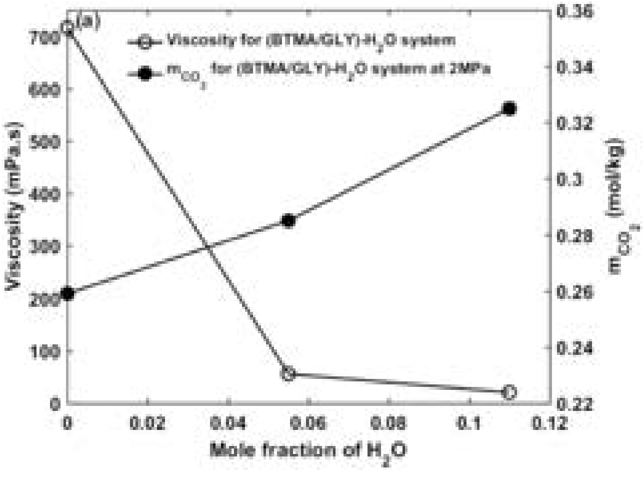


Fig. 11. The quantity of CO_2 absorbed at one hour, along with the viscosity of the BTMA/GLY (1:2) mixture, influenced by the addition of a small amount of water.

Table 10
Performance of [MEA.Cl][EDA]=1:3 with various water content.

DES	Water Contents (%)	wt% of CO_2 at the first uptake
[MEA.Cl][EDA]=1:3	0	25.2
	5	26.9
	7.5	27.3
	10	27.5
	20	28.1

5.3. Corroboration for the drop in melting point of synthesized DESs

A DES consists of large, nonsymmetric ions with a low lattice energy, resulting for the drop in melting point obtained. The charge delocalization that occurred resulted from the hydrogen bonding forming between hydrogen bond donor and hydrogen bond acceptor causes the melting point of the mixture decreases in comparison to the melting points of its constituents. The data for the decrease in melting points of the mixture as compared to its constituents rarely shown in most paper published for newly founded DESs hence the proof of the mixture formed is truly a deep eutectic solvent left a question mark among the readers as no concrete data for the drop in melting point is included in the research paper.

For the purpose of strengthening the claim that DES has lower

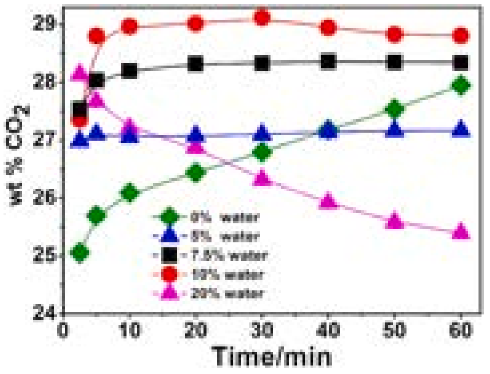


Fig. 12. Time-dependent CO_2 absorption process by [MEA.Cl][EDA]=1:3 with different water contents.

Fig. 13. CO_2 desorption from DESs via carbonic acid mechanism in the presence of 20 wt% water.

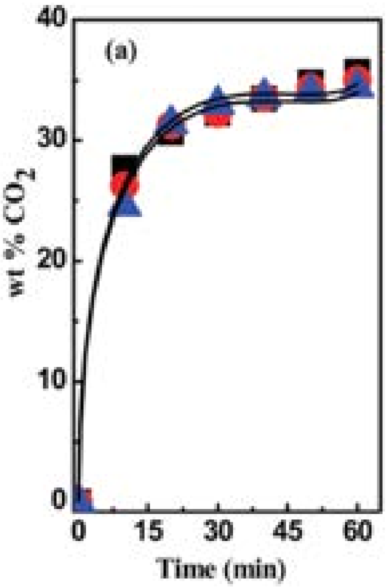


Fig. 14. CO_2 absorption kinetics in [MEA.Cl] [EDA] at 1:4 molar ratio + 10 wt % (black), 20 wt% (red) and 30 wt% (blue) water.

melting point as compared to its constituent, it is advisable for future studies to include the data for the melting point of the constituent and the mixture formed to give the idea that DES is truly formed as the result of the formation of hydrogen bonding. The date provided could proof the claim for future research not just for the purpose of carbon capture however also for other application such as metal processing, desulphurization, and pharmaceutical and medicinal purpose .

5.4. Classification of DESs based on its four categories

DESs is widely known to be separated into four categories which are Type I, II, III and IV. Despite this, most researchers don't include the categories of the DES synthesize in their studies. The limitations of their research make it difficult to compare the results of different studies. Additionally, it makes assessing the effectiveness of DES synthesizes in different contexts difficult. In particular, the categorization of the synthesized DES can help future scholars gain a greater understanding of how it can be used for purposes other than carbon dioxide separation in the future.

6. Conclusion

In this paper, various patents on the absorption of carbon dioxide via the use of green solvent which comprise of amine solvent, ionic liquid and deep eutectic solvent have been reviewed and discussed.

The use of solvents in the carbon capture process is significant, but there are interesting patents that address the problems associated with conventional solvents, particularly amine, which have serious drawbacks, requiring that a more suitable solvent be identified to replace it to avoid future issues, such as its high corrosiveness affecting its economic value. Additionally, ionic liquids, a greener solvent that has attracted researchers in the past decades, possess some of the issues making them hard to implement for carbon capture, the biggest being its high cost. Eventually, deep eutectic solvent was discovered and is considered a potential substitute for ionic liquid due to its ability to provide the benefits of ionic liquid while leaving behind the drawbacks. It has been demonstrated that amine-based deep eutectic solvents provide excellent CO_2 absorption.

Furthermore, several patents have been evaluated that address the possibility of using a third component, water for example to improve absorption performance. The observed result suggests that water plays a significant role in amine-based deep eutectic solvents, with the tertiary group exhibiting the greatest improvement among all groups.

CRediT authorship contribution statement

Muhammad Fizri Hazeem Ismail: Writing – review & editing, Writing – original draft, Conceptualization. **Asiah Nusaibah Masri:** Supervision, Conceptualization. **Norhana Mohd Rashid:** Funding acquisition. **Izni Mariah Ibrahim:** Conceptualization. **Sulafa Abdalmageed Saadaldeen Mohammed:** Visualization. **Wan Zaireen Nisa Yahya:** Resources.

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

The authors are unable or have chosen not to specify which data has been used.

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