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## Development and systematic evaluation of aqueous triazole chloride-based deep eutectic solvents for efficient CO<sub>2</sub> capture

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Deep eutectic solvents (DESs) have attracted considerable attention as promising alternatives to conventional solvents for mitigating CO<sub>2</sub> emissions due to their tunable structures, low volatility, and promising physicochemical properties. In this work, a series of [Triz]Cl/amine DESs were designed and synthesized and then formulated as 30 wt% aqueous solutions (30 wt% DES + 70 wt% H<sub>2</sub>O) to systematically investigate how the type of hydrogen bond donor (HBD) affects their physicochemical properties, thermal stability, and CO<sub>2</sub> capture performance, and to identify the most effective solvent; their CO<sub>2</sub> absorption capacity, absorption rate, thermal stability, and desorption efficiency were determined experimentally, and a novel stepwise evaluation strategy was employed for identification. [Triz]Cl/DETA was identified, exhibiting significantly enhanced performance, with CO<sub>2</sub> absorption capacity, absorption rate, thermal stability, and cyclic loading increased by 34%, 12%, 114%, and 39%, respectively, when compared with the conventional monoethanolamine (MEA). Its viscosity (both before and after CO<sub>2</sub> absorption), oxidative stability, and corrosion resistance were further studied, confirming the superior performance, and the reaction mechanism was also elucidated. This work provides valuable insights into the structure–property relationships of DESs and establishes [Triz]Cl/DETA-based solvents as promising candidates for efficient and sustainable CO<sub>2</sub> capture applications.

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### Green foundation

1. We performed structure–function analysis for HBD in aqueous [TrizCl]-based DESs and identified [Triz]Cl/DETA that outperforms MEA while lowering volatility and corrosion, yielding design rules (optimization of amine density, flexibility, and H-bonding).
2. 30 wt% [Triz]Cl/DETA delivers 0.17 g-CO<sub>2</sub> per g-solvent,  $K_a = 0.183 \text{ min}^{-1}$ , a desorption efficiency of 57% at 110 °C, and a cyclic loading of 0.09 g-CO<sub>2</sub> per g-solvent; the post-loading viscosity is  $\sim 7.5 \text{ mPa s}$ . The corrosion rates are markedly lower than those of 30 wt% MEA (unloaded:  $1.2 \times 10^{-4}$  vs.  $2.3 \times 10^{-4} \text{ mm a}^{-1}$ ; loaded:  $7.5 \times 10^{-4}$  vs.  $1.8 \times 10^{-3} \text{ mm a}^{-1}$ ). No FTIR-detectable oxidation was observed after 5 h of O<sub>2</sub>.
3. Regeneration temperature/energy (target  $\leq 100 \text{ °C}$ ) *via* composition tuning is reduced; durability (extension of O<sub>2</sub> exposure beyond 5 h) is verified; and corrosion is optimized by optimizing the DES–water ratio and materials selection, while quantifying solvent loss relative to that of MEA.

## 1. Introduction

The combustion of fossil fuels has driven industrial modernization but has also produced unprecedented anthropogenic

CO<sub>2</sub> emissions.<sup>1</sup> The concentration of CO<sub>2</sub> in the atmosphere has continued to rise and reached 422.5 ppm in 2024,<sup>2</sup> the first year on record with a global average temperature about 1.5 °C above pre-industrial levels.<sup>3</sup> It has been observed that the global per capita CO<sub>2</sub> emissions and the global mean temperature have risen substantially since the 19th century, demonstrating a clear positive correlation. Therefore, CO<sub>2</sub> mitigation and removal have become critical scientific and technological concerns for achieving global climate goals and maintaining climate stability.<sup>4</sup>

To mitigate these challenges, carbon capture, utilization, and storage (CCUS)<sup>5</sup> has been considered indispensable, owing to its application in large-scale industrial operation. Within the CCUS chain, CO<sub>2</sub> capture serves as a critical com-

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ponent, making the development of efficient CO<sub>2</sub> capture paramount. Among the available technologies for CO<sub>2</sub> capture, chemical-based absorption is widely regarded as the most promising one, due to its high selectivity, large absorption capacity, technological maturity, and flexibility for integration with existing industrial systems. For the chemical absorption routes, amine-based solvents, such as monoethanolamine (MEA), diethanolamine (DEA), methyl diethanolamine (MDEA), and their blends, are the most widely employed in research and industrial practice.<sup>5</sup> Among the studied amines, primary and secondary amines (*e.g.*, MEA and DEA) provide high absorption capacities and fast CO<sub>2</sub> capture, but their use is often constrained by volatility, corrosivity, and high regeneration energy demand; tertiary amines (*e.g.*, MDEA) exhibit lower volatility and reduced corrosivity, but their inherently low CO<sub>2</sub> absorption rates remain a significant drawback.<sup>6</sup> Hence, the development of alternative absorbents remains imperative to overcome the limitations of conventional systems.

Ionic liquids (ILs) exhibit exceptional properties, including high thermal stability, negligible volatility, and tunable structures, making them highly adaptable for diverse applications.<sup>7</sup> Nevertheless, the high production cost of conventional ILs remains a critical barrier to their large-scale deployment. Deep eutectic solvents (DESs) are a distinct class of IL analogues composed of a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA).<sup>8</sup> DESs not only retain many of the advantageous properties of ILs but also offer additional benefits, such as low cost and biocompatibility. Development of DESs has become more prominent than that of conventional ILs in recent years.

To develop DESs as absorbents for CO<sub>2</sub> capture, recent studies have highlighted that the choice of HBD is decisive.<sup>9</sup> For instance, Jiang *et al.*<sup>10</sup> showed that employing acylamido compounds as HBDs led to notably high capacities, and 1,5-diazabicyclo[4.3.0]non-5-ene (DBN)/2-imidazolidone (EU) (2 : 1) reached 0.23 g-CO<sub>2</sub> per g-solvent at 45 °C and 1 bar, attributed to multiple N-site interactions that enhance chemisorption. Likewise, within a chloride-amine family using a fixed HBA ([MEA]Cl), replacing the HBD from MEA to ethylenediamine (EDA) markedly increased the capacity, where [MEA]Cl/EDA achieved ~0.30 g-CO<sub>2</sub> per g-solvent under ambient pressure.<sup>11</sup> With choline chloride as the HBA, switching the HBD from urea to ethylene glycol reduces viscosity but also weakens chemisorption strength and decreases equilibrium loading, revealing a rate-capacity trade-off controlled by the HBD identity.<sup>12</sup> Similarly, the superbases 1,1,3,3-tetramethylguanidine (TMG) was used as the HBA and paired with polyol HBDs (*e.g.*, glycerol and ethylene glycol) to tune both the absorption kinetics and the energy required for desorption *via* shifting the dominant binding pathway (from carbamate to bicarbonate under humid conditions).<sup>13</sup>

Despite growing evidence highlighting the importance of the HBD component for the performance of DESs, previous research<sup>14</sup> has encountered three primary challenges. First, the formulation space was only partially investigated by focusing on EDA as the HBD and varying the types of HBAs, thus

leaving the vast spectrum of potential HBDs largely unexplored. Second, the EDA-based DESs demonstrated only moderate thermal stability, surpassing that of MEA, but still inadequate at relatively high temperatures and with extended cycling. Third, crucial durability indicators relevant to practical applications, such as oxidative stability, corrosion resistance, and cyclic stability, have not been systematically evaluated or sufficiently considered.

Additionally, for the DESs with outstanding CO<sub>2</sub> capture capacities, it is common for a sharp viscosity rise to occur following the CO<sub>2</sub> absorption, which has frequently been overlooked.<sup>15,16</sup> For example, neat [MEA]Cl/MDEA shows a multi-fold viscosity increase (from 275 to 1500 mPa s) upon CO<sub>2</sub> loading, which severely suppresses mass transfer and lowers the absorption rate.<sup>17</sup> To address this issue, water, an inexpensive and environmentally benign cosolvent, is highly effective at adjusting viscosity. For example, a 40 wt% [MEA]Cl/EDA + 60 wt% H<sub>2</sub>O solution maintained a low viscosity both before and after CO<sub>2</sub> absorption (4.4 and 13.3 mPa s at 25 °C, respectively).<sup>18</sup> Similarly, a [Ch]Cl/MEA solution containing 75 vol% H<sub>2</sub>O exhibited a high CO<sub>2</sub> absorption capacity of 0.19 g-CO<sub>2</sub> per g-solvent and low viscosity (2.2 mPa s) at 20 °C and 1.5 MPa.<sup>19</sup>

To address the research gaps and given prior evidence that 1,2,4-triazolium chloride ([Triz]Cl) demonstrates favorable CO<sub>2</sub> absorption/desorption efficiency and thermal stability, in this work, a series of [Triz]Cl-based DESs was designed and synthesized by pairing [Triz]Cl with structurally varied amine HBDs and then mixing with water as a cosolvent to form aqueous solutions. A stepwise method was used to identify promising DESs based on the CO<sub>2</sub> absorption capacity, absorption rate, thermal stability, and cyclic loading under identical conditions. The most effective DES was subsequently subjected to application-relevant evaluations, including viscosity measurements before and after CO<sub>2</sub> absorption, oxidative stability testing, corrosion compatibility assessment, and mechanistic analysis of the CO<sub>2</sub> capture. Through this approach, a practical design and screening strategy was developed to identify DESs as absorbents for CO<sub>2</sub> capture, and guidance directly relevant to industrial CO<sub>2</sub> capture deployment was provided.

## 2. Experimental

### 2.1. Materials

CO<sub>2</sub> (99.99%), 1,2,4-triazole (Triz, Macklin, 99%, CAS no. 288-88-0), hydrochloric acid (HCl, 37%, Macklin, CAS no. 7647-01-0), ethylenediamine (EDA, Lingfeng, 99.5%, CAS no. 107-15-3), diethylenetriamine (DETA, Macklin, 99%, CAS no. 111-40-0), tetraethylenepentamine (TEPA, Macklin, 95%, CAS no. 112-57-2), *N*-methyldiethanolamine (MDEA, Macklin, 98%, CAS no. 105-59-5), diethanolamine (DEA, Macklin, 99%, CAS no. 111-42-2), monoethanolamine (MEA, Macklin, 99%, CAS no. 141-43-5), 2-amino-2-methyl-propanol (AMP, Sigma-Aldrich, >95%, CAS no. 124-68-5) and aminoethylethanolamine (AEEA, Macklin, 99%, CAS no. 111-41-1) were used in this study. The structures of all studied HBDs are shown in Table 1.



**Table 1** Molecular characteristics and amine-group density of the selected HBDs

HBD	Molar mass (g mol <sup>-1</sup> )	Molecular structure	Number of amine groups	Molar amine density (mol g <sup>-1</sup> )
Ethylenediamine (EDA)	60.10		2	0.033
Diethylenetriamine (DETA)	103.17		3	0.029
Tetraethylenepentamine (TEPA)	189.3		5	0.026
2-Amino-2-methyl-propanol (AMP)	89.14		1	0.011
Aminoethylethanolamine (AEEA)	104.15		2	0.019
Monoethanolamine (MEA)	61.08		1	0.016
Diethanolamine (DEA)	105.14		1	0.010
N-Methyldiethanolamine (MDEA)	119.16		1	0.008

## 2.2. Preparation of DESs

The HBA, [Triz]Cl, was synthesized *via* a one-step anion-exchange method. During the synthesis, equimolar HCl was added dropwise into a round-bottom flask containing an aqueous 1,2,4-triazole solution, under stirring in an ice bath (1 °C) to minimize HCl volatilization. Afterward, the mixture was stirred at room temperature for 3 hours, followed by rotary evaporation at 60 °C and vacuum drying at the same temperature for 3 hours to remove excess water. To eliminate the remaining impurities, the sample was then connected to an ultra-high vacuum pump at 70 °C, yielding white crystalline [Triz]Cl. The DES, for example, [Triz]Cl/DETA, was prepared by mixing [Triz]Cl with DETA at a molar ratio of 1 : 5, followed by stirring at 60 °C for 3–5 hours to obtain a clear and homogeneous solvent. Subsequently, [Triz]Cl/DETA was diluted with H<sub>2</sub>O at a mass ratio of 3 : 7 to produce the aqueous [Triz]Cl/DETA solution for CO<sub>2</sub> capture. Other aqueous DES solutions were prepared following the same procedure.

## 2.3. Characterization

<sup>13</sup>C NMR spectra were measured using a 60 MHz NMR spectrometer (Spinsolve 60). Fourier Transform Infrared (FTIR, Nicolet 8700) spectroscopy was employed to identify functional groups of samples in the range of 400–4000 cm<sup>-1</sup> with a resolution of 4 cm<sup>-1</sup>. Thermal stability was evaluated *via* thermogravimetric analysis (TGA) using a TA SDT650 instrument, with DES samples heated from room temperature to 400 °C at a rate of 10 °C min<sup>-1</sup> under an N<sub>2</sub> flow of 60 mL min<sup>-1</sup>. Viscosity measurements of the solvents before and after CO<sub>2</sub> absorption were performed at 25 °C using an Anton Paar 2000 ME viscometer (accuracy ±0.35%). Oxidative stability was evaluated by comparing FTIR spectra recorded before and after 5 h of exposure to O<sub>2</sub> and N<sub>2</sub>.

## 2.4. CO<sub>2</sub> absorption capacity and rate

The CO<sub>2</sub> absorption capacity was measured using a gravimetric method,<sup>18</sup> following the procedure described in our previous work, as shown in Fig. S1.<sup>14</sup> Briefly, about 7 g of solvent was placed in a 15 mL gas absorption tube, which was immersed in a water bath maintained at a constant temperature. CO<sub>2</sub> gas was then bubbled into the solvent under atmospheric pressure at a flow rate of 60 mL min<sup>-1</sup>, controlled by a mass flow controller (YJ-700C). The entire gas absorption tube was weighed at regular intervals using an electronic analytical balance with an accuracy of ±0.1 mg. Absorption equilibrium was reached when there was no further increase in mass. Each experiment was repeated at least twice, and the average value was reported with an uncertainty of ±0.01 g-CO<sub>2</sub> per g-solvent. Before conducting experiments on the systems studied in this work, a 30 wt% MEA aqueous solution was investigated to ensure experimental reliability and accuracy, and a 30 wt% [Triz]Cl/EDA aqueous solution was also tested as a representative DES. Details of the uncertainty analysis and the corresponding raw data are provided in Fig. S2 of the SI.

The kinetics of the CO<sub>2</sub> capture process were analyzed, where the instantaneous CO<sub>2</sub> absorption rate ( $r_t$ , g-CO<sub>2</sub> per (g-solvent min)) and the apparent absorption rate constant ( $K_a$ , min<sup>-1</sup>) were calculated based on formulas derived from previous studies,<sup>20</sup> as shown below:

$$r_t = \frac{dn_t}{dt} \quad (1)$$

$$n_t = n_e(1 - e^{-K_a t}) \quad (2)$$

where  $n_t$  is the CO<sub>2</sub> capacity (g-CO<sub>2</sub> per g-solvent) at time  $t$  and  $n_e$  is the saturated CO<sub>2</sub> capacity. The value of  $K_a$  was obtained by fitting the experimental data.



## 2.5. Solvent regeneration

The solvent regeneration process was conducted in a three-neck flask containing CO<sub>2</sub>-saturated DES, which was heated to 110 °C in an oil bath and continuously stirred at 150 rpm, as shown in Fig. S1. To recover water vapor released alongside CO<sub>2</sub>, the gas stream was passed through a serpentine condenser maintained at 1 °C, followed by concentrated sulfuric acid to remove residual moisture.<sup>21</sup> The desorption rate ( $r_d$ , g-CO<sub>2</sub> per (g-solvent min)) was obtained from eqn (3), and then the CO<sub>2</sub> cyclic loading ( $n_d$ ) was obtained from the integral of  $r_d$  over time as shown in eqn (4). The CO<sub>2</sub> desorption efficiency ( $\eta$ ) was calculated from the cyclic loading and the saturated CO<sub>2</sub> capacity  $n_e$  as expressed in eqn (5):

$$r_d = \frac{Q_t \times M_{\text{CO}_2}}{m_s \times 22.4 \times 1000} \left( \frac{P \times (T_0 + 273.15)}{P_0 \times (T + 273.15)} \right) \quad (3)$$

$$n_d = \int_0^t r_d dt \quad (4)$$

$$\eta = \frac{n_d}{n_e} \times 100\% \quad (5)$$

where  $Q_t$  is the CO<sub>2</sub> flow rate at the outlet of the round-bottom flask (m<sup>3</sup> min<sup>-1</sup>);  $M_{\text{CO}_2}$  is the molar mass of CO<sub>2</sub>;  $P$  and  $T$  are the experimental pressure and temperature (kPa and °C);  $P_0$  and  $T_0$  are the standard pressure (101.325 kPa) and standard temperature (0 °C), respectively; and  $m_s$  is the solvent mass.

## 2.6. Corrosion analysis

To study the corrosive effects of the absorbents on steel, a three-electrode electrochemical system was used.<sup>22</sup> The setup consists of a 20# carbon steel working electrode (W.E.), an Ag/AgCl (saturated KCl) reference electrode (R.E.), and a platinum counter electrode (C.E.). Before testing, the steel samples were

cleaned with acetone and ethanol. Electrochemical measurements were performed using a CHI760E Electrochemical Workstation. Each specimen was stabilized at the open-circuit potential (OCP). Tafel polarization was recorded by scanning from -100 to +100 mV *versus* OCP at a rate of 10 mV s<sup>-1</sup>. The corrosion current density ( $i_{\text{corr}}$ ) was obtained by Tafel extrapolation, and the corrosion rate (CR, mm a<sup>-1</sup>) was calculated as

$$\text{CR} = \frac{3.27 \times 10^{-3} \times i_{\text{corr}} \times w}{A \times \rho} \quad (6)$$

where  $w$  is the equivalent weight of 20# carbon steel (g equiv.<sup>-1</sup>);  $A$  is the exposed surface area (cm<sup>2</sup>); and  $\rho$  is the density of 20# carbon steel (g cm<sup>-3</sup>).

## 3. Results and discussion

### 3.1. Structural and functional features of HBDs

HBDs play a critical role in the CO<sub>2</sub> capture performance of DESs, with representative examples and their corresponding absorption capacities summarized in Table 2. Depending on their functional groups, HBDs can participate in different types of hydrogen-bonding and CO<sub>2</sub> interaction mechanisms. The most studied classes include acylamides, polyols, carboxylic acids, alkanolamines, and polyamines. Acylamido-based HBDs, such as urea, contain both carbonyl (C=O) and amino (-NH<sub>2</sub>) groups, which form strong hydrogen bonds with HBAs, significantly lowering the eutectic temperature.<sup>23</sup> Moreover, as shown in Fig. 1(a), their amine groups can chemically interact with CO<sub>2</sub> to form carbamate-like intermediates, enhancing CO<sub>2</sub> capture efficiency.<sup>10</sup> However, the relatively high volatility and limited chemical stability of some acyla-

**Table 2** CO<sub>2</sub> absorption performance of DESs based on different types of HBDs

HBD type	DESs (molar ratio)	HBD	$T$ (K)/ $P$ (bar)	Capacity (g-CO <sub>2</sub> per g-solvent)	Ref.
Acylamido compounds	[Ch]Cl/urea (1 : 2)	Urea	313.15/123.4	0.04	23
	DBN/DMU (2 : 1)	Dimethylolurea (DMU)	318.15/1.0	0.04	10
	DBN/DMLU (2 : 1)	1,3-Dimethylurea (DMLU)	318.15/1.0	0.17	
	DBN/EU (2 : 1)	2-Imidazolidone (EU)	318.15/1.0	0.23	
	DBN/EU (3 : 1)	2-Imidazolidone (EU)	318.15/1.0	0.19	
Polyols	[TETA]Cl/DEG (1 : 2)	Diethylene glycol (DEG)	313.15/1.0	0.16	25
	[TETA]Cl/EG (1 : 3)	Ethylene glycol (EG)	313.15/1.0	0.18	
	[N <sub>2222</sub> ]Triz/EG (1 : 2)	Ethylene glycol (EG)	298.15/1.0	0.13	26
	[N <sub>2222</sub> ]Im/EG (1 : 2)	Ethylene glycol (EG)	298.15/1.0	0.13	
Carboxylic acids	[Ch]Cl/Gly (1 : 1)	Glycerol (Gly)	298.2/1.0	0.02	31
	[BHDE]Cl/L (1 : 2)	Lactic acid (L)	298.15/2.8	0.001	27
	[BHDE]Cl/Ac (1 : 2)	Acetic acid (Ac)	298.15/2.1	0.003	
	[N <sub>4444</sub> ]Cl/DECA (1 : 2)	Decanoic acid (DECA)	308.15/0.9	0.002	28
	[N <sub>8881</sub> ]Cl/DECA (1 : 2)	Decanoic acid (DECA)	323.15/0.9	0.002	
	[N <sub>8888</sub> ]Cl/DECA (1 : 2)	Decanoic acid (DECA)	323.15/0.9	0.002	
Alcohol amine	[TEPA]Cl/MEA (1 : 2)	Monoethanolamine (MEA)	298.15/1.0	0.41	9
	[DEA]Cl/MDEA (1 : 3)	Methyldiethanolamine (MDEA)	298.15/1.0	0.11	17
	[MDEA]Cl/MDEA (1 : 3)	Methyldiethanolamine (MDEA)	298.15/1.0	0.06	
Polyamine	[MEA]Cl/EDA (1 : 4)	Ethylenediamine (EDA)	328.15/1.0	0.36	29
	[MEA]Cl/DETA (1 : 3)	Diethyltriamine (DETA)	313.15/1.0	0.18	30
	[MEA]Cl/TETA (1 : 3)	Triethylenetetramine (TETA)	313.15/1.0	0.17	
	[MEA]Cl/TEPA (1 : 3)	Tetraethylenepentamine (TETA)	313.15/1.0	0.10	



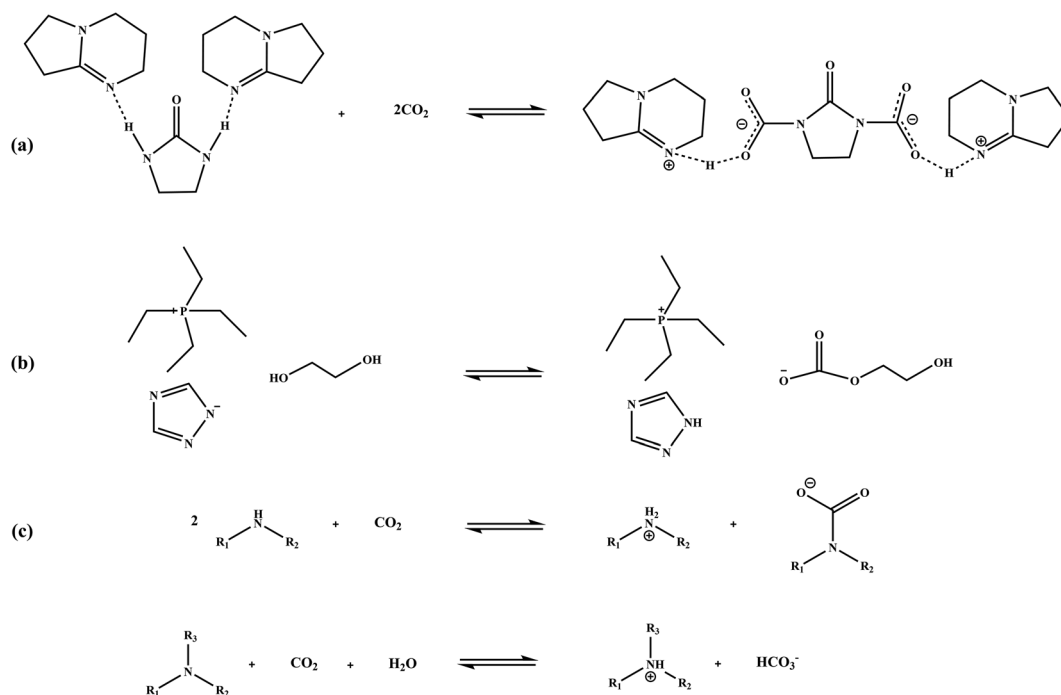


Fig. 1 Representative CO<sub>2</sub> capture pathways involving different HBD types in DESs: (a) acylamide, (b) polyol, and (c) amine.

mides under regeneration conditions constrain their large-scale applicability.

Polyol-based HBDs, such as ethylene glycol (EG) and glycerol, contain multiple hydroxyl (–OH) groups, which facilitate extensive hydrogen-bonding networks with HBAs, thereby promoting CO<sub>2</sub> absorption physically and chemically.<sup>24</sup> For example, when using EG and diethylene glycol (DG) as HBDs, and triethylenetetramine chloride ([TETA]Cl) as the HBA to prepare various DESs, it is found that EG or DG can activate the –NH– and –NH<sub>2</sub> groups in [TETA]Cl, thereby improving the basicity of the DESs and enhancing CO<sub>2</sub> capacity.<sup>25</sup> FTIR and NMR analyses have demonstrated that EG can chemically react with CO<sub>2</sub> to produce carbonate species, facilitated by activation of its hydroxyl oxygen through interaction with the cationic HBA (Fig. 1b).<sup>26</sup> In addition, polyol-based DESs exhibit high thermal stability and low volatility, which is beneficial for solvent regeneration and long-term stability.

Carboxylic acid-based HBDs can be categorized into hydrophilic and hydrophobic types, both primarily capturing CO<sub>2</sub> through physical absorption.<sup>27</sup> Hydrophilic acids (*e.g.*, lactic acid and formic acid) form dense hydrogen bonding networks *via* their –COOH groups, depressing melting points and increasing polarity to enhance CO<sub>2</sub> solubility. In contrast, hydrophobic acids (*e.g.*, dodecanoic acid) yield less polar DESs that demonstrate superior thermal stability and require lower energy for solvent regeneration.<sup>28</sup>

Amino alcohols and polyamines are among the most promising HBD families owing to their low cost, high reactivity toward CO<sub>2</sub>, and favorable physicochemical characteristics.<sup>17</sup> Amino alcohols possess both amine (–NH<sub>2</sub>) and hydroxyl

(–OH) groups, offering dual sites for hydrogen bonding and CO<sub>2</sub> activation,<sup>9</sup> while polyamines are rich in multiple nucleophilic amine sites, capable of forming strong hydrogen bonding networks with HBAs.<sup>29</sup> These features enable the formation of stable and dense hydrogen bonding networks with HBAs, thereby facilitating the self-assembly of low-melting-point DESs and providing abundant reactive sites for CO<sub>2</sub> interaction.<sup>30</sup> As depicted in Fig. 1(c), primary and secondary amines react directly with CO<sub>2</sub> to form carbamate species, while tertiary amines facilitate CO<sub>2</sub> hydration to produce bicarbonate ions in aqueous media. This chemical versatility enables amine-based DESs to achieve high absorption capacities and fast kinetics compared with other HBD types.

As summarized in Table 2, DESs incorporating amine-based HBDs consistently exhibit superior CO<sub>2</sub> absorption capacities. Therefore, in this work, amine-based HBDs were selected to systematically investigate the structure–property relationships. More specifically, EDA, DETA, TEPA, MEA, DEA, MDEA, AMP, and AEEA were chosen, which encompass a broad range of molecular architectures, from polyamines to alkanolamines and tertiary amines, enabling a systematic comparison of how the number of amine groups, steric hindrance, and hydroxyl substitution influence CO<sub>2</sub> absorption capacity, kinetics, and thermal stability.

### 3.2. Absorption performance of aqueous DESs

Based on the findings in section 3.1, eight amines with potentially superior CO<sub>2</sub> absorption capacities were selected as the HBDs for this study. For a DES solution, the ratio of HBA to HBD and the content of water will also influence the pro-



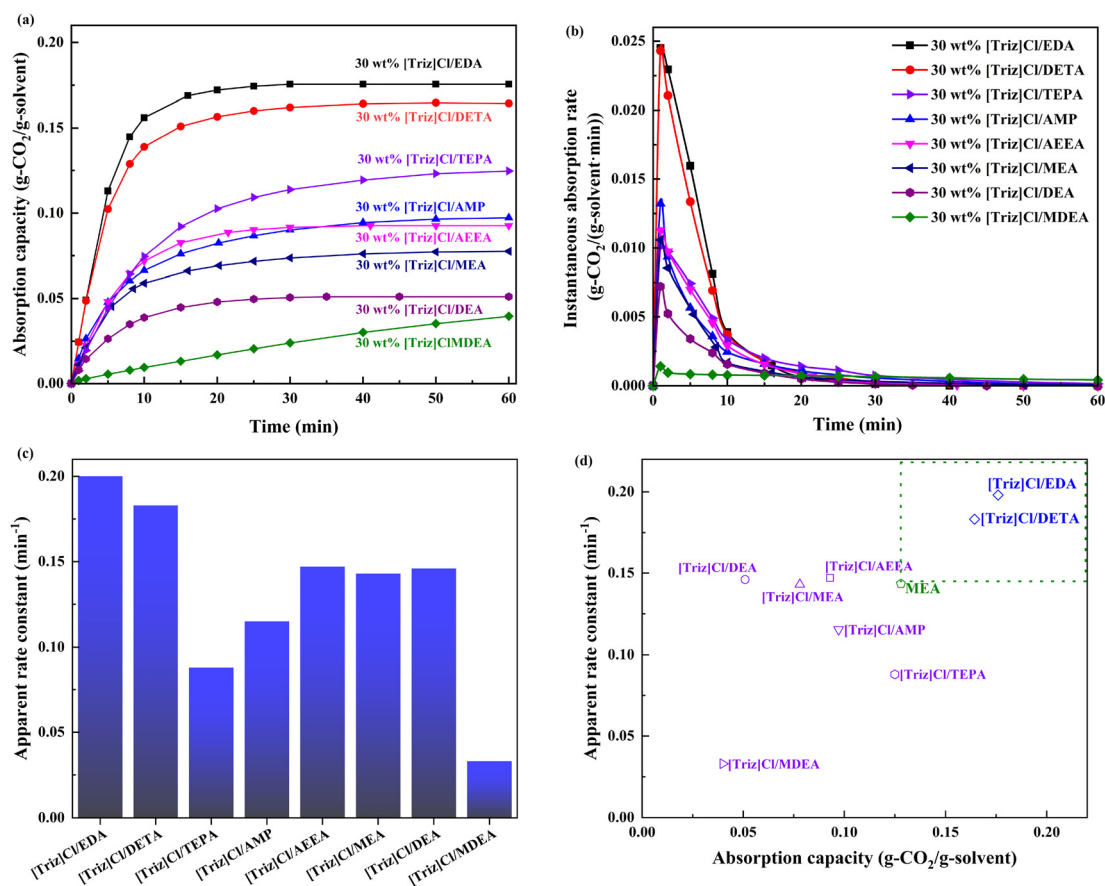
properties and performance. Previously, Shukla *et al.*<sup>32</sup> systematically investigated the properties of [Im]Cl/EDA at various HBA:HBD molar ratios (1:4, 1:5, and 1:6). The results showed that although [Im]Cl/EDA (1:5) exhibited a slightly lower CO<sub>2</sub> absorption capacity than [Im]Cl/EDA (1:6), its thermal degradation was significantly lower than that of the 1:4 and 1:6 counterparts. Additionally, the viscosity of the 30 wt% [Im]Cl/EDA (1:5) aqueous solution was found to be below 5 mPa s. Therefore, a 30 wt% DES aqueous solution

with an HBA:HBD molar ratio of 1:5 was prepared for all the studied DESs in this work to reflect practical application scenarios.

**3.2.1. Absorption capacity.** To elucidate the effect of HBD on the CO<sub>2</sub> absorption capacity, dynamic CO<sub>2</sub> absorption curves were measured over time. The results are shown in Table 3. For the DESs based on polyamine HBDs, aqueous [Triz]Cl/EDA, [Triz]Cl/DETA, and [Triz]Cl/TEPA solutions achieved the top 3 capacities of 0.18, 0.17, and 0.13 g-CO<sub>2</sub> per g-solvent, respectively, as shown in Fig. 2(a). For the alkanolamine-based DESs, [Triz]Cl/AMP (0.10 g-CO<sub>2</sub> per g-solvent) and [Triz]Cl/AEEA (0.09 g-CO<sub>2</sub> per g-solvent) exhibited relatively higher CO<sub>2</sub> absorption capacities than [Triz]Cl/MEA (0.08 g-CO<sub>2</sub> per g-solvent), [Triz]Cl/DEA (0.05 g-CO<sub>2</sub> per g-solvent), and [Triz]Cl/MDEA (0.04 g-CO<sub>2</sub> per g-solvent). This order corresponds to their molar amine group densities (except for [Triz]Cl/AMP), as shown in Table 3 and Fig. S3(a), indicating that higher amine group densities provide more reactive sites for CO<sub>2</sub> capture, thereby enhancing the absorption performance of DESs, such as [Triz]Cl/EDA. However, the relatively high CO<sub>2</sub> absorption capacity of [Triz]Cl/AMP, despite its low amine group density, may be attributed to the unique structure of AMP, where the structural synergy and

**Table 3** CO<sub>2</sub> absorption data of 30 wt% DES aqueous solutions

DESs	CO <sub>2</sub> absorption capacity (g-CO <sub>2</sub> per g-solvent)	Instantaneous rate (g-CO <sub>2</sub> per (g-solvent min))	Apparent rate constant $K_a$ (min <sup>-1</sup> )
[Triz]Cl/EDA	0.18	0.025	0.20
[Triz]Cl/DETA	0.17	0.024	0.18
[Triz]Cl/AMP	0.10	0.013	0.12
[Triz]Cl/AEEA	0.09	0.011	0.15
[Triz]Cl/MDEA	0.04	0.001	0.03
[Triz]Cl/MEA	0.08	0.011	0.14
[Triz]Cl/TEPA	0.13	0.010	0.09
[Triz]Cl/DEA	0.05	0.007	0.15



**Fig. 2** (a) CO<sub>2</sub> absorption curves, (b) instantaneous CO<sub>2</sub> absorption rate curves, (c) apparent rate constant, and (d) systematic comparison of CO<sub>2</sub> absorption capacity and apparent rate constant of aqueous DES solutions at 22 °C and 1 bar.



possible weak interactions between the hydroxyl group and CO<sub>2</sub> together enhance CO<sub>2</sub> solubility.<sup>33</sup> Moreover, the DESs incorporating polyamine-based HBDs exhibited significantly higher CO<sub>2</sub> absorption capacities compared to those with alkanolamine-based HBDs. Notably, [Triz]Cl/MEA exhibited a higher CO<sub>2</sub> absorption capacity than [Triz]Cl/DEA, indicating that primary amines possess stronger CO<sub>2</sub> binding ability than secondary amines. [Triz]Cl/MDEA showed the lowest capacity, consistent with the typical trend of primary > secondary > tertiary amines.<sup>34</sup>

**3.2.2. Absorption kinetics.** The instantaneous CO<sub>2</sub> absorption rates ( $r_t$ ) of DESs with different HBDs, alongside the corresponding apparent absorption rate constants ( $K_a$ ), were calculated by fitting the experimental data using eqn (1) and (2), with the results summarized in Table 3. As shown in Fig. 2(b), all DESs exhibited a characteristic two-stage absorption profile. In the initial 1–5 minutes, the instantaneous absorption rate increased rapidly due to the presence of abundant free active sites. Subsequently, the rate declined gradually, as saturation and diffusion limitations became dominant, requiring CO<sub>2</sub> molecules to diffuse deeper into the DES matrix to access the remaining reactive sites. Among all the DESs, [Triz]Cl/EDA and [Triz]Cl/DETA demonstrated the highest instantaneous CO<sub>2</sub> absorption rates (~0.025 and 0.024 g-CO<sub>2</sub> per (g-solvent min)), indicating rapid early-stage chemisorption. These were followed, in descending order, by [Triz]Cl/AMP, [Triz]Cl/AEEA, [Triz]Cl/MEA, [Triz]Cl/TEPA, [Triz]Cl/DEA, and [Triz]Cl/MDEA. However, it is noteworthy that the ranking of apparent rate constants does not fully match that of the instantaneous absorption rates, as shown in Fig. 2(c). For instance, [Triz]Cl/TEPA has a lower apparent rate constant than [Triz]Cl/DEA, but a higher instantaneous absorption rate. This discrepancy is expected, as the former reflects early stage chemisorption kinetics, while the latter accounts for the overall absorption process, including mass transfer and diffusion limitations.<sup>35</sup> Therefore, both parameters are complementary and should be considered jointly when evaluating DES performance for CO<sub>2</sub> capture.

To comprehensively compare the CO<sub>2</sub> absorption capacity and apparent rate constant of aqueous DESs with different HBDs, as well as to compare their performance relative to conventional MEA, the absorption performance data of all tested DESs are summarized in Fig. 2(d). The data demonstrate that [Triz]Cl/EDA and [Triz]Cl/DETA exhibit significantly superior performance in both CO<sub>2</sub> absorption capacity and apparent absorption rate constant compared to the other DESs and MEA. This may be attributed to their higher amine density and the lower steric hindrance. These findings highlight the critical role of HBD selection in optimizing CO<sub>2</sub> capture performance.

### 3.3. Thermal stability of DESs

As illustrated in Fig. 3, the thermal stability of [Triz]Cl-based DESs (neat, water-free samples) was evaluated using thermogravimetric analysis (TGA) and differential thermogravimetry (DTG). The onset temperature ( $T_{\text{onset}}$ ) and the peak temperature ( $T_{\text{peak}}$ ) were used to quantitatively evaluate the thermal stability of the DESs.  $T_{\text{onset}}$  is defined as the temperature at which the weight baseline intersects the tangent of the TGA curve, corresponding to the beginning of thermal decomposition, as shown in Fig. S4. In contrast,  $T_{\text{peak}}$  represents the temperature at which the sample exhibits the maximum decomposition rate and corresponds to the peak in the DTG curve.

The results show that all DESs exhibit higher thermal stability than pure MEA (Table 4), confirming that DES formation enhances solvent stability. Specifically, MEA exhibited the lowest stability, with a  $T_{\text{onset}}$  of 83 °C and a  $T_{\text{peak}}$  of only 110 °C. Among the samples, [Triz]Cl/TEPA demonstrated the highest  $T_{\text{onset}}$  (261 °C) and  $T_{\text{peak}}$  (295 °C), alongside minimal weight loss (0.7% at 110 °C). This superior stability is attributed to the high molecular complexity of TEPA, which facilitates the formation of extensive hydrogen-bonding networks with the HBA, thereby enhancing the structural integrity of the DES. In contrast, [Triz]Cl/EDA displayed the lowest thermal stability among the polyamine-based DESs, with a  $T_{\text{onset}}$  of only 90 °C and a substantial weight loss of 23%. This stark difference is primarily due to the low molecular weight and

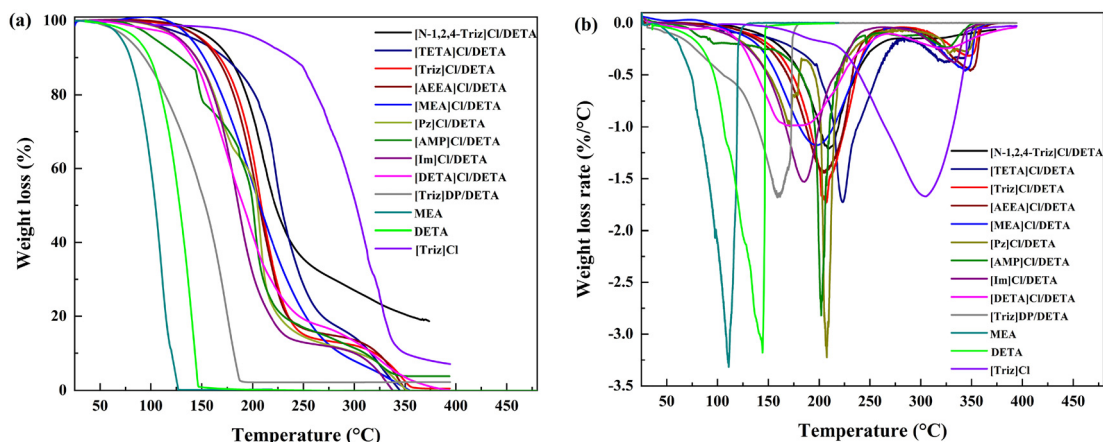


Fig. 3 (a) TGA and (b) DTG curves of pure DESs with different HBDs, compared with MEA, DETA, and [Triz]Cl.



**Table 4** Thermal stability of pure [Triz]Cl-based DESs, MEA, DETA, and [Triz]Cl

DES	$T_{\text{onset}}$ (°C)	$T_{\text{peak}}$ (°C)	Weight loss at 110 °C (%)
[Triz]Cl/EDA	90	109	23.0
[Triz]Cl/DETA	178	207	0.7
[Triz]Cl/TEPA	261	295	0.7
[Triz]Cl/MEA	155	186	1.6
[Triz]Cl/DEA	172	259	1.1
[Triz]Cl/MDEA	199	247	0.7
[Triz]Cl/AEEA	224	247	0.2
[Triz]Cl/AMP	160	171	5.7
MEA	83	110	40.0
DETA	101	141	13.5
[Triz]Cl	255	304	0.1

boiling point of EDA, resulting in high volatility and increased susceptibility to thermal degradation. [Triz]Cl/DETA, on the other hand, exhibited intermediate stability, with a  $T_{\text{onset}}$  of 178 °C, a  $T_{\text{peak}}$  of 207 °C, and negligible weight loss (0.7%), reflecting a balance between molecular complexity and volatility. [Triz]Cl/MDEA and [Triz]Cl/AEEA exhibited relatively high  $T_{\text{onset}}$  values of 199 °C and 224 °C, respectively, along with low weight losses (0.7% and 0.2%), indicating favorable thermal resilience. In contrast, [Triz]Cl/MEA and [Triz]Cl/AMP showed relatively low  $T_{\text{onset}}$  temperatures (155 °C and 160 °C, respectively) and higher weight losses (1.6% and 5.7%).

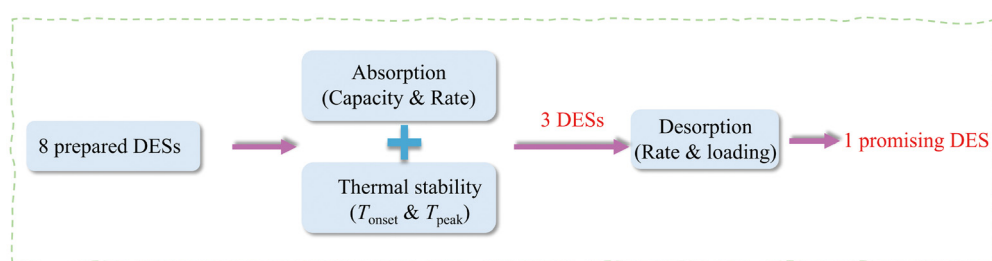
Overall, the thermal stability of DESs followed the trend: [Triz]Cl/TEPA > [Triz]Cl/AEEA > [Triz]Cl/MDEA > [Triz]Cl/DETA > [Triz]Cl/DEA > [Triz]Cl/AMP > [Triz]Cl/MEA > [Triz]Cl/EDA > MEA. These findings highlight that the thermal stability of DESs is predominantly governed by the physical properties and structural characteristics of the HBD, as well as the strength of hydrogen bonding between the HBD and HBA. For further analysis, Fig. S3(b) shows that the  $T_{\text{onset}}$  and  $T_{\text{peak}}$  of the DESs increase with the boiling point of the HBD. DESs based on higher-boiling, higher-molecular-weight HBDs (e.g., TEPA, AEEA, and MDEA) are more thermally stable than those using the lower-boiling HBDs (e.g., MEA and EDA). This suggests that stronger hydrogen-bonding networks and less volatile HBDs enhance the intrinsic thermal stability of the DESs.

### 3.4. First-step screening and desorption performance of aqueous DESs

As illustrated in Fig. 2(d), 30 wt% [Triz]Cl/EDA and [Triz]Cl/DETA demonstrate significantly superior CO<sub>2</sub> capture perform-

ance, exhibiting higher absorption capacities (0.18–0.17 g-CO<sub>2</sub> per g-solvent) and faster absorption rate constants (0.20–0.18 min<sup>-1</sup>) compared to other 30 wt% DES aqueous solutions (0.05–0.13 g-CO<sub>2</sub> per g-solvent; 0.03–0.15 min<sup>-1</sup>) and 30 wt% MEA (0.12 g-CO<sub>2</sub> per g-solvent; 0.16 min<sup>-1</sup>). These distinctive advantages have qualified 30 wt% [Triz]Cl/EDA and [Triz]Cl/DETA as primary candidates for subsequent investigations. Meanwhile, thermal stability analysis revealed that pure [Triz]Cl/TEPA and [Triz]Cl/AEEA possess exceptional thermal resistance with high  $T_{\text{onset}}$  values of 261 °C and 224 °C, respectively, ranking highest among all the evaluated DESs. However, [Triz]Cl/TEPA exhibited suboptimal absorption kinetics (0.09 min<sup>-1</sup>), rendering it industrially impractical. In contrast, although [Triz]Cl/AEEA exhibited a lower CO<sub>2</sub> absorption capacity and rate constant (0.09 g-CO<sub>2</sub> per g-solvent and 0.15 min<sup>-1</sup>) than MEA, its outstanding thermal stability ( $T_{\text{onset}}$  = 224 °C, 0.2 wt% weight loss at 110 °C) made it a more promising candidate. Therefore, [Triz]Cl/AEEA was selected as the third potential absorbent, alongside [Triz]Cl/EDA and [Triz]Cl/DETA, for further study (Fig. 4).

As a key step in the absorption–desorption cycle, CO<sub>2</sub> desorption is equally vital as absorption, as it governs the overall energy efficiency and long-term sustainability of the process. In this work, desorption at 110 °C was identified as the optimal condition, providing a favorable balance between the amount of CO<sub>2</sub> desorbed and the desorption rate, as shown in Fig. S5. To this end, the desorption performance of the CO<sub>2</sub>-saturated aqueous DES solutions was evaluated by measuring the time-dependent CO<sub>2</sub> desorption rate, desorption efficiency, and cyclic loading. As shown in Fig. 5(a), the peak desorption rates of [Triz]Cl/DETA, [Triz]Cl/EDA, [Triz]Cl/AEEA, and MEA are 0.011, 0.013, 0.009, and 0.009 g-CO<sub>2</sub> per (g-solvent min), respectively. In comparison, [Triz]Cl/EDA and [Triz]Cl/DETA exhibited desorption rates 44% and 22% higher than that of MEA, respectively, whereas [Triz]Cl/AEEA showed a similar desorption rate to MEA. Furthermore, as shown in Fig. 5(b), the desorption efficiencies of [Triz]Cl/DETA, [Triz]Cl/EDA, [Triz]Cl/AEEA, and MEA are 57%, 46%, 56%, and 57%, respectively. Correspondingly, their CO<sub>2</sub> cyclic loadings, presented in Fig. 5(c), are 0.09, 0.08, 0.05, and 0.07 g-CO<sub>2</sub> per g-solvent, respectively. As a result, [Triz]Cl/DETA demonstrated both the highest cyclic loading and the highest desorption efficiency, highlighting its potential as a highly regenerable absorbent. Notably, although [Triz]Cl/EDA exhibited a higher cyclic

**Fig. 4** Stepwise methodology adopted for the screening of DESs.

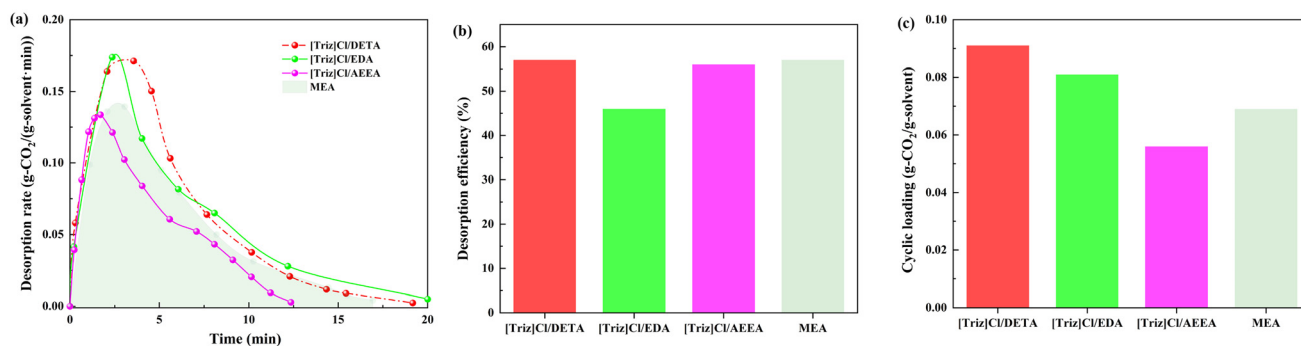


Fig. 5 (a) CO<sub>2</sub> desorption curves, (b) desorption efficiencies, and (c) CO<sub>2</sub> cyclic loading of aqueous DESs and MEA.

loading than MEA, it showed the lowest desorption efficiency. This may be attributed to the stronger CO<sub>2</sub> binding affinity of EDA, which impedes complete desorption under moderate thermal conditions.

### 3.5. Global comparison and second-step screening

Based on a comprehensive evaluation of key performance metrics, including CO<sub>2</sub> absorption capacity, apparent rate constant, thermal stability, desorption efficiency, and cyclic loading, as shown in Table 5, the 30 wt% [Triz]Cl/DETA aqueous solution was identified as the most promising absorbent. It exhibits high absorption performance ( $n_t = 0.17$  g-CO<sub>2</sub> per g-solvent,  $K_a = 0.18$  min<sup>-1</sup>), outstanding thermal stability ( $T_{onset} = 178$  °C with only 0.7% weight loss at 110 °C), and the highest cyclic CO<sub>2</sub> loading (0.09 g-CO<sub>2</sub> per g-solvent), while maintaining a desorption efficiency comparable to that of MEA.

To further integrate and compare the multiple performance indicators of DESs, a radar chart was constructed using normalized data, as shown in Fig. 6. All performance metrics were normalized to a 0–1 range using Min–Max scaling methods (eqn (S1) in the SI), with weight loss converted to residual mass. This comprehensive visualization enables a direct comparison of overall performance across different systems. Based on this analysis, the 30 wt% [Triz]Cl/DETA aqueous solution was also identified as the most promising absorbent (Fig. 4).

### 3.6. Further research on [TrizCl][DETA]

**3.6.1. Viscosity before and after CO<sub>2</sub> absorption.** The viscosity of the 30 wt% [Triz]Cl/DETA aqueous solution was measured before and after CO<sub>2</sub> absorption and compared with that of 30 wt% MEA aqueous solution, as shown in Fig. 7.

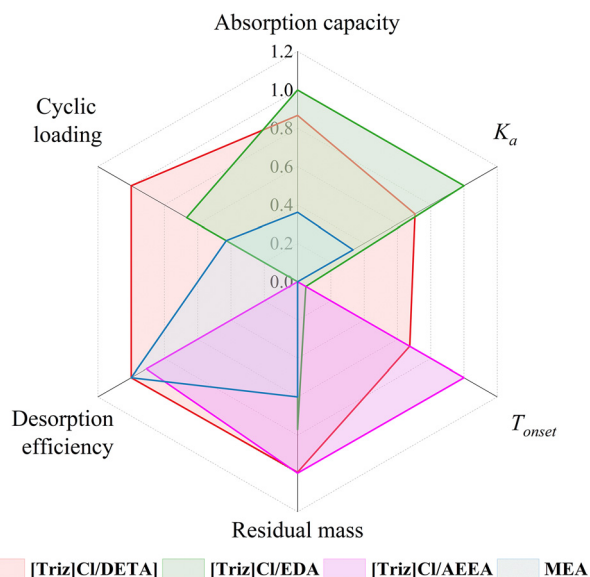


Fig. 6 Comprehensive performance comparison of CO<sub>2</sub> absorbents.

Upon CO<sub>2</sub> absorption, the viscosity of [Triz]Cl/DETA increased due to the formation of carbamate species, which enhanced hydrogen bonding and ionic interactions and thus restricted molecular mobility. The effect was more pronounced than in MEA because the polyamine structure of [Triz]Cl/DETA provides multiple reactive sites that facilitate the formation of denser hydrogen-bonding networks. Nevertheless, the viscosity of the [Triz]Cl/DETA solution after CO<sub>2</sub> absorption remained below 7 mPa s, which is sufficiently low for CO<sub>2</sub> capture.

**3.6.2. Oxidative stability test.** To evaluate the oxidative stability of the aqueous DES under practical conditions, its

Table 5 Comprehensive performance comparison of DESs

Absorbent	Absorption capacity $n_t$ (g-CO <sub>2</sub> per g-solvent)	Apparent rate constant $K_a$ (min <sup>-1</sup> )	$T_{onset}$ (°C)	Weight loss at 110 °C (%)	Desorption efficiency $\eta$ (%)	Cyclic loading $n_c$ (g-CO <sub>2</sub> per g-solvent)
[Triz]Cl/DETA	0.17	0.18	178	0.7	57%	0.09
[Triz]Cl/EDA	0.18	0.20	90	23	46%	0.08
[Triz]Cl/AEEA	0.09	0.15	224	0.2	56%	0.05
MEA	0.12	0.16	83	40	57%	0.07



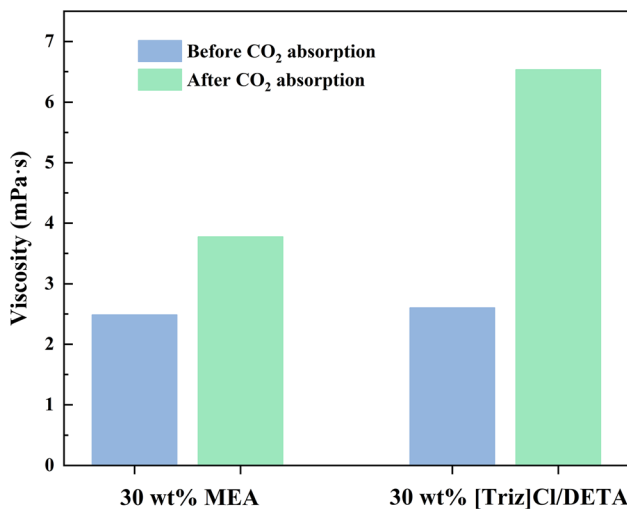


Fig. 7 Viscosity of 30 wt% [Triz]Cl/DETA and MEA aqueous solutions before and after CO<sub>2</sub> absorption.

potential reactivity toward common industrial gas components, N<sub>2</sub> and O<sub>2</sub>, was examined. The 30 wt% [Triz]Cl/DETA aqueous solution was maintained at 22 °C and continuously purged with either N<sub>2</sub> or O<sub>2</sub> for 5 hours; the samples were then analyzed by FTIR, and the spectra were compared with those of the fresh sample. As shown in Fig. 8, no noticeable variations were observed in the characteristic absorption bands after exposure to either gas, indicating that the chemical composition and molecular structure of the DES remained unchanged. Furthermore, to simulate desorption conditions, the solvent was exposed to O<sub>2</sub> at 110 °C for 5 hours, followed by FTIR analysis (Fig. 8). No new peaks or significant intensity

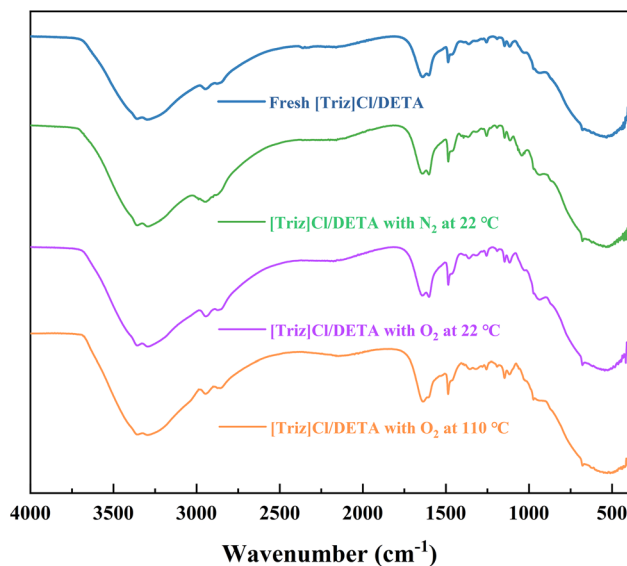


Fig. 8 FTIR spectra of 30 wt% [Triz]Cl/DETA aqueous solutions: fresh, after 5 h exposure to N<sub>2</sub> or O<sub>2</sub> at 22 °C, and after 5 h exposure to O<sub>2</sub> at 110 °C.

changes were detected, confirming that aqueous [Triz]Cl/DETA exhibits excellent oxidative stability even at 110 °C. These findings demonstrate the excellent oxidative stability of aqueous [Triz]Cl/DETA, suggesting its suitability for long-term gas treatment applications without oxidative degradation.

**3.6.3. Corrosion analysis.** Considering the critical influence of corrosion on the industrial applicability of CO<sub>2</sub> capture solvents, the corrosion behavior of 20# carbon steel was tested in both conventional 30 wt% MEA and the novel 30 wt% [Triz]Cl/DETA aqueous solutions at room temperature. The corrosion rates were obtained from the Tafel polarization measurements, as illustrated in Fig. 9(a). In the CO<sub>2</sub>-free state, the [Triz]Cl/DETA solution exhibited a substantially lower corrosion rate than MEA, measured at  $1.2 \times 10^{-4}$  and  $2.3 \times 10^{-4}$  mm a<sup>-1</sup>, respectively (Fig. 9(b)). Upon CO<sub>2</sub> loading, the corrosion rates of both systems increased; however, the rise was considerably less pronounced for [Triz]Cl/DETA. Specifically, its corrosion rate increased to  $7.5 \times 10^{-4}$  mm a<sup>-1</sup>, whereas that of MEA reached  $1.8 \times 10^{-3}$  mm a<sup>-1</sup>. These results clearly indicate that the aqueous [Triz]Cl/DETA system possesses higher corrosion resistance and is significantly less susceptible to CO<sub>2</sub>-induced corrosion than the conventional aqueous MEA.

**3.6.4. The role of HBA and HBD in [Triz]Cl/DETA.** To elucidate the CO<sub>2</sub> absorption mechanism of aqueous [Triz]Cl/DETA (30 wt% DES + 70 wt% H<sub>2</sub>O), both FTIR and <sup>13</sup>C NMR analyses were conducted before and after CO<sub>2</sub> absorption. As shown in Fig. 10(a), the emergence of new peaks at 1546 and 819 cm<sup>-1</sup> in the FTIR spectra can be attributed to C=O and C–O stretching vibrations, respectively, which are indicative of carbamate formation. Peaks at 1090 and 1162 cm<sup>-1</sup> were assigned to C–N and symmetric C–O stretching,<sup>36</sup> while a shoulder near 1327 cm<sup>-1</sup> reflected the characteristic vibration of carbamate C=O groups.<sup>11</sup>

In parallel, Fig. 10(b) shows the <sup>13</sup>C NMR spectra of [Triz]Cl/DETA before and after CO<sub>2</sub> absorption. A new peak at ~165 ppm appeared in the <sup>13</sup>C NMR spectrum of CO<sub>2</sub>-loaded [Triz]Cl/DETA, which was attributed to the carbonyl carbon of the carbamate species (–NHCOO–), confirming that chemical absorption occurred *via* the formation of zwitterionic intermediates, followed by stabilization through proton transfer. New signals were observed near 48 ppm, corresponding to the –CH<sub>2</sub>–N– groups formed after the CO<sub>2</sub> absorption. Moreover, a slight upfield shift near 45 ppm indicates changes in the local electronic environment of the amine carbon atoms, reflecting hydrogen-bond rearrangement and proton exchange during carbamate formation.<sup>37</sup> These observations collectively support the reaction of amine sites with CO<sub>2</sub>.<sup>38</sup>

To further clarify which component interacts with CO<sub>2</sub> and how the DES structure is formed, the FTIR spectra of 30 wt% DETA, 30 wt% [Triz]Cl, and 30 wt% [Triz]Cl/DETA were compared (Fig. S6). New bands at 2361 and 1255 cm<sup>-1</sup> appear only for 30 wt% [Triz]Cl/DETA, indicating specific hydrogen-bond interactions between the HBA ([Triz]Cl) and HBD (DETA). After CO<sub>2</sub> absorption, 30 wt% [Triz]Cl shows only minor spectral changes, whereas 30 wt% DETA clearly displays new carbamate bands, and the CO<sub>2</sub>-loaded spectrum of 30 wt% [Triz]Cl/DETA



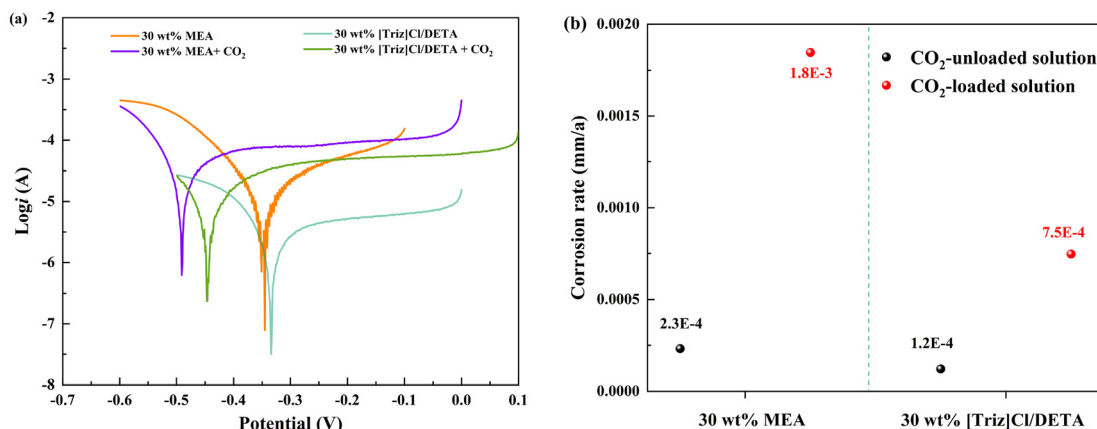


Fig. 9 (a) Tafel polarization curves of carbon steel in 30 wt% MEA and [Triz]Cl/DETA aqueous solutions; (b) corresponding corrosion rates determined from the Tafel extrapolation.

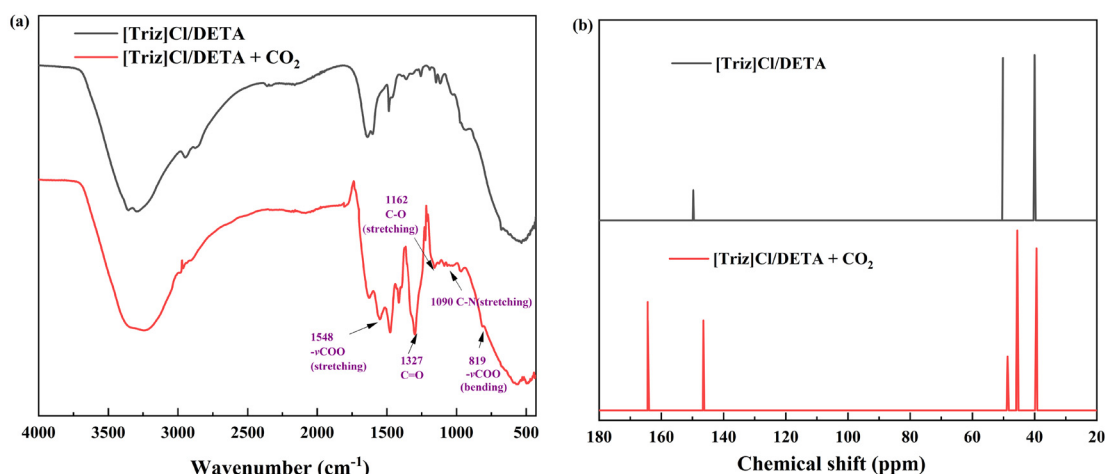


Fig. 10 (a) FTIR and (b)  $^{13}\text{C}$  NMR spectra of [Triz]Cl/DETA before and after  $\text{CO}_2$  absorption.

closely matches that of  $\text{CO}_2$ -loaded DETA. These results confirm that DETA provides the chemically active  $-\text{NH}-$  sites for  $\text{CO}_2$  binding, while [Triz]Cl mainly acts as the hydrogen-bond acceptor matrix rather than reacting directly with  $\text{CO}_2$ .

The respective roles of [Triz]Cl and DETA are also reflected in the macroscopic  $\text{CO}_2$  absorption behaviour and thermal stability. As shown in Fig. S7 (SI), 30 wt% DETA shows the highest  $\text{CO}_2$  capacity (0.20 g- $\text{CO}_2$  per g-solvent) but a slower absorption rate than 30 wt% [Triz]Cl/DETA, whereas 30 wt% [Triz]Cl exhibits only a very low  $\text{CO}_2$  capacity (0.02 g- $\text{CO}_2$  per g-solvent). In 30 wt% [Triz]Cl/DETA, the DETA content is 24.9 wt%. For comparison, the  $\text{CO}_2$  absorption capacity of 24.9 wt% DETA was estimated, proving a value of 0.16 g- $\text{CO}_2$  per g-solvent, which is almost the same as that of 30 wt% [Triz]Cl/DETA (0.17 g- $\text{CO}_2$  per g-solvent). Therefore, DETA provides the main chemically active  $-\text{NH}-$  sites for  $\text{CO}_2$  binding. The thermal stabilities of pure DETA and [Triz]Cl were also measured under the same conditions for comparison (Fig. 3). Both the  $T_{\text{onset}}$  and  $T_{\text{peak}}$  values of DETA are lower than those

of [Triz]Cl/DETA, whereas [Triz]Cl shows higher  $T_{\text{onset}}$  and  $T_{\text{peak}}$  values than both DETA and [Triz]Cl/DETA. These observations confirm that [Triz]Cl does not significantly absorb  $\text{CO}_2$  and primarily acts as a hydrogen-bond acceptor framework, thereby improving the physicochemical properties of the DES (e.g., thermal stability).

### 3.6.5. Sensible heat requirement for solvent regeneration.

A simplified metric was introduced to compare the sensible heat requirement during the [Triz]Cl/DETA regeneration. Briefly, the sensible heat required to raise the solvent from the absorption temperature (22 °C) to the desorption temperature (110 °C) was estimated, which was then normalized by the cyclic  $\text{CO}_2$  loading to obtain an indicative sensible heat requirement per unit  $\text{CO}_2$ . The detailed calculation procedure is provided in the SI (section 2). Based on this simplified analysis and assuming a specific heat capacity like that of typical aqueous amine solutions, the simplified sensible-heat index for 30 wt% [Triz]Cl/DETA is about 25% lower than that for 30 wt% MEA. Combined with the much lower mass loss at



110 °C (0.7% vs. 40%), these results indicate that [Triz]Cl/DETA is intrinsically favorable for energy-efficient regeneration.

It should be noted that this simplified metric reflects only the sensible-heat contribution. Indeed, the energy demand for solvent regeneration is a joint contribution of sensible heat to increase temperature, CO<sub>2</sub> absorption enthalpy, and energy used for vaporization. This work focused on the screening of effective solvents. Once the effective solvents are identified, in our future work, equilibrium and calorimetric measurements together with process-level simulations will be carried out to quantify the overall regeneration energy of the promising DES systems.

## 4. Conclusions

In this study, a series of DESs were synthesized using [Triz]Cl as the HBA and eight different amine-based compounds as HBDs to evaluate the relationship between HBD structure and CO<sub>2</sub> capture performance. The results demonstrated that the molecular structure and functionality of the HBD significantly influenced the CO<sub>2</sub> absorption capacity, absorption rate, thermal stability, and desorption performance. Specifically, short-chain polyamines such as EDA and DETA exhibited high CO<sub>2</sub> absorption capacities (0.18 and 0.17 g-CO<sub>2</sub> per g-solvent, respectively) owing to their dense -NH- active sites that facilitate carbamate formation; the thermal stability of the DESs follows the trend [Triz]Cl/TEPA > [Triz]Cl/AEEA > [Triz]Cl/MDEA > [Triz]Cl/DETA > [Triz]Cl/DEA > [Triz]Cl/AMP > [Triz]Cl/MEA > [Triz]Cl/EDA, emphasizing that stronger hydrogen-bond networks and higher molecular weights of the HBD enhance resistance to decomposition. Among them, 30 wt% [Triz]Cl/DETA exhibited the highest cyclic CO<sub>2</sub> loading (0.09 g-CO<sub>2</sub> per g-solvent) and desorption efficiency (57%), coupled with superior oxidative and corrosion stability, confirming its industrial applicability. FTIR and <sup>13</sup>C NMR analyses confirmed the formation of the carbamate species, providing clear evidence of chemical absorption of CO<sub>2</sub> in the [Triz]Cl/DETA systems. Overall, this work establishes clear structure–property correlations for DES-based CO<sub>2</sub> absorbents, demonstrating that optimizing amine density, molecular flexibility, and hydrogen-bonding interactions is key to balancing absorption performance, regeneration efficiency, and stability. These features suggest that the 30 wt% [Triz]Cl/DETA aqueous solution is a more energy-efficient and equipment-friendly CO<sub>2</sub> absorbent than the 30 wt% MEA aqueous solution, offering a clear green chemistry advantage.

## Author contributions

Qiangbing Shi: data curation and writing – original draft. Kaige Jia: writing – original draft. Xiangping Zhang: methodology and validation. Chuan Wang: validation. Paul Cobden: validation. Anna-Maria Beregi Amnéus: validation. David Muren: validation. Xiaoyan Ji: validation, conceptualization, supervision, and funding acquisition.

## Conflicts of 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 data supporting this article have been included as part of the supplementary information (SI). Supplementary information is available. Supplementary information: a schematic diagram of the CO<sub>2</sub> absorption and desorption apparatus, TG and DTG curves, CO<sub>2</sub> desorption behavior at different temperatures, and details of the min-max scaling method. See DOI: <https://doi.org/10.1039/d5gc05611j>.

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