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Characterisation and kinetic study of carbon dioxide absorption by an aqueous diamine solution

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HIGHLIGHTS

- The existence of –OH is negative to CO₂ absorption kinetics and capacity.
- The –CH₃ and increasing chain length improve the CO₂ absorption performance.
- The major species for MAPA–CO₂ system were analysed using FT-IR and ¹³C NMR.
- The mechanism of CO₂ capture into the MAPA solution was proposed.

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ABSTRACT

Aqueous monoamine solvents have been extensively studied for the purpose of CO₂ absorption to reduce emissions from sources such as industrial power stations. However, to improve the economic viability of carbon capture technologies, solvents with higher CO₂ absorption capacity and faster kinetics are urgently required. Diamines comprising two amino groups have potentially higher CO₂ absorption capacity and rates than monoamine solvents, such as monoethanolamine, and could be superior liquid absorbents for CO₂ absorption. In this study, we selected six linear diamines with a structure of NH₂(CH₂)_n-R (n = 2 or 3; R = NH₂, NHCH₃ or N(CH₃)₂) and four monoamines with a structure of NH₂(CH₂)_n-R (R = OH, CH₃ or CH(OH)CH₃). We then investigated the effect of diamine molecular structure on absorption kinetics and capacity using a bubble column, and confirmed the observed kinetic behaviours of selected diamines at different concentrations using a wetted-wall column and stopped-flow reactor. Under the conditions studied, all selected diamines had an absorption capacity of more than 0.78 moles of CO₂ per mole of amine, which far outstrips the capacity of monoamines. The hydroxyl group decreased the rate of CO₂ absorption, while the methyl group and longer chain lengths increased CO₂ absorption rate and capacity; the tertiary amino group exhibited the lowest kinetic performance. N-methylpropane-1,3-diamine (MAPA) had both the fastest absorption rate and the highest mass transfer coefficient. Using Fourier-transform infrared spectroscopy and ¹³C nuclear magnetic resonance, we elucidated the mechanism involved in the reaction of MAPA with CO₂. Our research provides a method for the future selection and design of new diamines for post-combustion CO₂ capture.

1. Introduction

Post-combustion carbon capture (PCC) from large emission sources, such as fossil-fuel power stations, is a straightforward way to curtail increasing atmospheric CO₂ concentrations [1]. At present, amine scrubbing is the leading CO₂ capture technology, and is also perceived as the most mature [2].

Various aqueous amines are used as absorption media for CO₂

capture. These include primary amines, such as monoethanolamine (MEA); secondary amines, such as diethanolamine (DEA); and tertiary amines, such as methyldiethanolamine (MDEA) [3]. The primary and secondary amines react with CO₂ via a zwitterion mechanism to form a carbamate as shown in the equations below, where B is a base and could be H₂O, OH[−] or an amine (Eqs. (1) and (2)) while tertiary amines react with CO₂ in the presence of water to form bicarbonate (Eq. (3)) [4]:



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Primary and secondary amines have fast CO₂ absorption kinetics, but low CO₂ absorption capacities (up to 0.5 mole/mole). Tertiary amines, on the contrary, have high CO₂ absorption capacities (up to 1 mole/mole) and low heat requirements for regeneration, but low CO₂ absorption kinetics [5,6]. Although the field of amine scrubbing has significantly advanced, the technology still has inherent drawbacks, such as a large energy penalty and high capital investment. These limitations are crucial barriers to the practical implementation of PCC [7,8]. For amine solvents, a greater absorption capacity minimises sensible heat losses, while a fast absorption kinetics reduces the required amount of packing for the same amount of CO₂ removal, hence reducing capital costs [9,10]. Therefore, developing an ideal solvent with high CO₂ absorption capacity and fast absorption kinetics is essential for a breakthrough in CO₂ absorption technologies, potentially reducing both the parasitic power demand of CO₂ capture and the capital investment.

Ionic liquids (ILs) are interesting alternatives for CO₂ absorption; they have negligible vapour pressure, high thermal stability and high CO₂ absorption capacity [11–13]. Unfortunately, IL viscosities are much higher than those of traditional aqueous amines, which significantly increases the diffusion coefficient of gases in solutions [14,15] and greatly reduces CO₂ absorption rates. CO₂ can also be captured by solid sorbents based on amines immobilised on porous solids, such as zeolites, activated carbons, mesoporous silicas and metal–organic frameworks [16,17]. These amines have demonstrated excellent CO₂ capture capabilities. However, solid amine sorbents are less well developed than aqueous amines and their industrial application is yet to be realised. Hybrid solvents of primary or secondary amines and tertiary amines could combine the advantages of each amine, providing both faster reaction kinetics and higher CO₂ absorption capacity. Several studies on CO₂ absorption with blended amines have been reported [18,19]. For example, Zhang et al. observed that higher concentrations of bicarbonate ions were formed and lower regeneration energy was achieved by adding a small amount of 1-dimethylamino-2-propanol (a tertiary amine) into an MEA solution [20]. In another study, Luo et al. found that blended amine solutions of MEA/*N,N*-diethylethanolamine demonstrated higher CO₂ absorption/regeneration performance and cyclic capacities than individual conventional amines [21]. However, although this approach has merit, its limitations cannot be ignored. Such mixtures typically possess the volatility, toxicity and stability of the worst-performing component in the formulation [22].


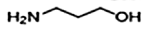

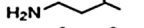
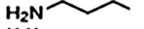
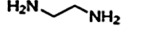


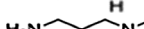

Diamines have two amine groups in one molecule, both of which can interact with CO₂ molecules. Therefore, they are expected to have better absorption capacity and kinetics than monoamines [23]. Using diamines in CO₂ absorption also represents a more energy-efficient prospect for PCC. Compared with monoamines, less solvent (mass) is required for diamines to achieve similar total concentrations of reactive nitrogen (amine). Thus, to obtain the same amount of CO₂ removal, less material needs to be circulated and heated in the stripper, thereby reducing the sensible heat requirement, pump work, and size and overall cost of the cross-exchanger [24]. An example of this scenario is the cyclic diamine-piperazine, which has been widely recognised for its rapid reactivity and low energy consumption for CO₂ absorption [25]. Kim et al. even confirmed that aqueous piperazine-derivative solutions had higher absorption capacities and lower heats of absorption than that of MEA solution at 313 K [26]. Machida et al. observed that 3-(diethylamino)propylamine, which contains a primary amino group for an increased CO₂ absorption rate and a tertiary amino group for an increased CO₂ absorption capacity in one molecular structure, had a higher absorption rate and capacity and lower absorption heat than MEA, MDEA and DEA [27]. Moreover, Cao et al. investigated a diamine (2-[(3-aminopropyl)methylamino]ethanol), which exhibited a faster

CO₂ absorption rate and larger equilibrium loading than MEA and MDEA [28]. While there is still significant scope for the development of alternative diamine solvents, further work associated with diamine screening and the design of diamine molecules is desirable. Considering that amine screening is very time consuming and expensive, a pre-evaluation method that can link an amine's performance to its molecular structure must be developed.

Extensive research has already been conducted to investigate the relationship between CO₂ absorption capacity and amine structure [29–31]. The studies of Didas et al. and Perinu et al. found that the type of amino group affects the CO₂ chemisorption process. Different amino groups have different basicity, which influences the strength of the interaction between CO₂ molecules and the amino groups; the chain length of the amine between the amino groups also significantly influences CO₂ absorption capacity [29,30]. Singh et al. reported that the absorption capacity of diamines increased with the increasing chain length [31]. Other functional groups of amine molecules should also be taken into consideration. For example, El Hadri et al. found that the methyl group is an electron donor that increases the molecule's CO₂ absorption capacity, and that the hydroxyl group is also a strong electron attractant, the effect of which on the nitrogen is to decrease CO₂ loading [3]. CO₂ absorption kinetics is also important for the screening of suitable amine candidates, because the kinetics of the reaction significantly affect both the absorption rate and the design of the PCC absorber. However, little work has been conducted on the effect of amine molecular structure on CO₂ absorption kinetics; in particular, a comprehensive investigation of the structure–kinetics relationships of diamines for CO₂ absorption is lacking. Therefore, a further understanding of the relationship between the molecular structures of diamine-based solvents and their kinetic performance is needed to develop better solvents for CO₂ absorption. An improved understanding will also help guide future diamine screening and design work.

In the present work, we investigated the effect of chain length and amino type on the CO₂ absorption performance of six linear diamines with a structure of NH₂(CH₂)_{*n*}-R, (*n* = 2 or 3; R = NH₂, NHCH₃ or N(CH₃)₂). In addition, we tested four monoamines with the structure of NH₂(CH₂)_{*n*}-R, (R = OH, CH₃ or CH(OH)CH₃) to elucidate the effect of functional groups on CO₂ absorption. Table 1 lists all amines used in this work along with their abbreviated forms, molecular weights and molecular structures. Our study had three main parts. First, we used a bubble column to determine the initial absorption rate (*r*₀) and absorption capacity of CO₂ using different aqueous amines. The initial CO₂ absorption rate was defined as the absorption rate in the first minute: when the CO₂ loading in each amine can be assumed to be zero, and the absorption capacity corresponds to the maximum molar amount of CO₂ absorbed per mole of amine. Understanding the link between the amine structure, initial absorption rate and CO₂ solubility

Table 1
Amines used in this work.

Amine solution	Abbreviation	Mol wt	Molecular structure
Monoethanolamine	MEA	61.08	
3-Amino-1-propanol	3AP	75.11	
4-Amino-2-butanol	4AB	103.16	
1-Aminobutane	1BA	73.14	
Ethylenediamine	EDA	60.1	
1,3-Diaminopropane	1,3-PDA	74.13	
<i>N</i> -methylthylenediamine	MEDA	74.13	
<i>N</i> -methylpropane-1,3-diamine	MAPA	88.15	
2-Dimethylaminoethylamine	DMAEA	88.15	
3-Dimethylamino-propylamine	DMAPA	102.18	

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