Short Communication



Diamine based water-lean CO₂ solvent with extra high cyclic capacity and low viscosity

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Abstract: The industrial application of emerging water-lean solvents to CO₂ capture from flue gas is challenged by their high viscosity. In this work, we report a novel water-lean CO₂ solvent which possesses lower viscosity and higher CO₂ cyclic capacity than other water-lean solvents reported in the literature. The new solvent consists of *N*, *N*-dimethyl-1, 2-ethanediamine (DMEDA), physical cosolvent *N*-methyl-2-pyrrolidone (NMP) and up to 15% water (named ENH). We evaluated the effect of the solvent composition on the viscosity, CO₂ cyclic capacity and regeneration energy of ENH and compared it with the reference monoethanolamine (MEA) based solvents. It was found that ENH containing 5% H₂O (ENH-5%H₂O) with a CO₂ loading of 0.767 mol CO₂·mol amine⁻¹ had a viscosity of 7.603 mPa·S at 40 °C, which was comparable with that of traditional blended amines. Excellent cyclic capacity performance was also observed, with ENH-5% H₂O showing a 140% improvement compared to aqueous MEA. Regeneration energy of ENH-5% H₂O was estimated to be 2.418 GJ·tCO₂⁻¹ which is 36% lower than the 30 wt. % aqueous MEA solvent. © 2021 Society of Chemical Industry and John Wiley & Sons, Ltd.

Additional supporting information may be found online in the Supporting Information section at the end of the article.

Keywords: CO₂ capture; water-lean; diamine; viscosity; cyclic capacity

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Introduction

ithin the field of CO₂ capture, utilization, and storage (CCUS) for the mitigation of greenhouse gas emissions, CO₂ capture using aqueous amine-based absorbents is regarded as the most technically mature approach. Recently, water-lean solvents, which mix amines with organic diluents and contain low or no water, have gained considerable interests due to their lower energy requirement for regeneration and ability to be used in existing processes with minimal modification.

Previous investigations have found that water-lean solvents contribute to energy savings compared to traditional aqueous solvents. The regeneration energy of monoethanolamine (MEA) blended with ethylene glycol (EG) was found to be 23.8–31.3% lower than that of aqueous MEA.¹ Lv *et al.* found that a nonaqueous solvent containing 2-amino-2-methyl-1-propanol (AMP), (2-(2-aminoethylamino)ethanol and *N*-methyl-2-pyrrolidone (NMP) achieved a low regernation energy (2.09 GJ·tCO₂⁻¹) which was approximately half that of aqueous MEA.²

However, water-lean solvents are generally challenged by their high viscosity.³⁻⁶ It was reported that water-lean solvents containing alcohol derivatives had high viscosity values, even up to ~1300 mPa⋅S at 40 °C with CO₂ loading⁷ which would significantly reduce the mass and heat transfer efficiency in the CO₂ capture process. Moreover, alcohol derivatives were easily oxidized in an oxygen-containing environment, leading to a more severe degradation.8 Thus, organic solvents, for example sulfolane (SFL), N-methyl-2-pyrrolidone (NMP) and poly (ethylene glycol) dimethylether (NHD), with high thermal stability and low vapor pressure were selected to form water-lean systems. SFL, NMP, and NHD based solvents can help lower the high viscosity of water-lean solvents to a certain extent⁹⁻¹¹ A relatively low viscosity (3.93 mPa·S) was achieved for the water-lean DETA/SFL solvent at a CO₂ loading of 2.21 mol·L⁻¹ (equal to 1.105 mol CO₂·mol amine⁻¹) at 348 K.9 7m MEA mixed with NMP and H₂O in a mass ratio of NMP: H₂O of 19:1 (equal to 3.5wt. % H₂O) had a viscosity of 15.4 mPa·S with a CO₂ loading of 0.45 mol CO₂·mol amine⁻¹.¹⁰ It was illustrated that multiple factors such as NMP fraction influenced on the viscosity performance of MEA-NMP- H_2O . However, that is still 5 times more viscous than 30 wt% aqueous MEA. A viscosity of 37.4 mPa·S was achieved in the lower phase of biphasic solvent containing 1 M

3-dimethylaminopropylamine (DMAPA) mixed with NHD and $\rm H_2O$ (volume ratio: 8:2). It was indicated that DMAPA with no hydroxyl group had weaker hydrogen bonding effect, leading to a relatively lower viscosity than MEA in the same volume ratio of NHD and $\rm H_2O$.¹¹

In this work, we report a novel, high performing water-lean solvent with a lower viscosity than those reported water lean solvents in the literature. It is composed of a di-amine, N, N-dimethyl-1, 2-ethanediamine (DMEDA), an organic physical solvent NMP and up to 15% water. Aqueous DMEDA was reported to exhibit a higher absorption rate and cyclic capacity compared with aqueous MEA.¹² We carried out several experiments to investigate the performance of water-lean solvents including viscosity, cyclic capacity, and regeneration energy. 30 wt% aqueous MEA solvent was used as a benchmark for comparison. To our knowledge, it is the first time that the DMEDA based water-lean solvent has been reported for CO₂ capture. The promising results shown in this work will stimulate more research work to further develop this solvent and water-lean solvents in general.

Experimental

Materials

Carbon dioxide (CO₂, 99.9%) and nitrogen (N₂, 99.9%) gases were supplied by BOC Australia. Monoethanolamine (MEA, 99% Merck), *N*,*N*-dimethyl-1,2-ethanediamine (DMEDA, 99% Sigma), 1-methyl-2-pyrrolidone (NMP, 99% Chem-supply) were used as received and their CAS numbers and molecular structures are provided in Table S1. Deionized water was used to prepare the water-lean solutions. The solvents in this study all contained 30 wt. % of amines, and the water content varied among 5, 10, 15, and 70%, with the balance made up by NMP (Table S2).

Viscosity and CO₂ absorption enthalpy measurement

The details for viscosity and CO₂ absorption enthalpy measurement were previously presented by Xiao *et al.*¹² and Luo *et al.*¹³

Desorption test

The CO₂ desorption device was described in our previous work.¹⁴ All the materials used were weighed

by an analytical balance to the planned quantities in the preparation of the solvents. The weight percentage concentration was used in this work. The rich solvent was obtained after 24 hr absorption in a jacketed flask at 40 °C under 101kPa CO₂ partial pressure. The stirring rate was 800 rpm controlled by a magnetic stir plate. Prepared rich solvent was directly heated at 90 °C with the same stirring rate. Seven samples (0.5 mL) were collected during 60 min of desorption time and analyzed by NMR. The NMR facility and analysis procedure were described by Xiao *et al.*¹²

Condensate water measurement

The regeneration energy of CO_2 solvent (Q_{reg} , $GJ \cdot tCO_2^{-1}$) was composed of three parts: reaction heat (Q_{reac}), sensible heat (Q_{sens}) and latent heat (Q_{latent}), as given in Eq. 1. The calculation equation of each part is shown in Eqn 2.^{15–17}

$$Q_{\text{reg}} = Q_{\text{reac}} + Q_{\text{sens}} + Q_{\text{latent}} \tag{1}$$

$$Q_{\text{reg}} = \frac{-H_{\text{abs}}}{M} + \frac{C_p m_{\text{sol}} \Delta T}{m_{\text{CO}_2}} + \frac{n_w}{M \times n_{\text{CO}_2}} \Delta H_w^{\text{vap}} \quad (2)$$

where, H_{abs} is the reaction heat of CO₂ absorption, kJ·mol· CO_2^{-1} , M is the molecular weight of CO_2 , 44 g·mol⁻¹, C_p is the heat capacity of rich solvents, kJ·kg⁻¹·K⁻¹, m_{sol} is the weight of rich solvents, kg, ΔT is the temperature difference between lean and rich solvents which is assumed as 10K, m_{CO2} is the weight of CO₂ absorbed, g, $\Delta H_{\rm w}^{\rm vap}$ is the heat of water evaporation, kJ·mol⁻¹, n_w and n_{CO2} is the amount of water evaporated and CO2 desorbed, mol which was determined using the apparatus shown in Fig. S1. Fresh solvent (500 g) was stirred with pure CO₂ gas presaturated with water flowing at a rate of 100 mL min⁻¹ for 6 hr. Then, 100g of the obtained rich solvent was injected into a 250 mL three necked flask and heated at 90 °C controlled by an oil bath for 1 hr. The stirring rate was maintained at 800 rpm during the whole experiment. Water vapor was condensed and collected by a condenser. A refrigerating circulator supplied cooling water at 3 °C to the condenser. An electronic soap film flowmeter (BL-1000, range 1–1000 mL min⁻¹, accuracy 1%) was used to monitor the CO₂ desorption rate throughout the whole desorption process. The amount of CO₂ desorbed was measured by a CO₂ loading test device based on neutralization reaction, the detail of which was described by Zhu et al. 18

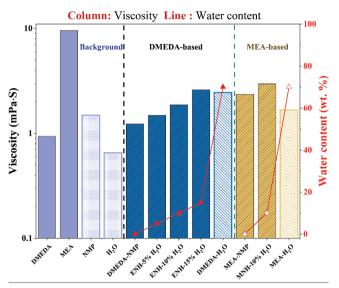


Figure 1. Viscosity of CO₂ free water-lean and aqueous solvents at 40 °C. The data for MEA-H₂O is from reference.²²

Results and discussion

Viscosity

The viscosity of fresh ENH water-lean solvents comprising different water content (0–15%) at 313 K is shown in Fig. 1. Aqueous MEA and MNH water-lean solvents were selected for comparison. It was found that the viscosity of the ENH solvent series dropped appreciably with a decrease in water content in the measured water fraction range (0–15%). A decline of 53% was achieved through decreasing water content of the ENH solvent from 15 to 0%, and DMEDA-NMP had a viscosity 26% lower than aqueous MEA.

It is known that the viscosity of a mixed solvent is strongly affected by the viscosity of each component (background viscosity) and interactions among molecules in the mixture. The results show that the co-solvent NMP (1.49 mPa·S) has a viscosity 56% higher than H₂O (0.65mPa·S). The measured viscosity of four DMEDA-based water-lean solvents was different from the expectation that the system viscosity would decrease as the fraction of water and NMP increased and decreased, respectively. In these cases, the impact of background viscosity changes attributed to the small change of water content (0-15%) was negligible. It is believed that the hydrogen bond between water and amine played a key role in the viscosity changes¹⁹ of the DMEDA-based water-lean system. The increased water fraction enhanced hydrogen bond formation with the amine, resulting in an increase in viscosity. Whereas, the DMEDA-H₂O

aqueous solvent had a relatively lower viscosity compared with that of ENH-15% $\rm H_2O$. This suggests that further addition of water contributed more to background viscosity after the amine was fully solvated by water to form hydrogen bonds.

A different trend was observed in MEA-based solvents. The viscosity of MEA blends decreased sharply by the addition of water and/or NMP. MEA has one electron withdrawing hydroxyl group and one electron donating primary amino group. The two functional groups match well to form strong inter-molecular hydrogen bonds.²⁰ These hydrogen bonds could result in MEA molecules connected in linear, branched, or mixed ways, resulting in a viscous mixture. However, this intermolecular hydrogen bond is disrupted when water or NMP is introduced. The nitrogen and oxygen atoms in NMP are locked in an amide group, hence suppressing hydrogen bond formation with either amino or hydroxyl groups of MEA under general CO₂ capture process conditions. The hydroxyl group of MEA can behave as either hydrogen acceptor or donor in hydrogen bond formation.²¹ The hydroxyl group in MEA (pKa 15.61) should behave as a hydrogen acceptor when fully solvated by water (pKa 14) to form much weaker hydrogen bonds compared to that formed with an amino group. Hence, a decrease of viscosity was observed in all MEA-based solvents, in which MEA formulated with H₂O resulted in a lower viscosity than that with NMP.

Regarding the influence of amine structure, MEA based solvents always resulted in a higher viscosity than DMEDA based ones with the same water content (0 and 10%) in these water-lean systems. In addition to the background viscosity difference between MEA and DMEDA, the ability to form hydrogen bonds also plays a crucial role in viscosity performance. In nonaqueous systems, the intermolecular bonding interaction was preferred in MEA-NMP relative to DMEDA-NMP, leading to a higher viscosity of the MEA-NMP solvents. In the 10% H₂O system, the hydroxyl group of MEA was regarded as a hydrogen acceptor from water and is more hydrophilic than tertiary amino group of DMEDA, which contributed to hydrogen bonding with water. As a consequence, a more extensive hydrogen bonding network was able to form in MNH-10%H₂O leading to a larger viscosity.

Although DMEDA-NMP had the lowest viscosity without CO₂ loading, carbamate precipitated out as solid during CO₂ uptake, hence it is unsuitable for

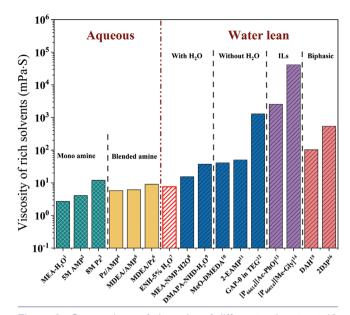


Figure 2. Comparison of viscosity of different solvents at 40 °C with rich loading (Except two ionic liquids which were tested at 30 °C). Rhombic pattern: aqueous solvent, no pattern: blended solvent, diagonal pattern: water-lean solvent.¹ loading: 0.5 mol $CO_2 \cdot mol$ amine-¹,² loading: 0.20 mol $CO_2 \cdot mol$ amine-¹,³ loading: 0.10 mol $CO_2 \cdot mol$ amine-¹,⁴ loading: 0.13 mol $CO_2 \cdot mol$ amine-¹,⁴ loading: 0.13 mol $CO_2 \cdot mol$ amine-¹,⁵ loading: 0.75 mol $CO_2 \cdot mol$ amine-¹,⁶ loading: 0.13 mol $CO_2 \cdot (mol \ N)^{-1},^{7}$ loading: 0.767 mol $CO_2 \cdot mol$ amine-¹,⁶ loading: 1.06 mol $CO_2 \cdot mol$ amine-¹,¹⁰ loading: 0.7 mol $CO_2 \cdot mol$ amine-¹,¹⁰ loading: 0.7 mol $CO_2 \cdot mol$ amine-¹,¹⁰ loading: 0.7 mol $CO_2 \cdot mol$ amine-¹,¹⁰ loading: 1.20 mol $CO_2 \cdot mol$ amine-¹,¹⁰ loading: 0.90 mol $CO_2 \cdot kg$ -¹,¹⁰ loading: 0.564 mol $CO_2 \cdot mol$ amine-¹.

application in common absorption and desorption equipment. This indicates that the presence of some water is important to ensure the system remains as a single phase upon reaction with CO₂. Therefore, ENH-5%H₂O which had the second smallest viscosity of the fresh solvents was chosen to test the viscosity after absorption of 15 kPa CO₂ at 40 °C for 5 hr and compared with published typical mono-amine solvents, blended solvents, and water-lean solvents (Fig. 2). Some CO₂ loading results of the published solvents quoted in Fig. 2 were displayed in different units in the original paper. They were converted to the unit of mol CO₂·mol amine⁻¹ for unification. The abbreviation and composition of all solvents are listed in Table S3. ENH-5% H₂O with a CO₂ loading of 0.767 mol CO₂·mol amine⁻¹ had a viscosity of 7.603 mPa·S, which is the lowest viscosity among reported

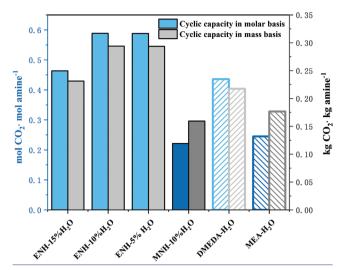


Figure 3. Cyclic capacity performances of different solvents.

water-lean solvents so far. Compared to aqueous amine systems, the viscosity of ENH-5% H_2O solvent was close to MDEA/AMP blends and 36.6 % smaller than that of aqueous Pz. This enables the use of ENH in existing industrial absorber and heat exchanger equipment. Compared to water free solvents (e.g., GAP-0 in tri-ethylene glycol (TEG)) and Ionic Liquids (ILs), the determined viscosity of ENH-5% H_2O is 2–5 orders of magnitude smaller. This is because that the background viscosity of GAP-0 and ILs is much bigger than small alkylamine molecules like DMEDA.

Cyclic capacity

Cyclic capacity of DMEDA and MEA-based solvents is shown in Fig. 3. As the difference of cyclic capacity in two different units was unapparent, only cyclic capacity in mole basis was used below for an easy discussion.

Figure 3 shows that ENH solvents achieved improvements in cyclic capacity compared with aqueous DMEDA-H₂O. ENH solvents with smaller water content (5%, 10%) resulted in higher cyclic capacity, in which ENH-5%H₂O showed an improvement of 140% with respect to the aqueous MEA-H₂O solvent.

The introduction of a nonprotonic polar co-solvent NMP greatly effects the cyclic capacity of the studied amines. It was reported that NMP can physically absorb CO₂ at higher pressure (> 2MPa). However, solubility of CO₂ at atmosphere pressure was quite small²³ which was considered negligible in this case. Thus, we postulate that the low

polarity of NMP is the key reason for the large cyclic capacity when it's used as a diluent in a water lean solvent based on a basic rule "like seeks like." Comparing to H₂O, the nonprotonic polar co-solvent NMP has weak ability to stabilize the high-polarity product: bicarbonate/carbonate, resulting in a significant reduction of bicarbonate/carbonate amount. While for the carbamate comprising a lower polarity than bicarbonate/carbonate, NMP can stabilize it to a certain extent to ensure its formation during CO₂ absorption. Hence carbamate was detected as the major absorption product in water lean solvents by NMR analysis. However, in the desorption process, the weaker solvation ability of NMP was declined by the higher temperature and further reduced the stability of the polar carbamate molecule, resulting in an easier decomposition of carbamate to release more CO₂. Hence, this is the reason why ENH-15%H₂O with lowest NMP ratio gained lowest cyclic capacity compared with other two ENH solvents. However, comparable cyclic capacities were observed in ENH-5%H₂O and ENH-10%H₂O due to the differing contributions from bicarbonate and carbamate. The 5% higher water content in ENH-10%H₂O allowed more bicarbonate formation while further stabilized the carbamate molecule. These two impacts made similar but opposite contribution to the cyclic capacity, resulting the similar overall cyclic capacities between ENH-5%H₂O and ENH-10%H₂O.

In contrast, MEA-H₂O obtained a slightly higher cyclic capacity than MNH-10%H₂O. This is because more bicarbonate/carbonate was formed in rich MEA-H₂O and its decomposition dominated in the desorption process. MNH-10%H₂O formed smaller amount of bicarbonate/carbonate than MEA-H₂O, however achieved similar cyclic capacity. This also demonstrates that the introduction of NMP is able to boost cyclic capacity due to carbamate decomposition under mild conditions.

To further explain these results, the free energy barriers of CO₂ desorption in water-lean and aqueous systems were computed by Gaussian 09. The details of calculation method are given in the supporting information.

Optimized structures of reactant complex, transition state and free energy of CO_2 desorption reaction of aqueous and water-lean system are displayed in Fig. S2. It was found that the free energy barrier of CO_2 desorption in an aqueous system (91.18 kJ·mol⁻¹) was significantly higher than that in the DMEDA-based

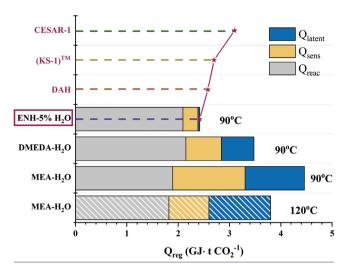


Figure 4. The regeneration energies of ENH-5% H_2O , DMEDA- H_2O , MEA- H_2O , DAH, (KS-1) TM and CESAR-1 solvents.

water-lean system (78.30 kJ·mol⁻¹). It is known that the reaction with a lower energy barrier requires less energy to proceed, which aligns to the results of larger cyclic capacity observed in the water-lean system.

Regeneration energy

ENH-5%H $_2$ O which has a satisfactory viscosity and large cyclic capacity has potential to reduce energy demand compared to aqueous solvents. Hence, ENH-5%H $_2$ O was chosen to compare the calculated regeneration energy with two aqueous solvents DMEDA-H $_2$ O and MEA-H $_2$ O. Parameters used for energy calculation are given in Table S4. The regeneration energy measured here was based on a lower desorption temperature (90 °C) which was not optimal for MEA-H $_2$ O so that the energy consumption result of MEA-H $_2$ O desorbed at 120 °C 15 was also displayed in Fig. 4.

An obvious decrease of sensible heat was obtained in the di-amine based solvents (DMEDA-H₂O and ENH-5%H₂O), which was attributed to their larger cyclic capacity compared with aqueous MEA at both 90 and 120 °C. Moreover, organic co-solvent NMP has a significantly lower heat capacity than water. Hence, an extra decrease (57.9 %) of sensible heat was achieved in ENH-5%H₂O compared with the DMEDA-H₂O aqueous solvent. ENH-5%H₂O with smaller water content further suppressed the water vaporization, achieving a quite low latent heat of 0.034 GJ·tCO₂⁻¹. A regeneration energy of 2.418 GJ·t CO₂⁻¹ was calculated for the ENH-5%H₂O case, which was 36% and 30%

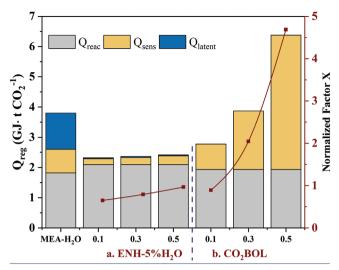


Figure 5. Normalized regeneration energies for ENH-5%H₂O and CO₂BOL with different parameters of the heat exchanger α .

lower than MEA (120 °C) and DMEDA aqueous solvents, respectively. In Fig. 4, ENH-5%H₂O was also compared for the regeneration energy with reported solvents such as proprietary solvents:(KS-1) TM, ²⁴ biphasic solvents: DAH¹⁵ and new benchmark²⁵: CESAR-1.²⁶ ENH-5%H₂O has the smallest regeneration energy suggesting good potential improved performance in industrial application.

In an industrial CO_2 capture process, higher viscosity of solvents can significantly reduce the heat transfer efficiency of lean/rich exchangers, which may result in a larger sensible heat requirement.²⁷ A normalized factor χ was introduced^{28,29} to correct the required sensible heat compared with MEA-H₂O (Eqn S5).

CO₂BOL (IPADM-2BOL).^{30–32} which is a novel CO₂ binding organic liquid with high cyclic capacity but high viscosity, was chosen to illustrate the impact of viscosity on sensible heat together with ENH-5%H₂O (Fig. 5). The parameters of CO₂BOL used for the energy calculation are listed in Table S5. The regeneration energy of ENH-5%H₂O was insignificantly affected by the parameters of the heat exchangers (α) and was approximately half of that of MEA-H₂O,¹⁵ In contrast, the CO₂BOL performed differently in regeneration. Due to its more viscous property, the regeneration energy of CO₂BOL rose rapidly with increasing α . As α increased from 0.1 to 0.5, the sensible heat of CO₂BOL increased fivefold, resulting in a regeneration energy over 6.0 GJ·tCO₂⁻¹ which was far larger than that of aqueous MEA. It was

demonstrated that viscosity had a significant effect on regeneration energy and is an important property to be considered during solvent development.

Conclusion

Water-lean solvents consisting of DMEDA, organic physical co-solvent NMP and H2O (named ENH) were evaluated in this work. Several merits such as low viscosity, high cyclic capacity and low regeneration energy were demonstrated in ENH water-lean solvents. For ENH-5%H₂O with CO₂ loading of 0.767 mol CO_2 ·mol amine⁻¹, the observed viscosity is only 7.603 mPa·S at 40 °C. Improvement in cyclic capacity of 140% for ENH-5%H₂O was achieved in comparison with 30 wt% aqueous MEA. The regeneration energy of ENH-5%H₂O was estimated to be 2.418 GJ·tCO₂⁻¹ which is 36 % lower than that of aqueous 30 wt% aqueous MEA. Normalized regeneration energy highlighted the impact of viscosity on sensible heat. ENH-5%H₂O of lower viscosity showed a similar regeneration energy no matter the value of heat exchangers parameter (α) was, and this can significantly reduce the size/surface area requirement of heat exchangers.

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References

- Kang MK, Jeon SB, Cho JH, Kim JS and Oh KJ, Characterization and comparison of the CO₂ absorption performance into aqueous, quasi-aqueous and non-aqueous MEA solutions. *Int J Greenh Gas Control* 63(July):281–288 (2017). https://doi.org/10.1016/j.ijggc.2017.05.020.
- Bihong L, Kexuan Y, Xiaobin Z, Zuoming Z and Guohua J, 2-amino-2-methyl-1-propanol based non-aqueous absorbent for energy-efficient and non-corrosive carbon dioxide capture. Appl Energy 264(January):114703 (2020). https://doi.org/10.1016/j.apenergy.2020.114703.
- Song JH, Yoon JH, Lee H and Lee KH, Solubility of carbon dioxide in monoethanolamine + ethylene glycol + water and monoethanolamine + poly(ethylene glycol) + water. J Chem Eng Data 41(3):497–499 (1996). https://doi.org/10.1021/je9502758.
- Svensson H, Edfeldt J, Zejnullahu Velasco V, Hulteberg C and Karlsson HT, Solubility of carbon dioxide in mixtures of 2-amino-2-methyl-1-propanol and organic solvents. *Int J Greenh Gas Control* 27:247–254 (2014). https://doi.org/10.1016/j.ijggc.2014.06.004.

- Barzagli F, Lai S and Mani F, A new class of single-component absorbents for reversible carbon dioxide capture under mild conditions. *ChemSusChem* 8(1):184–191 (2015). https://doi.org/10.1002/cssc.201402421.
- Malhotra D, Cantu DC, Koech PK, Heldebrant DJ, Karkamkar A, Zheng F et al., Directed hydrogen bond placement: Low viscosity amine solvents for CO₂ capture. ACS Sustain Chem Eng 7(8):7535–7542 (2019). https://doi.org/10.1021/acssuschemeng.8b05481.
- Perry RJ and Davis JL, CO₂ capture using solutions of alkanolamines and aminosilicones. *Energy and Fuels* 26(4):2512–2517 (2012). https://doi.org/10.1021/ef201963m.
- Shoukat U, Baumeister E, Pinto DDD and Knuutila HK, Thermal stability and corrosion of tertiary amines in aqueous amine and amine-glycol-water solutions for combined acid gas and water removal. *J Nat Gas Sci Eng* 62:26–37 (2019). https://doi.org/10.1016/j.jngse.2018.11.025.
- Wang L, Yu S, Li Q, Zhang Y, An S and Zhang S, Performance of sulfolane/DETA hybrids for CO₂ absorption: Phase splitting behavior, kinetics and thermodynamics. *Appl Energy* 228(March):568–576 (2018). https://doi.org/10.1016/j.apenergy.2018.06.077.
- Yuan Y and Rochelle GT, CO2 absorption rate in semi-aqueous monoethanolamine. Chem Eng Sci 182:56–66 (2018). https://doi.org/10.1016/j.ces.2018.02.026.
- Qiu Y, Lu H, Zhu Y, Liu Y, Wu K and Liang B, Phase-change CO₂ absorption using novel 3-dimethylaminopropylamine with primary and tertiary amino groups. *Ind Eng Chem Res* 59(19):8902–8910 (2020). https://doi.org/10.1021/acs.iecr.9b06886.
- Xiao M, Cui D, Yang Q, Liang Z, Puxty G, Conway W et al., Advanced designer amines for CO₂ capture: interrogating speciation and physical properties. Int J Greenh Gas Control 82:8–18 (2019). https://doi.org/10.1016/j.ijggc.2018.12.021.
- Luo W, Yang Q, Conway W, Puxty G, Feron P and Chen J, Evaluation and modeling of vapor-liquid equilibrium and CO₂ absorption enthalpies of aqueous designer diamines for post combustion capture processes. *Environ Sci Technol* 51(12):7169–7177 (2017). https://doi.org/10.1021/acs.est.7b00379.
- Zhang R, Yang Q, Liang Z, Puxty G, Mulder RJ, Cosgriff JE et al., Toward efficient CO₂ capture solvent design by analyzing the effect of chain lengths and amino types to the absorption capacity, bicarbonate/carbamate, and cyclic capacity. Energy and Fuels 31(10):11099–11108 (2017). https://doi.org/10.1021/acs.energyfuels.7b01951.
- Liu F, Fang M, Dong W, Wang T, Xia Z, Wang Q et al., Carbon dioxide absorption in aqueous alkanolamine blends for biphasic solvents screening and evaluation. Appl Energy 233–234(August 2018):468–477 (2019). https://doi.org/10.1016/j.apenergy.2018.10.007.
- Zhao B, Liu F, Cui Z, Liu C, Yue H, Tang S et al., Enhancing the energetic efficiency of MDEA/PZ-based CO₂ capture technology for a 650MW power plant: process improvement. Appl Energy 185:362–375 (2017). https://doi.org/10.1016/j.apenergy.2016.11.009.
- Nwaoha C, Idem R, Supap T, Saiwan C, Tontiwachwuthikul P, Rongwong W et al., Heat duty, heat of absorption, sensible heat and heat of vaporization of 2-amino-2-methyl-1-propanol (AMP), piperazine (PZ) and monoethanolamine (MEA) tri-solvent blend for carbon dioxide

- (CO₂) capture. *Chem Eng Sci* **170**:26–35 (2017). https://doi.org/10.1016/j.ces.2017.03.025.
- Zhu D, Fang M, Zhong L, Zhang C and Luo Z, Semi-batch experimental study on CO₂ absorption characteristic of aqueous ammonia. *Energy Procedia* 4:156–163 (2011). https://doi.org/10.1016/j.egypro.2011.01.036.
- Ma Y, Liu Y, Su H, Wang L and Zhang J, Relationship between hydrogen bond and viscosity for a series of pyridinium ionic liquids: Molecular dynamics and quantum chemistry. *J Mol Liq* 255:176–184 (2018). https://doi.org/10.1016/j.molliq.2018.01.121.
- Hendricks SB, Wulf OR, Hilbert GE and Liddel U, Hydrogen bond formation between hydroxyl groups and nitrogen atoms in some organic compounds. J Am Chem Soc 58(10): 1991–1996 (1936). https://doi.org/10.1021/ja01301a050.
- Sadakiyo M, Yamada T and Kitagawa H, Hydroxyl group recognition by hydrogen-bonding donor and acceptor sites embedded in a layered metal - organic framework. *J Am Chem Soc* 133(29):11050–11053 (2011). https://doi.org/10.1021/ja203291n.
- Amundsen TG, Øi LE and Eimer DA, Density and viscosity of monoethanolamine + water + carbon dioxide from (25 to 80)°C. J Chem Eng Data 54(11):3096–3100 (2009). https://doi.org/10.1021/je900188m.
- Murrieta-Guevara F, Romero-Martinez A and Trejo A, Solubilities of carbon dioxide and hydrogen sulfide in propylene carbonate, N-methylpyrrolidone and sulfolane. *Fluid Phase Equilib* 44(1):105–115 (1988). https://doi.org/10.1016/0378-3812(88)80106-7.
- Van Straelen J and Geuzebroek F, The thermodynamic minimum regeneration energy required for post-combustion CO₂ capture. *Energy Procedia* 4:1500–1507 (2011). https://doi.org/10.1016/j.egypro.2011.02.017.
- Feron PHM, Cousins A, Jiang K, Zhai R and Garcia M, An update of the benchmark post-combustion CO₂-capture technology. Fuel 273(May 2019):117776 (2020). https://doi.org/10.1016/j.fuel.2020.117776.



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- Sanchez Fernandez E, Goetheer ELV, Manzolini G, Macchi E, Rezvani S and Vlugt TJH, Thermodynamic assessment of amine based CO₂ capture technologies in power plants based on european benchmarking task force methodology. *Fuel* 129:318–329 (2014). https://doi.org/10.1016/j.fuel.2014.03.042.
- Xin K, Gallucci F and van Annaland MS, Optimization of solvent properties for post-combustion CO₂ capture using process simulation. *Int J Greenh Gas Control* 99(June):103080 (2020). https://doi.org/10.1016/j.ijggc.2020.103080.
- Liu F, Fang M, Yi N, Wang T and Wang Q, Biphasic behaviors and regeneration energy of a 2-(Diethylamino)-Ethanol and 2-((2-Aminoethyl)Amino) ethanol blend for CO₂ capture. Sustain Energy Fuels 3(12):3594–3602 (2019). https://doi.org/10.1039/c9se00821q.
- Li L, Voice AK, Li H, Namjoshi O, Nguyen T, Du Y et al., Amine blends using concentrated piperazine. Energy Procedia 37:353–369 (2013). https://doi.org/10.1016/j.egypro.2013.05.121.
- Malhotra D, Koech PK, Heldebrant DJ, Cantu DC, Zheng F, Glezakou VA et al., Reinventing design principles for developing low-viscosity carbon dioxide-binding organic liquids for flue gas clean up. ChemSusChem 10(3):636–642 (2017). https://doi.org/10.1002/cssc.201601622.
- Zheng F, Heldebrant DJ, Mathias PM, Koech P, Bhakta M, Freeman CJ et al., Bench-scale testing and process performance projections of CO₂ capture by CO₂-binding organic liquids (CO2BOLs) with and without polarity-swing-assisted regeneration. Energy and Fuels 30(2):1192–1203 (2016). https://doi.org/10.1021/acs.energyfuels.5b02437.
- Koech PK, Zhang J, Kutnyakov IV, Cosimbescu L, Lee SJ, Bowden ME et al., Low viscosity alkanolguanidine and alkanolamidine liquids for CO₂ capture. RSC Adv 3(2):566–572 (2013). https://doi.org/10.1039/c2ra22801q.

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their chemical reaction behaves and capacity in CO_2 capture process. She carried out studies to enhance the related fundamental knowledge and to address application concerns in CO_2 capture. She has also designed solvents with high efficiency and advanced properties to the CO_2 capture at large scales.



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Graeme studied chemistry and computer science at The University of Newcastle (AU). He then did his PhD in chemistry finishing in 2004. 2005–2007 were spent as a postdoc first in Sweden and then Switzerland. In 2007 Graeme returned to Newcastle and

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Research Focus: The chemical and physical processes involved in the separation of carbon dioxide and other acid gases from gas mixtures, and their potential transformation into valuable materials. Both from the perspective of measuring what is happening and building predictive models to simulate what is happening. By understanding in detail the mechanisms of these processes improvements can be made to the gas absorbing materials, chemical transformation steps and the design of the overall separation process. These improvements come from a combination of chemical insights and simulations used to guide the research focus. Carrying out this research couples chemical kinetics, equilibria and thermodynamics with mass and heat transfer processes.