



Advances in Gas Processing

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Mohammed Jaber F. Al Marri | Fadwa T. Eljack

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Advances in Gas Processing

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4th International Gas Processing Symposium

Edited by

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List of Contents

Preface.....	xi
--------------	----

Carbon Management

Solubility of CO₂ in Aqueous Amine Solutions: a Study to Select Solvents for Carbon Capture from Natural-gas Power Plant

Jiafei Zhang, Danlu Tong, Paul S. Fennell, J. P. Martin Trusler 1

CO₂ Capture Using an Aqueous Formulated Solvent Containing Ethylaminoethanol, N- Methyl-2-Pyrrolidone, and Hydroxyl Radical Scavengers: Study of Solvent Degradation and Absorption Kinetics

Rahul R. Bhosale, Shahd S. Gharbia, Jamila Folady, Mehak Jilani, Dareen Dardor, Anand Kumar, Leo L. P. van den Broeke, Vijaykumar V. Mahajani, Sukanta Kumar Dash..... 11

Preparation of Polyethylenimine Impregnated Mesoporous Precipitated Silica for CO₂ Capture

Dang Viet Quang, Aravind V. Rayer, Nabil El Hadri and Mohammad R.M. Abu-Zahra 21

Thermodynamic Analysis of Solar Fuel Production via Thermochemical H₂O and/or CO₂ Splitting Using Tin Oxide Based Redox Reactions

Rahul R. Bhosale, Shiva Yousefi, Jamila Folady, Mehak Jilani, Dareen Dardor, Shahd S. Gharbia, Anand Kumar, Leo J. P. van den Broeke, Ivo Alxneit, Rajesh Shende 39

Synthesis of a New Cu-Aluminosilicate Catalyst for CO₂ Capture and Conversion to Hydrocarbons

Reyad Shawabkeh, Ibnehwaleed Hussein, Waqar Ahmad, Adeem Rana 49

Development of Amine-blend Systems for CO₂ Post-Combustion Capture

Nabil EL Hadri, Dang Viet Quang, Aravind V. Rayer, Mohammad R. M. Abu-Zahra 59

Gas Conversion

Single Step Synthesis of Transition Metal Nanoparticles in Aqueous Phase for Catalytic Applications

Anand Kumar, Anchu Ashok, Yussuf Olasunkanmi Kuti, Rahul R Bhosale, Mohd Ali H. Saleh, Leo J.P. van den Broeke..... 69

Modeling Fischer-Tropsch Product Distribution of a Cobalt-based Catalyst in Different Reaction Media

Shaik Afzal, Jan Blank, Rehan Hussain, Nimir O. Elbashir 81

A Comparative Study of the Effects of Water on Methane Oxidation over Pd@CeO₂/Al₂O₃ and Pd@TiO₂/Al₂O₃ Catalysts

Sardar Ali, Mohammed J. Al-Marri, Ahmed Gamal Abdul Monem, Yara I. Arafat, Mahmoud M. Khader 89

Conversion of Methane in the Liquid Compression Reactor

Maxim Glushenkov, Gerard Fuite, Theo van der Meer, Alexander Kronberg 99

Development of Nickel-based Catalysts for Methane Steam Reforming

Sardar Ali, M.M. Zagho, Mohammed J. Al-Marri, Yara I. Arafat, Mahmoud M. Khader 111

Energy Mix

Solar Fuel Production via Non-Stoichiometric CexZryHfzO2-δ Based Two-Step Thermochemical Redox Cycle

Rahul R. Bhosale, Mehak Jilani, Dareen Dardor, Shahd S. Gharbia, Jamila Folady, Anand Kumar, Leo L. P. van den Broeke Fangjian Lin, Ivo Alxneit..... 117

Investigation of Laminar Flame Speeds of Methane-LPG Air Mixtures

Ahmed S. Ibrahim, Mohammed A. Abdalwahab, Othman S. Abulaban, Samer F. Ahmed 125

Emerging Opportunities for Natural Gas Treatment and CO₂ Capture

Jaap F. Vente and Wim G. Haije 133

Thermochemical Conversion of CO₂ into Solar Fuels Using Ferrite Nanomaterials

<i>Rahul R. Bhosale, Dareen Dardor, Shahd S. Gharbia, Jamila Folady, Mehak Jilani, Anand Kumar, Leo L. P. van den Broeke, Fangjian Lin, Ivo Alxneit</i>	141
---	-----

Operational Excellence

Electrodeposition of Ni-B-Zn Alloy Coatings and their Characterization <i>R. A. Shakoor, Ramazan Kahraman, U. S. Waware, Yuxin Wang, Wei Gao</i>	149
---	-----

Qatargas GHG Management Strategy and Technology Assessment <i>Afzal A. Subedar, Khalifa Ahmed Al-Sulaiti, Rashid Mohammed Al-Rashdi, Ibrahim Bawazir, Muh Ilyasak</i>	159
--	-----

Removal of Metal Ions and Heat Stable Salts From Industrial Lean Amine Solvent Using Polymeric Hydrogels From Gas Sweetening Unit <i>Priyabrata Pal and Fawzi Banat</i>	173
--	-----

NGL Recovery Processes and Operating Issue- Case Study <i>Sunil P. Solanki and Ibrahim Al-Hassan</i>	185
---	-----

Flow Accelerated Corrosion (FAC) in boilers / HRSGs <i>Shashidhar V. and Patnaik, D</i>	197
--	-----

Gas Solubility in Aqueous Solutions Under Two-Phase (H-L _w) Hydrate Equilibrium Conditions <i>Ioannis N. Tsimpanogiannis, Ioannis G. Economou, Athanassios K. Stubos</i>	205
---	-----

Versatile Gas Monitoring System on the Heterogeneous Znyq SoC Platform <i>Amine Ait Si Ali, Abbes Amira, Faycal Bensaali, Mohieddine Benammar, Muhammad Hassan, Amine Bermak</i>	213
---	-----

A Front-end EPC Gen-2 Passive UHF RFID Transponder for Embedded Gas Sensor <i>Mohamed Zgaren and Mohamad Sawan</i>	223
---	-----

Membrane Gas Desorption for Natural Gas Treating <i>Annemieke van de Runstraat, Earl L.V. Goetheer, Daphne E. Bakker, Alexey V. Volkov, Abdelbaki Benamor</i>	233
--	-----

Olefin / Paraffin separation using Task-Specific Materials based on Ionic Liquids.

Johan Jacquemin, Yun Zheng, Yu Wang, Jingjing Zhou, Jillian Thompson, Mert Atilhan, David Rooney, Majeda Khraisheh 243

Environmental and Sustainability

Tracking of GHG Emissions and Tax Implication During Normal/Abnormal Situations – Ethylene Process Base Case Industrial Application

Fahd M. Mohammed, Monzure-Khoda Kazi, Ahmed M. AlNouss, Fadwa T. Eljack 251

Qatargas Flare Reduction Program

Mohsin Raja, Ibrahim Bawazir, Ihab Abdelmohsen, Khalid Bashir, and Khalifa Ahmed Al-Sulaiti 261

Adsorption of H₂S from Natural Gas using treated Oil Fly Ash

Zaheer Aslam, Reyad A. Shawabkeh, Ibnelwaleed A. Hussien I. 273

Treatment of Produced Water from Oil & Gas Operations by Membrane Distillation

Altaf Hussain, Joel Minier-Matar, Arnold Janson, Samer Adham 285

Application of Membrane Bioreactor Technology for Produced Water Treatment

Arnold F. Janson, Ana Santos, Altaf Hussain, Joel Minier-Matar, Simon Judd, Samer Adham 293

Qatargas LDAR Fugitive VOC Emissions Program

Farhan A. Khan, Khalifa A. Al Sulaiti, Khalid Bashir 301

Application of Forward Osmosis to Reduce Produced Water Injection Volumes

Joel Minier-Matar, Altaf Hussain, Arnold F. Janson, Anthony G. Fane, Samer Adham 309

Life Cycle Assessment of North American Shale Gases

Ian J. Laurenzi 317

Environmental Analysis of Different Scenarios of Power Generation in Iran with Natural Gas, Vacuum Residue and Syngas Feedstock <i>Mousa Meratizaman, Sina Monadizadeh, Majid Amidpour</i>	327
--	-----

Design and Modeling

Carbon Management: Regional Solutions Based on Carbon Dioxide Utilization and Process Integration <i>Anand Kumar, Anchu Ashok, Mohammed S. Shurair, Kutti Yussuf, Leo J.P. van den Broeke</i>	337
---	-----

Atomistic Simulations of Clathrate Hydrates <i>Joseph Costandy, Vasileios K. Michalis, Athanassios K. Stubos, Ioannis N. Tsimpanogiannis, Ioannis G. Economou</i>	351
--	-----

Molecular Thermodynamic Models for CO ₂ and Mixtures: Recent Developments and Applications for Process Design <i>Sally El Meragawi, Othonas A. Moultos, Ioannis N. Tsimpanogiannis, Nikolaos I. Diamantonis, Athanassios Z. Panagiotopoulos and Ioannis G. Economou</i>	361
--	-----

Design Philosophy GPC High Pressure Pilot Plant <i>Annemieke van de Runstraat, Erwin Giling, Abdelbaki Benamor, Earl L.V. Goetheer</i>	371
---	-----

Real-time Monitoring of Solvent Composition for Acid Gas Absorption Processes <i>Leen van der Ham, Andries van Eckeveld, Abdelbaki Benamor, Earl Goetheer</i>	379
---	-----

Modeling of Reaction Front Movement in Combustion Synthesis for Catalyst Preparation <i>Noura A. Dowass, Anchu Ashok, Yussuf Olasunkanmi Kuti, Mohammed Salah Shurair, Anand Kumar, Rahul R. Bhosale, Mohd Ali H. Saleh, Leo J. P. van den Broeke</i>	391
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Index	401
-------------	-----

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Preface

Natural gas has been a valuable energy commodity for many centuries. Natural is considered the fastest growing segment of the energy market. The increased share of natural gas in the energy market is generally attributed to its low environmental impact and convenience in utilization as a fuel or feedstock in a wide range of petrochemical industry. It is also a key player in enhancing the efficiencies of power generation system. The challenge of energy security and environment requires solutions that are within reach; and natural gas is a flexible energy source that has the potential to complement emerging alternative technologies. Qatar is considered is one of the world's pioneers in implementing new processing technologies on a very large scale and accumulated a significant experience in operational excellence; and as such Qatar is positioned as one of the leading LNG and GTL exporters in the world. The situation highlighted above lead to an inspired 4th international gas processing symposium covering the state-of-the-art advances in the area of natural gas utilization, sustainability and excellence in processing. The 4th international gas processing symposium held in Doha, Qatar from October 26 to 27, 2014.

A key future theme in the Symposium was Natural Gas and World's Energy Mix because of its growing importance of the global energy market. The Symposium also covered the following gas processing applications in parallel sessions:

- Environment Sustainability
- Carbon Management
- Energy Mix
- Gas Conversion Processes (GCP)
- Design & Modeling
- Operational Excellence I (OEI)
- Industrial Case Studies Session

The 4th international gas processing symposium combines an interesting variety of papers from academia and industry, giving a unique perspective into state of the art matters related to gas process which are presented in the chapters of this symposium proceedings book.

The organizing committee of the 4th international gas processing symposium wishes to express gratitude to people who made this event

possible and pleasant. Grateful acknowledgment is given to His Highness Sheikh Tamim bin Hamad Al-Thani, Emir of the State of Qatar. The support of her Excellency, Professor Misnad, President of Qatar University. The strong support of College of Engineering and the Gas processing Center staff at Qatar University is also gratefully acknowledged.

The organizing committee also wishes to extend their gratitude to the symposium sponsors: Qatar Petroleum, Dolphin Energy, OryxGTL, QAFAC, RasGas, ExxonMobil and professional partners: European Federation of Chemical Engineers (EFCE), Gas Processing Association (GPA)-GCC, American Institute of Chemical Engineers (AIChE), Elsevier (publisher), Industrial & Engineering Chemistry Research (I & EC Research)(Special Issue)

Finally, the editors would like to thank the authors who shared their knowledge and expertise in papers constituting the core content of this book.

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Solubility of CO₂ in aqueous amine solutions: a study to select solvents for carbon capture from natural-gas power plant

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Abstract

New solvents or solvent formulations are required for the capture of CO₂ from natural gas power plants, the flue gases from which typically contain low concentrations of CO₂ coupled with relatively high concentrations of O₂. Information concerning vapour-liquid equilibrium, in particular CO₂ loading capacity, is important for the selection of solvents to be employed for absorption. A static-analytic apparatus has been developed to acquire such data at temperatures between (313 and 393) K and various CO₂ partial pressures. A tertiary amine, 2-dimethylaminoethanol (DMAE) and its blended solutions with piperazine (PZ) were studied. The density and viscosity of these amine solutions were also measured to yield a better understanding of their performance in mass transfer and fluid dynamics.

Keywords: CO₂ absorption, amines, vapour-liquid equilibrium, density, viscosity

1. Introduction

Anthropogenic CO₂ emissions from the combustion of fossil fuels have caused a significant increase in atmospheric greenhouse-gas concentrations and are beginning to change the global climate [1]. CO₂ capture and storage (CCS) is proposed as an intermediate solution to mitigate global warming and to allow continued use of fossil fuels. Amine-based CO₂ absorption has hitherto been the technology of choice for post-combustion capture (PCC) from fossil-fuel power stations, but challenges such as high energy consumption and solvent degradation still impede the implementation of conventional solvent scrubbing technologies for flue gas treatment [2-4]. The use of natural gas for power generation has greatly increased in recent years for both economic and environmental reasons. As shown in Table 1, the flue gas from natural gas power systems has a lower partial pressure of CO₂ (p_{CO_2}) and a higher concentration of O₂ compared to that from coal-fired power plant [5]. It is therefore necessary to select gas-specific solvents adapted for capturing CO₂ from natural gas combined cycle (NGCC) power plant. Such solvents should present a high CO₂ capacity at low p_{CO_2} and a good chemical stability, especially with regards to oxidative degradation.

Table 1. Typical flue gas compositions from coal and natural gas combustion

(unit: %)	CO ₂	N ₂	O ₂	H ₂ O	NO _x	SO _x	Ar
Coal-fired	12-16	75-80	2-4	10-15	400 ppm	150 ppm	0.82
Gas-fired	3-5	70-75	10-12	7-10	<50 ppm	<10 ppm	0.89

2. Amines for CO₂ absorption

Amine solvents with their high capacity to absorb dilute CO₂ have been used commercially for gas sweetening. MEA is the most well-known alkanolamine and has been employed for the separation of acid gases in industrial processes for more than 80 years [6]. It is characterised by high absorption capacity, fast reaction kinetics, good aqueous miscibility and low cost. However, this solvent shows several disadvantages such as high energy requirement (3.5 to 4 GJ/ton-CO₂), solvent degradation, and corrosion issues. Therefore, it is essential to develop new amine solvents that meet the following criteria:

- Reduced energy consumption
- Fast reaction kinetics and high capacity with respect to CO₂
- Minimal environmental impact
- Low degradation and corrosivity

N-methyldiethanolamine (MDEA), as a tertiary amine, has been used for selective absorption of H₂S and CO₂ in many industrial processes [7]. Some MDEA-based solvents developed by Dow Chemicals were claimed to be more economic with regard to energy consumption than MEA-based solvents, primarily owing to the lower enthalpy of reaction [8]. MDEA also exhibits advantages of low volatility and excellent resistance against thermal and oxidative degradation. In addition, other tertiary amines *e.g.* 2-dimethylaminoethanol (DMAE) and 2-diethylaminoethanol (DEAE) were also proposed for PCC [9], but slow reaction kinetics prove to be one of the main shortcomings of this category.

Sterically hindered amines, *e.g.* 2-Amino-2-methyl-1-propanol (AMP), are considered as promising candidates for acid-gas scrubbing [10]. They present high CO₂ capacity and low degradability, but high vaporisation loss is a potential weakness. Other sterically hindered amines *e.g.* KS-1 commercialised by MHI [11] and 2-(isopropylamino)ethanol [12] were claimed to be superior compared to MEA in the PCC process.

Piperazine (PZ), a diamine with fast reaction kinetics, high CO₂ loading and good chemical stability, has also been recommended for PCC [13]. However, it has been mainly used as an absorption promoter in solvent formulations because of its limited aqueous solubility and high price. Additionally, other polyamines *e.g.* 2-(1-piperazinyl)-ethylamine (PZEA) [14] and Cansolv's proprietary solvent DC105 [15], have proved more effective for CO₂ capture from NGCC.

Blended amine solvents are proposed to combine the advantages of each component. The optimal recipe typically comprises a principal solvent with high net CO₂ capacity and an activator exhibiting fast reaction kinetics to improve the efficiency of both the absorption and regeneration processes. A successful example is activated MDEA (aMDEA) developed by BASF [16], which presents the merits of moderate energy requirement, excellent chemical stability and improved reactivity. In this work, we investigate two additional blended solvent systems: AMP+PZ and DMAE+PZ.

3. Experimental

A static-analytic apparatus, shown in Figure 1, has been employed for accurate VLE measurements on CO₂-amine-H₂O systems. The experiments were carried out in a 100 ml stainless steel vessel at various temperatures from (313 to 393) K and total pressures up to 1 MPa. Both the gas and liquid phases were analysed by a Perkin-Elmer Clarus 500 gas chromatograph (GC) to determine the coexisting-phase compositions. The apparatus was validated in

earlier work [17] but the GC analysis system with gas sampling valve was improved in this study.

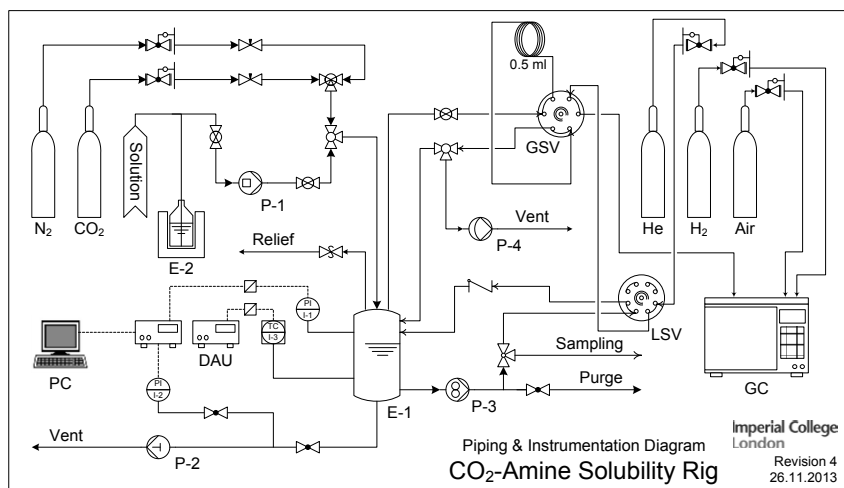


Figure 1. P&I Diagram of the static-analytic apparatus for VLE

DAU: data acquisition unit; E-1: jacketed reaction vessel; E-2: degasser; GC: gas chromatograph; GSV: gas sampling valve; LSV: liquid sampling valve; P-1: high pressure generator; P-2: rotary vane vacuum pump (0.01 kPa); P-3: gear pump; P-4: diaphragm vacuum pump (0.8 kPa); PC: computer.

MEA and PZ were purchased from VWR, and DMAE and AMP were supplied by Acros Organics. All of the tested amines were of at least 99 mass% purity. Aqueous amine solutions were prepared gravimetrically, and CO₂-saturated solutions were prepared from these by bubbling CO₂ through them at ambient pressure in a gas washing bottle. Partially loaded solutions were obtained by diluting CO₂-saturated solutions with fresh amine solution in the desired ratios. CO₂ loading (α_{CO_2}) was determined by GC analysis with an estimated uncertainty of 2%.

Density (ρ) was measured using an Anton Paar DMA 5000 M vibrating-tube densimeter in the temperature (T) range of (298 to 353) K. The uncertainty is estimated to be 0.05 kg/m³. Kinematic viscosity (ν) was determined using a certified U-tube capillary viscometer (PSL) with an estimated uncertainty of 0.3%. Dynamic viscosity (μ) was calculated from the relation $\mu = \nu \cdot \rho$.

4. Vapour-Liquid Equilibrium

The apparatus was initially validated by measuring the solubility of CO_2 in 30 wt% aqueous MEA solution at temperature between (313 and 393) K. As shown in Figure 2, our results are in satisfactory agreement with the literature data of Jou *et al.* [18]. Further measurements were carried out for the tertiary amine DMAE and blended DMAE+PZ solutions. Figure 3 illustrates the good solubility of CO_2 in DMAE-based solutions at 313 K and excellent CO_2 regenerability at 393 K. The partial replacement of DMAE with PZ improves the CO_2 solubility slightly.

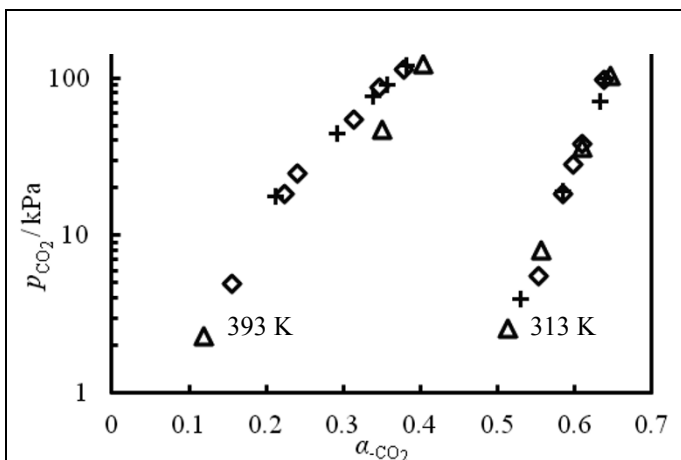


Figure 2. Solubility of CO_2 in 30 wt% MEA solution ($\diamond$ this work, $+$ Tong *et al.*, 2012, Δ Jou *et al.*, 1995).

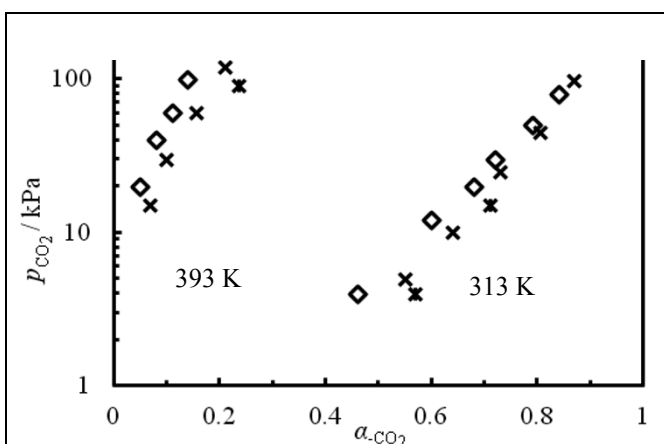


Figure 3. Solubility of CO_2 in 30 wt% DMAE ($\diamond$), 25 wt% DMAE+5 wt% PZ ($\times$) and 20 wt% DMAE+10 wt% PZ ($*$) solutions.

The cyclic loading capacity of CO₂ and alkalinity at low p_{CO_2} conditions were also studied to select specific amine solvents for NGCC-CCS. The proposed solvent formulations all have a higher net CO₂ capacity in comparison with the benchmark MEA and MDEA solvents (see Figure 4). Addition of the activator PZ in the solvent to replace part of the principle amine (AMP or DMAE) can not only accelerate the absorption rate but also improve the loading capacity, primarily due to the double amino function groups in a single molecule. Both the blended solvents AMP+PZ [19] and DMAE+PZ exhibit a good regenerability at the desorption condition.

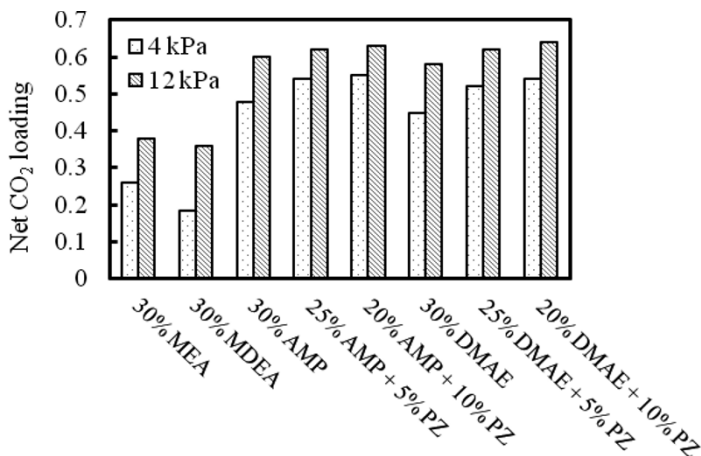


Figure 4. Comparison of net CO₂ capacity among the screened amine solvents.

5. Other thermophysical properties

To facilitate engineering calculations, it is essential to know the physical properties of solvents, and so properties such as the density and viscosity of aqueous amine solutions have been widely studied in the literature. However, very few literature data are available for CO₂-loaded aqueous amine systems. Our study therefore focuses on the effect of dissolved CO₂ on key thermophysical properties of aqueous amine solutions.

Density and viscosity are important for the process and plant design, especially relating to fluid dynamics calculations for the absorption column, gas-liquid mass transfer resistance and the selection of pumps and packings. The measurement systems were first validated with 30 wt% MEA solutions. As seen in Figure 5, the dynamic viscosity of MEA solutions is significantly increased by the dissolution of CO₂ and the measured data agree well with the values predicted from Weiland's correlation [20]. The maximum deviation is less than 3%.

Further measurements were conducted for DMAE and AMP solutions, and the effect of PZ addition and CO_2 loading was studied here. To suppress the temperature dependence of the data, the ratio of measured density ρ to the density of water (ρ_w) is presented in Figure 6, which illustrates that the ratio ρ/ρ_w is simply a linear function of T .

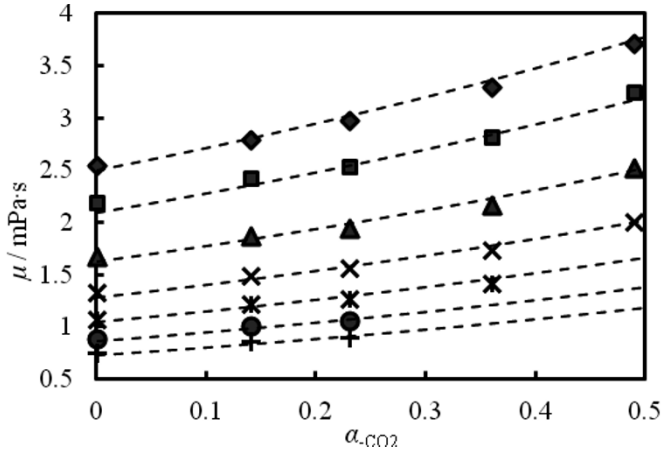


Figure 5. Dynamic viscosity of 30 wt% MEA solutions with various CO_2 loadings (◆ 298K, ■ 303K, ▲ 313K, × 323K, * 333K, ● 343K, + 353K, dashed lines: Weiland's correlation).

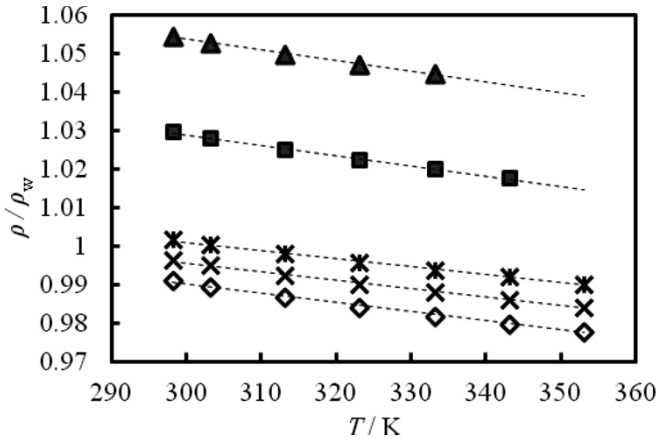


Figure 6. Density ratio of DMAE-based solutions to water with the influence of PZ addition and CO_2 dissolution (◇ 30 wt% DMAE, ■ 30 wt% DMAE + 0.8 mol/L CO_2 , ▲ 30 wt% DMAE + 1.4 mol/L CO_2 , × 25 wt% DMAE + 5 wt% PZ, * 20 wt% DMAE + 10 wt% PZ, dashed lines: linear fits).

The dynamic viscosity μ of 30 wt% DMAE, plotted in Figure 7, is much higher than the benchmark 30 wt% MEA. Both PZ addition and CO₂ loading enhance the viscosity, but the effect is quite weak in DMAE-based solutions. However, in the AMP-based solutions, μ is strongly enhanced by CO₂ dissolution (see Figure 8). In this case, both the DMAE- and AMP-based solutions can create a higher pressure drop along the absorption column compared to MEA; in addition, the packing at the bottom of the absorber should have a higher porosity than that at the top, since the viscosity of loaded AMP solution is almost double of that for the lean solution.

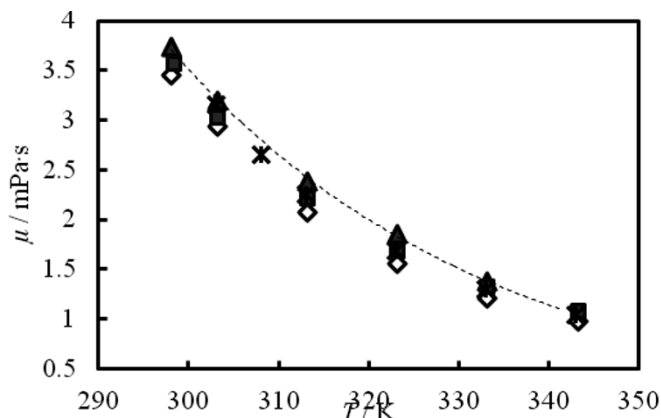


Figure 7. Dynamic viscosity of DMAE-based solutions ($\diamond$ 30 wt% DMAE, $\blacksquare$ 30 wt% DMAE + 0.8 mol/L CO₂, $\blacktriangle$ 30 wt% DMAE + 1.4 mol/L CO₂, $*$ 20 wt% DMAE + 10 wt% PZ, dashed line: exponential fit).

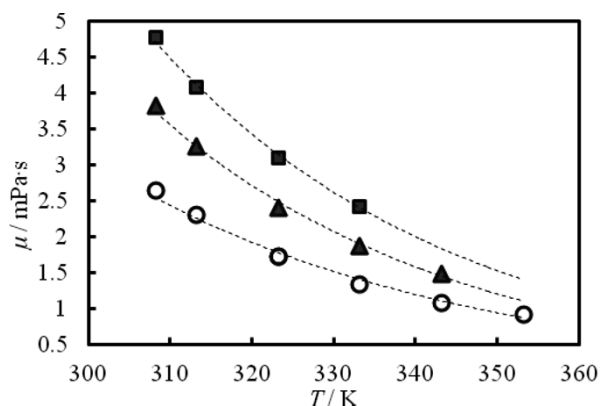


Figure 8. Dynamic viscosity of 20 wt% AMP+10 wt% PZ solutions ($\circ$ without CO₂, $\blacktriangle$ with 1.2 mol/L CO₂, $\blacksquare$ with 2.4 mol/L CO₂, dashed lines: exponential fits).