

Density and Viscosity of Partially Carbonated Aqueous Solutions Containing a Tertiary Alkanolamine and Piperazine at Temperatures between (298.15 and 353.15) K

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ABSTRACT: Measurements for the density and viscosity of partially carbonated solutions containing water, piperazine (PZ) and a tertiary amine, which was either dimethylaminoethanol (DMAE) or 2-diethylaminoethanol (DEAE), were conducted with total amine mass fractions of 30 % and 40 % over a temperature range from (298.15 to 353.15) K. Density and viscosity correlations of these mixtures were developed as functions of amine mass fraction, CO₂ loading and temperature. For both systems investigated, the average absolute relative deviations of the experimental data from these correlation are approximately 0.2 % for density and 3 % for viscosity. The correlations will be useful for thermodynamic analysis and computer simulations of carbon capture processes utilizing these promising blended amine systems.

KEYWORDS: carbon capture; carbon dioxide; density; diethylethanolamine; dimethylethanolamine; piperazine; viscosity.

INTRODUCTION

Recent and predicted future changes in the global climate due to anthropogenic greenhouse-gas emissions pose dangers to biological cycles and environmental systems.¹ The most significant emissions arising from combustion of fossil fuels. As fossil fuels are likely to remain dominant in the lowest-cost decarbonised global energy mix throughout the 21st century, it is important to implement carbon capture and storage (CCS) processes to curb CO₂ emissions.¹ Amine-based chemical absorption is the most mature technology to capture acidic gases and has been used extensively for gas sweetening since being patented more than 85 years ago.² However, solvents are not yet optimised for capturing CO₂ from the low-pressure flue gases of power plants and the energy penalty is still too great to make widespread industrial application economically viable³⁻⁵ with the current (low) prices for CO₂ emission. The energy penalty may be significantly reduced by selecting solvents that have a high CO₂ cyclic capacity and low or moderate energy requirements for regeneration. Aqueous solutions of 2-dimethylaminoethanol (DMAE) and 2-diethylaminoethanol (DEAE) have been identified as promising solvents, since they combine those properties.^{3, 6}

A fast rate of reaction with CO₂ is also beneficial, as this reduces absorber size and therefore capital cost. However, DEAE and DMAE are tertiary amines, which do not react directly with CO₂, but catalyse the hydration of CO₂ to form bicarbonates – a reaction that is relatively slow.⁷ However, the addition of small amounts of piperazine (PZ) as an activator enable a third parallel reaction path to form bicarbonate. CO₂ and PZ reacts rapidly to carbamate, which then transfers the CO₂ to the tertiary amine to form bicarbonate, while regenerating itself to PZ.⁷⁻⁹ Owing to the effective transport of CO₂ to the tertiary amine this process is referred to as the shuttle mechanism.⁹

The addition of small amounts of piperazine (PZ) as an activator was reported to enhance overall kinetics,¹⁰ as in parallel a faster carbamate reaction occurs. Firstly, PZ reacts rapidly with CO₂ to form a carbamate; secondly, the carbamate transfers CO₂ to the tertiary amine by the formation of an unstable zwitterion, releasing PZ; and finally, hydrolysis of the unstable zwitterion results in bicarbonate formation.¹⁰ Furthermore, tertiary amines also have a high chemical stability against thermal and oxidative degradation.¹¹ This is desirable as it keeps emission of possibly-toxic degradation products low³ and reduced operating expenses, owing to less solvent make-up being required. Finally, inexpensive production from renewable feedstocks makes DEAE and DMAE additionally attractive.⁷ However, accurate process simulation is necessary to predict the overall performance of different solvents in the capture and compression processes. This requires precise knowledge of chemical and thermophysical properties. Density and viscosity are both important for hydrodynamic calculations, which in turn are needed for efficient design of pumps and heat exchangers.¹² The solvent viscosity also influences solute diffusivities,¹³⁻¹⁵ which in turn affect overall CO₂ absorption rates in the column.

A few viscosity data are available for both unloaded and carbonated aqueous solutions containing (DEAE + PZ);¹⁶ however, these pertain to just two temperatures (303.15 K and

323.15 K) and to PZ mass fractions ≤ 7.5 %. For the (DMAE + PZ) system, neither density nor viscosity data are available. However, some publications provide data on partially-carbonated solutions containing either DMEA¹⁷ or DEAE^{6, 17-19} or PZ¹⁵. Given the lack of published data and of correlations that can be used over useful ranges of amine concentration and CO₂-loading, an experimental study was conducted to expand the available data and new empirical correlations were developed for modelling use.

EXPERIMENTAL DETAILS

Solvent Preparation and Determination of CO₂ loading

The chemical sample used are detailed in **Table 1**. Deionized (DI) water was obtained from a Millipore Direct (Q-UV) system that produced water of electrical resistivity $\geq 18 \cdot 10^6 \Omega \cdot \text{cm}$ at $T = 298.15 \text{ K}$. The water was degassed in an ultrasonic bath prior to use. Aqueous amines stock solutions were prepared gravimetrically in batches of 250 g using an electronic balance (Ohaus Pioneer, model PA 41) with a resolution of 0.001 g and glass bottles with a relatively narrow throat, which was covered with a lid whenever possible to prevent absorption of atmospheric CO₂. Owing to the relatively large batch size, the expanded uncertainty at 95 % confidence of the amine mass fractions was estimated to be 0.0002. This also takes the purities of the reagents into account.

The components studied in this work are numbered as follows: 1 = water, 2 = DEAE or DMAE, 3 = PZ, and 4 = CO₂. Definitions of the composition variables used here follow the work of Zhang et al.¹⁷ where mass fractions are defined on a CO₂-free basis as $w_i = m_i/(m_1+m_2+m_3)$, the CO₂ loading as $\alpha = n_4/(n_2+n_3)$ and the equivalent CO₂ molality as $b_4 = n_4/(m_1 + m_2 + m_3) = \alpha(w_2/M_2 + w_3/M_3)$, where m_i is the mass and n_i is the amount of component i . CO₂-saturated samples were obtained by bubbling CO₂ through the solvent in a gas washing bottle for a period of about 5 h, as described in the literature.^{6, 17, 20, 21} A heated bath maintained the solvent temperature at 303 K during saturation. The CO₂ that entered the gas-washing bottle was previously saturated with water vapour to suppress evaporation from the sample. Analysis of the saturated solutions indicated maximum loadings of between $\alpha = 0.62$ and $\alpha = 0.75$. Then, samples with four intermediate CO₂ loadings were prepared for each solvent composition by weighing and mixing amounts of loaded and unloaded solvent at $T = 303 \text{ K}$. All samples were kept in sealed bottles in the dark until needed, to prevent degassing. The CO₂ loading was determined before each experiment by gas

chromatograph (GC) with a Perkin Elmer (model Clarus 500) chromatograph calibrated as described by Tong et al.²² The estimated expanded uncertainty of the reported values is $U(\alpha) = \text{Max}(0.02, 0.04 \cdot \alpha)$.

Degassing during property measurement is a potentially significant issue that can bias experimental data. To determine if degassing occurred, the CO₂ loading prior and post property measurement was measured. If the ‘prior’ and ‘post’ values deviated by more than 0.01, it was assumed that degassing had occurred and the data were discarded. At the higher CO₂ loadings investigated, degassing was likely to occur before the highest experimental temperature was reached. Therefore, the CO₂ loading was additionally analysed between viscosity measurements. As a slight vertical CO₂ loading gradient across the viscometer was suspected in the case of degassing, the entire sample was withdrawn, cooled, carefully stirred and analysed in the GC; this, necessitated refilling the viscometer with fresh solvent before continuing measurements. All measurements were carried out at the ambient pressure of (0.101 ± 0.003) MPa.

Density Measurements

A vibrating tube densimeter (Anton Paar, DMA 5000) was used for density measurements. This instrument was calibrated using ambient air and pure degassed water at $T = 293.15$ K, as specified by the manufacturer. Expanded uncertainties at 95 % probability, as specified by the manufacturer, are $U(\rho) = 5 \cdot 10^{-6}$ g/cm³ for density and $U(T) = 0.01$ K for temperature. For an amine solution with mass fraction uncertainties of $U(w_2) = U(w_3) = 0.0002$, the overall expanded uncertainty of the density was estimated to be $U(\rho) = 1 \cdot 10^{-4}$ g·cm⁻³ at 95 % confidence. The equipment was validated by comparing experimental densities of aqueous solutions with DMAE mass fractions of 30 % with those from the literature¹⁷ and single point validations with air and

water were conducted prior to each measurement, as reported by others.^{17, 20} If bubble formation was observed, density measurements were stopped.

Viscosity Measurements

Kinematic viscosity measurements were conducted using a capillary U-Tube viscometer (PSL Rheotek, type BS/U size A), which was calibrated with degassed DI-water and a S20 viscosity standard (Paragon Scientific). A Julabo 18V oil bath with a Huber (CC-K6) chiller and a Julabo ME controller were used for temperature control. The temperature was monitored with a Fluke 5615 precision thermometer and the expanded uncertainty of the temperature was $U(T) = 0.01$ K. The overall expanded uncertainty of the viscosity was estimated to be $U(\eta) = \text{Max}(0.01 \text{ mPa}\cdot\text{s}, 0.02\eta)$.

RESULTS AND DISCUSSION

Experimental Data

The measurement system for density and viscosity was validated in our previous work¹⁷ by comparing experimental measurements for partially carbonated aqueous monoethanolamine solutions with data and correlations from the literature. In this work, additional check measurements were made on partially carbonated DMAE and DEAE solutions. The results were in good agreement with data from our earlier study¹⁷. The results of the density and viscosity measurements for the (DMAE + PZ) solvent system are presented in **Tables 2 and 3**, respectively, while results of density and viscosity measurements for the (DEAE + PZ) system are shown in **Tables 4 and 5**, respectively. It can be seen that the densities of both unloaded systems at lower temperatures are almost identical. The difference in density, for example, between the systems is

only 0.025 % at $T = 298.15$ K and with $w_2 = 0.2$ and $w_3 = 0.1$. An increase in temperature leads to a decrease of density in both systems. A temperature increase from 298.15 K to 353.15 K, for example, decreases the density of the unloaded systems with $w_2 = 0.2$ and $w_3 = 0.1$ containing DMAE and DEAE by 3.7 % and 3.8 %, respectively. Furthermore, it can be seen that the densities of both systems increase with a decrease in CO₂ loading. At $T = 298.15$ K and with $w_2 = 0.2$ and $w_3 = 0.1$, an increase from $\alpha = 0$ to $\alpha = 0.60$ increases the density of the system containing DMAE by 9.0 %, while an increase from $\alpha = 0$ to $\alpha = 0.56$ increases the density of the system containing DEAE by 7.0 %.

The viscosities of both unloaded systems at lower temperatures differs by less than 10 %. As expected, an increase in temperature decreases the viscosity of both systems significantly. For example, a change from 303.15 K to 353.15 K with $w_2 = 0.3$ and $w_3 = 0.1$ decreases the viscosity by 78 % and 79 % for the systems containing DMAE and DEAE, respectively. An increase in CO₂ loading leads to a significant increase in viscosity in both. Increasing the CO₂ loading of the system containing DMAE from $\alpha = 0$ to $\alpha = 0.46$, increased the viscosity by 45 %, while an increase in CO₂ loading of the system containing DEAE from $\alpha = 0$ to $\alpha = 0.53$, increased the viscosity by 62 %.

All correlations proposed in this work were developed with the objective of minimising the absolute average relative deviation ($\Delta_{\text{AARD},X}$) while also keeping the number of adjustable parameters reasonably small. The maximum absolute relative deviation ($\Delta_{\text{MARD},X}$) was also used as a measure of quality. These quantities were calculated as follows:

$$\Delta_{\text{AARD},X} = \frac{1}{N} \sum_{i=1}^N \frac{|X_{\text{exp}} - X_{\text{model},i}|}{X_{\text{exp}}} \quad (1)$$

$$\Delta_{\text{MARD},X} = \max\left(\frac{|X_{\text{exp}} - X_{\text{model},i}|}{X_{\text{exp}}}\right), \quad (2)$$

in which X denotes density or viscosity.

Density correlations

Density correlations must represent the non-ideality of the solvent solution, where the influences of effects such as the formation of hydration shells are accounted for. Partially carbonated solvent contains primarily reaction products of the initial species, such as CO_3^{2-} , $\text{PZ}(\text{COO}^-)_2$, PZCOO^- , HCO_3^- and DMEA^+ , which also influence density. Furthermore, the concentrations of those reaction products vary non-linearly with the mass-fraction of reagents and temperature.¹²

Weiland et al.¹² modelled the densities of partially carbonated solvents in terms of excess properties. Here, the total molar volume is modelled as the mole-fraction-weighted sum of the molar volumes of amines, water and dissolved CO_2 plus an excess molar volume function which represents deviations due to non-ideal mixing. Zhang et al.¹⁷ showed that this approach represents the densities of CO_2 -free solutions of either DMAE or DEAE very well. In principle, such an approach can easily be extended to solutions of more than one amine. However, PZ is a solid over the entire temperature range investigated and, while the sub-cooled liquid density can be estimated by extrapolation, this approach is not recommended. Therefore, we follow a simpler approach, similar to that proposed by Liu et al.²³ for CO_2 -free aqueous Ammonia and PZ, with a 6-parameters equation. The structure of the correlation method was slightly modified, by taking the density of water (ρ_w) into the denominator to minimise the effect of temperature, as suggested by Zhang et al.¹⁷ The resulting correlation has the following structure:

$$\rho_0/\rho_w = 1 + \frac{a_1 w_2 + a_2 w_3}{(T/T_0)} + \frac{a_3 w_2 + a_4 w_3}{(T/T_0)^2} + a_5 w_2 + a_6 w_3, \quad (3)$$

where ρ_0 and ρ_w are the densities of the unloaded amine and water, respectively, w_2 and w_3 are the mass fractions of the tertiary amine and piperazine, respectively, a_i with $i = 1$ to 6 are parameter that are specified in **Table 6**, T is the temperature and $T_0 = 298.15$ K. The density of water was calculated according to the following correlation based on literature data,²⁴

$$\rho_w /(\text{g} \cdot \text{cm}^{-3}) = 0.74017 + 0.59299 \cdot \left(\frac{T}{T_0}\right) - 0.33547 \cdot \left(\frac{T}{T_0}\right)^2. \quad (4)$$

This approach was found to have the lowest $\Delta_{\text{AARD},\rho}$ among all other 6 parameter equations investigated. As in our earlier work,¹⁷ the density of the loaded mixtures (ρ_L) was found to depend linearly on the product of CO₂ loading and amine mass fraction. Consequently, we represent the densities of partially carbonated solutions with the following correlation:

$$\rho_L = \rho_0(T, w_2, w_3)[1 + \alpha(c_1 w_2 + c_2 w_3)], \quad (5)$$

where ρ_L is the density of the loaded mixtures. The parameters c_1 and c_2 are presented in **Table 6**. The supporting information compares all density data with our model, while an example for each system is provided in **Figures 1** and **2**.

Viscosity correlations

Weiland et al.¹² studied viscosities of aqueous alkanolamine solutions and correlated them with an equation that contained an exponential function of temperature in the denominator – a variation of

the equation $\ln(\eta) = A + B/T$ that was originally proposed by Guzman²⁵ and Andrade.²⁶ However, Weiland et al.,¹² did not include the viscosity of pure water and used terms that did not contain amine mass fraction. The latter leads to deviations of calculated values for very dilute systems and therefore they had to restrict the correlation range to total amine mass fractions ≥ 0.2 . Zhang et al.¹⁷ reported that the viscosity of partially carbonated mixtures of water and tertiary amines, relative to that of pure water, could be well expressed by a modified Vogel-Fulcher-Tammann equation in which the parameters generally depend upon amine mass fraction and CO₂ loading. In order to extend this approach to the present blended amines systems, the viscosity is expressed as follows:

$$\ln \left[\frac{\eta(T, w_2, w_3, \alpha)}{\eta_w(T)} \right] = A(w_2, w_3, \alpha) + \frac{B(w_2, w_3, \alpha)}{(T/T_0) + C}, \quad (6)$$

where η and η_w are viscosities of the loaded mixtures and of water, respectively, T is the temperature, A and B are functions of the mass fractions, w_2 , w_3 , and of the CO₂ loading, α , and C is an independent parameter. The viscosity of pure water, η_w , was calculated from the simple equation,

$$\ln \left[\frac{\eta_w}{\text{mPa} \cdot \text{s}} \right] = -3.6957 + \frac{1.9011}{(T/T_0) - 0.4689}, \quad (7)$$

recommended by Zhang et al.¹⁷ The optimal structure and parameters for a correlation in the form of Eq. (6) were determined using a symbolic regression tool (Eureqa, Nutonian Inc.) that simultaneously searches for equation parameters and equation form, by scanning raw data for “meaningful and non-trivial invariants.”²⁷ The viscosity correlations developed in this way for the two mixtures containing DMAE and DEAE differ slightly. For the (DMAE + PZ) system, we have

$$\ln\left(\frac{\eta}{\eta_w}\right) = a_1 w_2 w_3 + a_2 w_3^3 + \frac{b_1 w_2 + b_2 w_3}{T/T_0 - c} + d_1 \alpha w_2 w_3 + d_2 \alpha w_2^2 + d_3 \alpha^2 w_3, \quad (8)$$

where w_2 and w_3 are the mass fractions of DMAE and PZ, respectively. For the (DMAE + PZ) system the correlation is

$$\ln\left(\frac{\eta}{\eta_w}\right) = a_1 w_2 w_3 + a_2 w_2^2 + \frac{b_1 w_2 + b_2 w_3}{T/T_0 - c} + d_1 \alpha w_2 w_3 + d_2 \alpha w_2^2 + d_3 \alpha^2 w_2, \quad (9)$$

where w_2 and w_3 are the mass fractions of DEAE and PZ, respectively. The parameters in Eqs. (8) and (9) are given in **Table 7**. Both correlations represent the experimental results reasonably well: for (DMAE + PZ), $\Delta_{\text{AARD},\eta} = 3 \%$ and $\Delta_{\text{MARD},\eta} = 12 \%$; while for (DEAE + PZ), $\Delta_{\text{AARD},\eta} = 2 \%$ and $\Delta_{\text{MARD},\eta} = 13 \%$. Generally, the larger deviations occur at high loadings, possibly because here small variations in α impact the sample viscosity significantly. However, no general pattern for the deviations can be found, indicating that those occur due to experimental variations rather than due to an inadequacy of the suggested correlations. We provide a full graphical comparison of our experimental data and the models in the supporting information and provide an example for the (DMAE + PZ) and (DEAE + PZ) systems in **Figures 3** and **4**, respectively.

As mentioned in the introduction, a few viscosity data exist in the literature for the partially carbonated solvents investigated in this work.^{6, 15-17} We will not compare our correlations to the viscosity data available for the carbonated single amine system containing only PZ¹⁵, since our correlations are based on experiments during which we always used significant amounts of a tertiary amine. In **Figures 5** and **6**, we compare the available literature data for partially-carbonated aqueous solutions containing either DMAE¹⁷ or DEAE^{6, 17} with the correlation

developed in this work and one can observe excellent agreement. In **Figure 7**, we compare recently published data for the partially-carbonated system DEAE+PZ¹⁶ with the correlations developed in this work. Our correlation represents their data at higher amine mass fractions well. However, the comparison is less favourable for lower amine mass fractions. Values for average and maximum deviations between all correlations and the experimental data are provided in **Table 8**.

CONCLUSIONS

We have measured the density and viscosity of partially carbonated aqueous amine solutions containing either DMAE or DEAE as the principal component and PZ as a promoter. The measurements were carried out under conditions such that $(w_2 + w_3) \leq 0.4$, $w_3 \leq 0.15$, $\alpha \leq 0.75$, and $298.15 \leq T/K \leq 353.15$. Compact empirical correlations were developed to represent the experimental data in this domain. The density correlations represent the experimental data for both unloaded and carbonated solutions, with Δ_{AARD} of 0.1 % for DMAE + PZ and 0.2 % for DEAE + PZ. To represent the viscosity data, a correlation based on a Vogel-Fulcher-Tammann equation was developed, with Δ_{AARD} of 3 % for DMAE + PZ and 2 % for DEAE +PZ. All correlations correctly model the effects of temperature, mass fraction and CO₂ loading across the ranges studied.

SUPPORTING INFORMATION

In the Supporting Information, we present graphical comparisons of the new experimental viscosity and density data with the correlation models developed in this work. We also show similar comparisons for the experimental data of Zhang et al.¹⁷ and Fu et al.¹⁶

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FIGURES

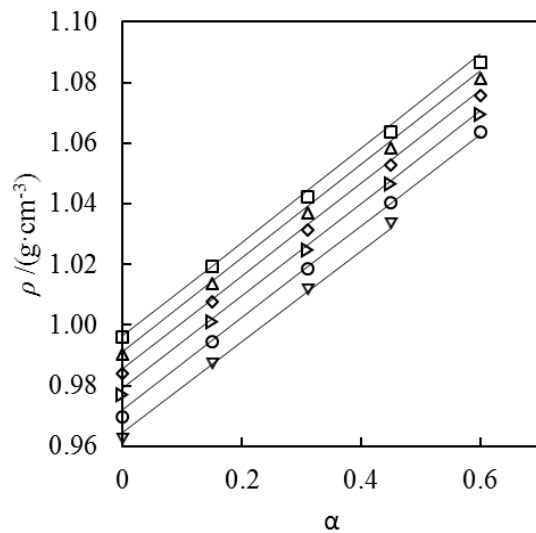


Figure 1. Density $\rho(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DMAE and PZ as a function of CO₂-loading α for CO₂-free mass fractions $w_2 = 0.2$ and $w_3 = 0.1$: □, $T = 303.15$ K; △, $T = 313.15$ K; ◇, $T = 323.15$ K; ▷, $T = 333.15$ K; ○, $T = 343.15$ K; ▽, $T = 353.15$ K; —, eq.(3).

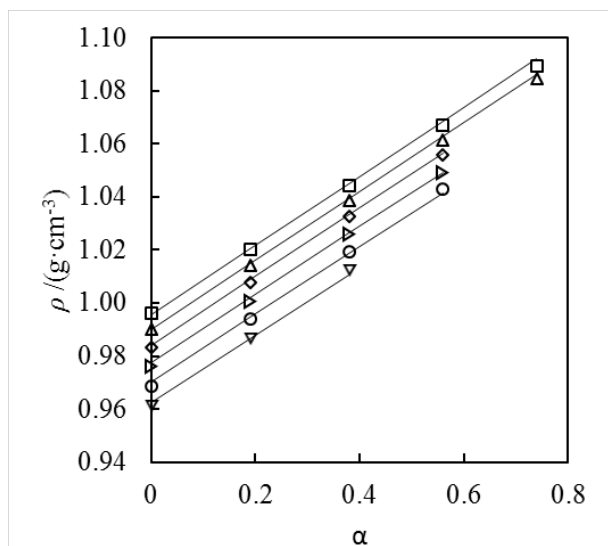


Figure 2. Density $\rho(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DEAE and PZ as a function of CO₂-loading α for CO₂-free mass fractions $w_2 = 0.20$ and $w_3 = 0.10$: □, $T = 303.15$ K; △, $T = 313.15$ K; ◇, $T = 323.15$ K; ▷, $T = 333.15$ K; ○, $T = 343.15$ K; ▽, $T = 353.15$ K; —, eq.(3).

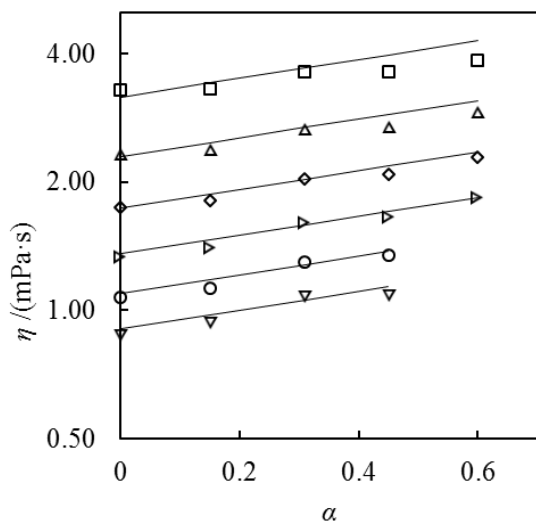


Figure 3. Viscosity $\eta(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DMAE and PZ as a function of CO₂-loading α for CO₂-free mass fractions $w_2 = 0.20$ and $w_3 = 0.10$: \square , $T = 303.15$ K; \triangle , $T = 313.15$ K; \diamond , $T = 323.15$ K; \triangleright , $T = 333.15$ K; \circ , $T = 343.15$; ∇ , $T = 353.15$ K; —, eq.(8).

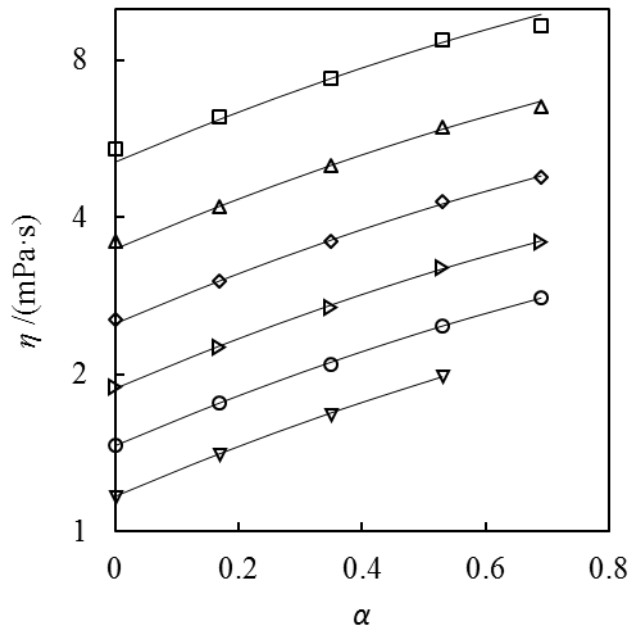


Figure 4. Viscosity $\eta(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DEAE and PZ as a function of CO₂-loading α for CO₂-free mass fractions $w_2 = 0.30$ and $w_3 = 0.10$: \square , $T = 303.15$ K; \triangle , $T = 313.15$ K; \diamond , $T = 323.15$ K; \triangleright , $T = 333.15$ K; \circ , $T = 343.15$; ∇ , $T = 353.15$ K; $-$, eq.(9).

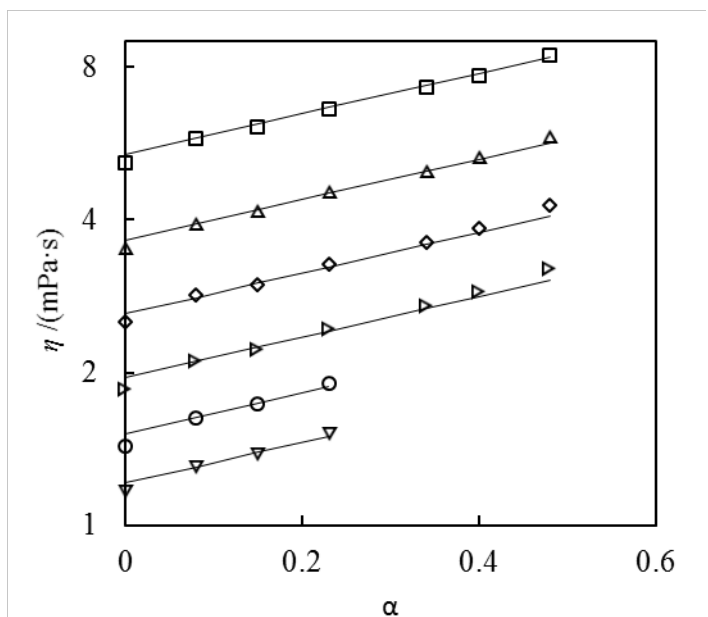


Figure 5. Viscosity $\eta(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DMAE as a function of temperature T , CO₂-loading α and CO₂-free weight fraction $w_2 = 0.45$.

Experimental data from Zhang et al.:¹⁷ □, $T = 303.15$ K; △, $T = 313.15$ K; ◇, $T = 323.15$ K; ▷, $T = 333.15$ K; ○, $T = 343.15$ K; ▽, $T = 353.15$ K; Correlation in this work: —, eq.(8).

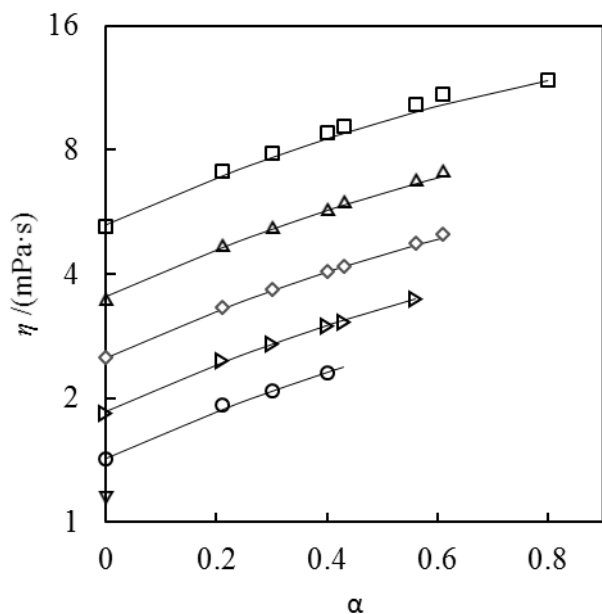


Figure 6. Viscosity $\eta(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DEAE as a function of temperature T and CO₂-loading α at a CO₂-free amine mass fraction $w_2 = 0.45$.

Experimental data from Zhang et al.:¹⁷ \square , $T = 303.15$ K; \triangle , $T = 313.15$ K; \diamond , $T = 323.15$ K; \triangleright , $T = 333.15$ K; \circ , $T = 343.15$ K; ∇ , $T = 353.15$ K; Correlation in this work: —, eq.(9).

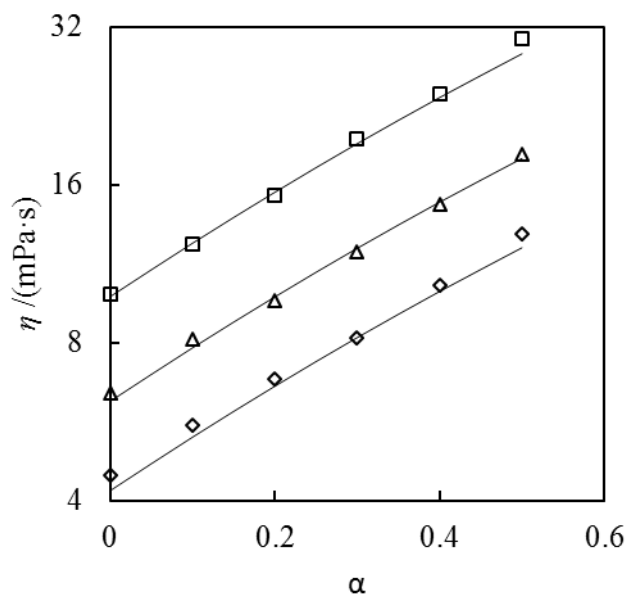


Figure 7. Viscosity $\eta(T, w_2, w_3, \alpha)$ of CO₂-loaded aqueous amine solutions containing DEAE and PZ as a function of temperature T and CO₂-loading α at CO₂-free amine mass fractions $w_2 = 0.4999$ and $w_3 = 0.0751$. Experimental data from Fu et al.:¹⁶ □, $T = 303.15$ K; △, $T = 313.15$ K; ◇, $T = 323.15$ K; Correlation in this work: —, eq.(9).

TABLES

Table 1. Description of chemical samples, where x denotes mole fraction.

Reagent	Purity	Supplier
N,N-(Dimethylamino)-ethanol	$x \geq 0.995$	Sigma-Aldrich
2-(Diethylamino)-ethanol	$x \geq 0.99$	Acrōs Organics
Piperazine	$x \geq 0.99$	Alfa-Aesar
Carbon dioxide	$x \geq 0.99995$	BOC
Water	Electrical resistivity > 18 M Ω ·cm at $T = 298$ K	Millipore Direct-Q UV3 apparatus
Viscosity Standard S20		Paragon Scientific

Table 2. Density ρ of partially carbonated aqueous solution containing DMAE and PZ at pressure $p = 0.101$ MPa, temperature T , DMAE mass fraction w_2 and PZ mass fraction w_3 on CO₂-free basis and CO₂ loading α and equivalent CO₂ molality b_3 .^a

			T / K	298.15	303.15	313.15	323.15	333.15	343.15	353.15
w_2	w_3	α	b_3 /(mol·kg ⁻¹)	$\rho / (\text{g} \cdot \text{cm}^{-3})$						
0.1000	0.1000	0.00	0.00	1.00195	0.99986	0.99478	0.98923	0.98305	0.97622	0.96875
		0.16	0.37	1.0173	1.01484	1.01006	1.00474	0.99906	0.99288	0.98624
		0.31	0.71	1.03221	1.03013	1.02562	1.02055	1.01495	1.0089	1.00242
		0.46	1.05	1.04722	1.04515	1.04061	1.03558	1.03004	1.02406	
		0.59	1.35	1.06017	1.05806	1.0535	1.04846	1.04296	1.03705	1.03072
0.1500	0.1500	0.00	0.00	1.00411	1.00137	0.99555	0.98929	0.9826	0.97551	0.96803
		0.13	0.45		1.02088	1.01537	1.00943	1.00306	0.9963	0.98916
		0.29	0.99		1.04438	1.03909	1.03343	1.02738	1.02093	1.01412
		0.44	1.51		1.06784	1.06278	1.05735	1.05152	1.04535	1.03878
		0.66	2.26		1.10084	1.09591	1.0906	1.08497	1.07896	1.07262
0.2000	0.1000	0.00	0.00	0.99890	0.99611	0.99018	0.98381	0.97702	0.96985	0.96229
		0.15	0.51	1.02184	1.0192	1.01362	1.00761	1.00117	0.99433	0.98713
		0.31	1.06	1.04491	1.04238	1.03705	1.03127	1.02506	1.01848	1.01155
		0.45	1.53	1.06621	1.06371	1.05847	1.05278	1.04671	1.04028	1.03341
		0.60	2.04	1.08899	1.08652	1.08129	1.0757	1.06976	1.0635	
0.2500	0.0500	0.00	0.00	0.99341	0.99057	0.98455	0.9781	0.97123	0.96398	0.95635
		0.16	0.54	1.01689	1.01415	1.00839	1.00218	0.99556	0.98855	0.98117
		0.32	1.08	1.04058	1.03788	1.03212	1.02594	1.01938	1.01248	1.00524
		0.45	1.52	1.0601	1.05732	1.05148	1.04529	1.03876	1.03193	1.02479
		0.60	2.03	1.08231	1.0796	1.07391	1.06792	1.06163	1.05504	
0.3000	0.0000	0.00	0.00	0.98798	0.98508	0.97895	0.97238	0.96541	0.95805	0.95031

		0.18	0.61	1.01383	1.01074	1.00424	0.99736	0.99015	0.98263	0.97483
		0.34	1.14	1.03776	1.03454	1.02782	1.0208	1.01352	1.006	0.99827
		0.49	1.65	1.06016	1.05693	1.05028	1.04336	1.03622	1.02882	1.02117
		0.72	2.42	1.09353	1.0908	1.08516				
0.2500	0.1500	0.00	0.00		0.99812	0.9911	0.98379	0.97616	0.96823	0.96
		0.15	0.68		1.02857	1.02216	1.01544	1.00842	1.00108	0.99344
		0.29	1.32		1.05869	1.05276	1.04651	1.03992	1.03304	1.02584
		0.45	2.05		1.08827	1.08261	1.07663	1.07034	1.06375	1.05686
		0.65	2.95		1.12697	1.12156	1.11589	1.10992		
0.3000	0.1000	0.00	0.00	0.99535	0.99193	0.98487	0.97751	0.96986	0.96188	0.95362
		0.16	0.72	1.02707	1.02393	1.01744	1.01065	1.00354	0.99612	0.9884
		0.31	1.40	1.05695	1.05399	1.04786	1.04139	1.0346	1.0275	1.02011
		0.46	2.08	1.08701	1.08409	1.07804	1.07171	1.06512	1.05825	1.0511
		0.61	2.76	1.11592	1.11314	1.10738	1.10137	1.09508		
0.3500	0.0500	0.00	0.00	0.98936	0.98592	0.97882	0.97141	0.9637	0.95569	0.94737
		0.15	0.68	1.01865	1.0154	1.00871	1.0017	0.99435	0.98673	0.9788
		0.3	1.35	1.04799	1.04475	1.03809	1.03112	1.02391	1.01647	1.00868
		0.45	2.03	1.0776	1.07434	1.06768	1.06082	1.05375	1.04645	1.03896
		0.6	2.07	1.10715	1.10411	1.09788	1.09128	1.08439		
0.4000	0.0000	0.00	0.00	0.9838	0.98032	0.97317	0.96572	0.95796	0.9499	0.94153
		0.16	0.72	1.01399	1.01037	1.00295	0.99531	0.98746	0.9794	0.97108
		0.31	1.39	1.04319	1.03949	1.03197	1.02427	1.01645	1.00846	1.00026
		0.45	2.02		1.06705	1.05966	1.05214	1.04451	1.03674	
		0.59	2.65		1.09495	1.08807	1.08107	1.07388		

^a Expanded uncertainties at 95% confidence are $U(T) = 0.02$ K, $U(p) = 0.003$ MPa, $U(w_2)=U(w_3)=0.0002$, $U(\alpha)=\text{Max}(0.02, 0.04 \cdot \alpha)$ for $\alpha > 0$, and $U(\rho) = 1 \cdot 10^{-4} \text{ g} \cdot \text{cm}^{-3}$.

Table 3. Viscosity η of partially carbonated aqueous solution containing DMAE and PZ at pressure $p = 0.101$ MPa, temperature T , DMAE mass fraction w_2 and PZ mass fraction w_3 on CO₂-free basis and CO₂ loading α and equivalent CO₂ molality b_3 .^a

			T / K	293.15	303.15	313.15	323.15	333.15	343.15	353.15
w_2	w_3	α	b_3 /(mol·kg ⁻¹)	η /(mPa·s)						
0.1500	0.1500	0.00	0.00		3.31	2.35	1.74	1.34	1.06	0.87
		0.13	0.45			2.59	1.97	1.51	1.22	0.99
		0.29	0.99			2.78	2.11	1.66	1.34	1.10
		0.44	1.51			2.97	2.31	1.81	1.48	1.22
		0.66	2.26			3.04	2.48	1.99	1.64	1.37
0.2000	0.1000	0.00	0.00	4.70	3.28	2.33	1.74	1.34	1.07	0.87
		0.15	0.51	4.82	3.31	2.39	1.81	1.41	1.13	0.93
		0.31	1.06	5.18	3.63	2.67	2.04	1.61	1.30	1.07
		0.45	1.53		3.62	2.70	2.08	1.66	1.35	1.08
		0.60	2.04		3.86	2.92	2.29	1.84		
0.2500	0.0500	0.00	0.00	4.56	3.08	2.20	1.65	1.28	1.02	0.88
		0.16	0.54	4.92	3.26	2.37	1.79	1.40	1.13	0.93
		0.32	1.08	4.88	3.57	2.66	2.01	1.59	1.28	1.06
		0.45	1.52		3.53	2.63	2.03	1.63	1.32	1.13
		0.60	2.03		3.77	2.84	2.23	1.78	1.46	
0.3000	0.0000	0.00	0.00		2.92	2.01	1.51	1.18	0.94	0.78
		0.18	0.61		2.96	2.15	1.63	1.28	1.03	0.85
		0.34	1.14		3.09	2.27	1.74	1.37	1.11	0.92
		0.49	1.65		3.20	2.38	1.84	1.46	1.19	
0.2500	0.1500	0.00	0.00			3.915	2.765	1.995	1.50	1.235
		0.15	0.68			4.64	3.28	2.47	1.90	1.51

		0.29	1.32			5.08	3.64	2.75	2.15	1.72
		0.45	2.05			5.87	4.33	3.31	2.59	2.09
		0.65	2.95			6.21	4.67			
0.3000	0.1000	0.00	0.00	8.37	5.24	3.53	2.52	1.88	1.46	1.14
		0.16	0.72	9.56	6.28	4.25	3.08	2.33	1.81	1.44
		0.31	1.40		6.44	4.63	3.38	2.57	2.02	1.62
		0.46	2.08		7.60	5.41	3.99	3.10	2.41	1.95
0.3500	0.0500	0	0.00	7.96	5.04	3.47	2.46	1.84	1.43	1.14
		0.15	0.68	8.72	5.42	3.75	2.73	2.06	1.60	1.29
		0.3	1.35	9.12	6.07	4.23	3.12	2.36	1.84	1.48
		0.45	2.03		6.62	4.70	3.46	2.66	2.10	1.68
		0.6	2.70		7.02	5.07	3.81	2.95		
0.4000	0.000	0	0.00		4.64	3.19	2.31	1.74	1.35	1.08
		0.16	0.72	7.57	4.95	3.44	2.51	1.90	1.48	1.20
		0.31	1.39		5.74	4.03	2.95	2.24		
		0.45	2.02	8.93	5.99	4.25	3.16	2.41		

^a Expanded uncertainties at 95% confidence are $U(T) = 0.02$ K, $U(p) = 0.003$ MPa, $U(w_2)=U(w_3)=0.0002$, $U(\alpha) = \text{Max}(0.02, 0.04 \cdot \alpha)$ for $\alpha > 0$, and $U(\eta) = \text{Max}(0.01, 0.02\eta)$.

Table 4. Density ρ of partially carbonated aqueous solution containing DEAE and PZ at pressure $p = 0.101$ MPa, temperature T , DEAE mass fraction w_2 and PZ mass fraction w_3 on CO₂-free basis and CO₂ loading α and equivalent CO₂ molality b_3 .^a

		T / K		298.15	303.15	313.15	323.15	333.15	343.15	353.15
w_2	w_3	α	b_3 /(mol·kg ⁻¹)	$\rho / (\text{g} \cdot \text{cm}^{-3})$						
0.1000	0.1000	0.00	0.00	0.99769	0.99629	0.99292	0.98879	0.98391	0.97828	0.97189
		0.17	0.34	1.01582	1.01362	1.00875	1.00332	0.99744	0.99108	0.98426
		0.33	0.66	1.03001	1.02788	1.02319	1.01798	1.01226	1.00605	0.9994
		0.50	1.01	1.04306	1.04096	1.03642	1.0313	1.02568	1.0196	
		0.61	1.23	1.05548	1.0534	1.04889	1.04384	1.03831	1.03234	
0.1500	0.1500	0.00	0.00		1.0015	0.99544	0.98892	0.98198	0.97464	0.96685
		0.16	0.48		1.02235	1.01661	1.01044	1.00385	0.99687	0.98951
		0.32	0.97		1.04373	1.03829	1.03244	1.02616	1.01949	1.01245
		0.47	1.42		1.06374	1.05861	1.05301	1.04703	1.04066	
		0.62	1.87		1.08353	1.07856	1.07314	1.06737		
0.2000	0.1000	0.00	0.00	0.99915	0.99621	0.98999	0.98331	0.97623	0.96877	0.96094
		0.19	0.54	1.02309	1.02021	1.01406	1.00772	1.00097	0.99383	0.98632
		0.38	1.09	1.0469	1.04426	1.03869	1.03269	1.02625	1.01942	1.01222
		0.56	1.61	1.06944	1.0669	1.06156	1.05579	1.04962	1.04309	
		0.74	2.12	1.09189	1.08948	1.08448				
0.2500	0.0500	0	0.00	0.99315	0.99005	0.98362	0.97677	0.96952	0.96189	0.95389
		0.19	0.52	1.01531	1.01235	1.0061	0.99943	0.99235	0.98489	0.97708
		0.49	1.33	1.05155	1.04861	1.04249	1.03597	1.02909	1.02189	1.01432
		0.54	1.47	1.05721	1.05433	1.04827	1.04183	1.03505	1.02796	1.02057
		0.75	2.04	1.08245	1.07996	1.0747	1.06906	1.06301		
0.3000	0.0000	0	0.00	0.98724	0.9841	0.97749	0.97047	0.96306	0.95526	0.9471
		0.19	0.49	1.0084	1.00502	0.99789	0.99036	0.98253	0.97441	0.96603
		0.36	0.92	1.02758	1.02402	1.01657	1.0088	1.00079	0.99261	0.98426

		0.52	1.33	1.04459	1.04106	1.03369	1.02603	1.01818	1.01018	
		0.75	1.92	1.07055	1.06773	1.06176	1.05544	1.04882		
0.2500	0.1500	0	0.00		0.9962	0.98891	0.98132	0.97343	0.96523	0.95673
		0.15	0.58		1.0219	1.01501	1.00783	1.00034	0.99255	0.98446
		0.31	1.20		1.0483	1.04182	1.03502	1.02792	1.02049	1.01275
		0.47	1.82		1.0759	1.06983	1.06343	1.05673	1.04969	1.04237
		0.61	2.36		1.10032	1.09459	1.08854	1.08218	1.07553	
0.3000	0.1000	0	0.00	0.99346	0.98989	0.98251	0.97485	0.96687	0.95858	0.95002
		0.17	0.63	1.02154	1.01809	1.01103	1.00372	0.99608	0.98815	0.97991
		0.35	1.30	1.05081	1.04758	1.0409	1.0339	1.02656	1.01894	1.011
		0.53	1.97	1.08015	1.07706	1.07068	1.06399	1.05701	1.04975	
		0.69	2.57	1.10581	1.10305	1.0973	1.09118	1.08475		
0.3500	0.0500	0	0.00	0.98696	0.98336	0.97591	0.96815	0.96007	0.95167	0.94296
		0.18	0.64	1.01479	1.01119	1.00388	0.99632	0.98845	0.98027	0.97179
		0.33	1.18	1.03741	1.03389	1.02664	1.0191	1.01128	1.00322	0.99487
		0.49	1.75	1.06229	1.05873	1.05143	1.04394	1.03628	1.02841	1.02029
		0.75	2.68	1.10288	1.09969	1.09376	1.08774	1.08169		
0.4000	0.0000	0	0.00	0.98018	0.97655	0.96901	0.96113	0.95294	0.94442	0.93557
		0.2	0.68	1.00947	1.00544	0.99724	0.98882	0.98026	0.97149	0.96252
		0.36	1.23	1.03276	1.02859	1.02013	1.01159	1.00297	0.99425	0.98539
		0.48	1.64	1.05066	1.04655	1.03815	1.0297	1.02123	1.01271	
		0.58	1.98	1.06487	1.06084	1.05269	1.04448	1.03626	1.02798	

^a Expanded uncertainties at 95% confidence are $U(T) = 0.02$ K, $U(p) = 0.003$ MPa, $U(w_2)=U(w_3)=0.0002$, $U(\alpha)=\text{Max}(0.02, 0.04 \cdot \alpha)$ for $\alpha > 0$, and $U(\rho) = 1 \cdot 10^{-4}$ g·cm⁻³.

Table 5. Viscosity η of partially carbonated aqueous solution containing DEAE and PZ at pressure $p = 0.101$ MPa, temperature T , DEAE mass fraction w_2 and PZ mass fraction w_3 on CO₂-free basis and CO₂ loading α and equivalent CO₂ molality b_3 .^a

			T / K	303.15	313.15	323.15	333.15	343.15	353.15
w_2	w_3	α	b_3 /(mol·kg ⁻¹)	η /(mPa·s)					
0.1000	0.1000	0.00	0.00	2.06	1.58	1.19	0.96	0.79	0.66
		0.17	0.34	2.13	1.61	1.25	1.02	0.84	0.70
		0.33	0.66	2.18	1.66	1.30	1.05	0.88	0.74
		0.5	1.01	2.23	1.71	1.37	1.12	0.91	
		0.61	1.23	2.27	1.79	1.43	1.16		
0.1500	0.1500	0.00	0.00	3.53	2.48	1.83	1.41	1.11	0.91
		0.16	0.48	3.78	2.72	2.01	1.55	1.24	1.02
		0.32	0.97	4.02	2.9	2.18	1.7	1.36	1.12
		0.47	1.42	4.27	3.12	2.38	1.87	1.50	
		0.62	1.87	4.53	3.34	2.58	2.03		
0.2000	0.1000	0.19	0.54	3.61	2.58	1.94	1.51	1.21	0.99
		0.38	1.09	3.93	2.86	2.18	1.70	1.37	1.12
		0.56	1.61	4.19	3.07	2.34	1.84	1.49	
		0.74	2.12	4.21	3.2				
0.2500	0.0500	0.00	0.00	3.22	2.27	1.69	1.31	1.05	0.86
		0.19	0.52	3.47	2.49	1.86	1.46	1.16	0.96
		0.49	1.33	3.95	2.86	2.16	1.68	1.35	1.11
		0.54	1.47	3.92	2.93	2.27	1.81	1.47	
		0.75	2.04	4.09	2.97	2.25	1.76		
0.3000	0.0000	0.00	0.00	3.00	2.13	1.60	1.25	1.00	0.83
		0.19	0.49	3.29	2.34	1.75	1.36	1.09	0.89

		0.36	0.92	3.53	2.51	1.88	1.46	1.17	
		0.52	1.33	3.73	2.67	2.00	1.55	1.24	
		0.75	1.92	3.81	2.82	2.17	1.71		
0.2500	0.1500	0.00	0.00	3.53	2.48	1.83	1.41	1.12	0.9
		0.15	0.58	6.63	4.45	3.17	2.35	1.82	1.44
		0.31	1.20	7.59	5.13	3.67	2.74	2.14	1.69
		0.47	1.82	8.89	6.05	4.35	3.26	2.53	
		0.61	2.36	9.85	6.76	4.92	3.73		
0.3000	0.1000	0.00	0.00	5.40	3.60	2.55	1.90	1.46	1.16
		0.17	0.63	6.21	4.20	3.02	2.26	1.76	1.40
		0.35	1.30	7.38	5.01	3.60	2.69	2.09	1.67
		0.53	1.97	8.75	5.95	4.28	3.21	2.48	1.97
		0.69	2.57	9.29	6.52	4.76	3.60	2.80	
0.3500	0.0500	0.00	0.00	4.94	3.33	2.40	1.79	1.39	1.12
		0.18	0.64	5.74	3.90	2.86	2.14	1.66	1.33
		0.33	1.18	6.59	4.49	3.24	2.43	1.88	1.51
		0.49	1.75	7.59	5.15	3.68	2.75	2.13	1.7
		0.75	2.68	8.46	5.93	4.38	3.35		
0.4000	0.0000	0	0.00	4.51	3.08	2.23	1.76	1.45	1.2
		0.2	0.68	5.34	3.63	2.62	1.97	1.52	1.21
		0.36	1.23	6.12	4.14	2.98	2.24	1.73	1.39
		0.48	1.64	6.62	4.51	3.22	2.41	1.85	
		0.58	1.98	7.30	4.94	3.50	2.59		

^a Expanded uncertainties at 95% confidence are $U(T) = 0.02$ K, $U(p) = 0.003$ MPa, $U(w_2)=U(w_3)=0.0002$, $U(\alpha) = \text{Max}(0.02, 0.04 \cdot \alpha)$ for $\alpha > 0$, and $U(\eta) = \text{Max}(0.01, 0.02\eta)$.

Table 6. Parameter for density correlations

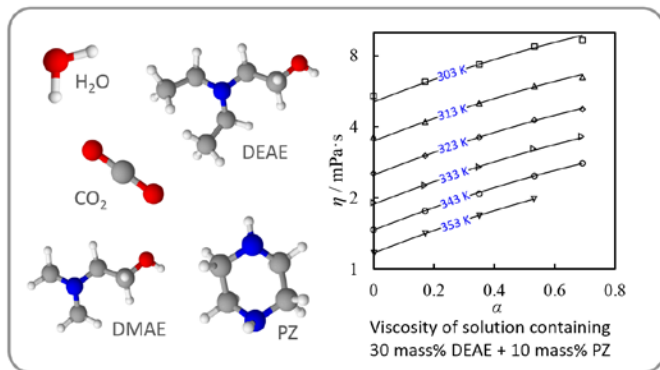
System	a_1	a_2	a_3	a_4	a_5	a_6	c_1	c_2
DMAE+PZ	0.09625	0.06439	0.10848	0.06439	-0.24454	-0.05322	0.49722	0.56252
DEAE+PZ	0.09590	0.11027	0.10353	0.11459	-0.25102	-0.13960	0.37759	0.55387

Table 7. Parameter for viscosity correlations

System	a_1	a_2	b_1	b_2	c	d_1	d_2	d_3
DMAE+PZ	5.44842	20.8208	1.28896	1.20257	0.713037	16.1799	4.54780	0.244159
DEAE+PZ	1.27764	-0.983822	1.46521	1.62252	0.700887	18.2417	6.87725	-1.07296

Table 8. Average and maximum deviations for density and viscosity correlations.

System	work	density correlations (CO ₂ -free)		density correlation (overall)		viscosity correlation (overall)	
		$\Delta_{AARD,\rho}$	$\Delta_{MARD,\rho}$	$\Delta_{AARD,\rho}$	$\Delta_{MARD,\rho}$	$\Delta_{AARD,\eta}$	$\Delta_{MARD,\eta}$
DMAE+PZ	This work	0.12%	0.30%	0.12%	0.49%	2.9%	11.9%
	Jiafei et al. ¹⁷	0.12%	0.41%	0.13%	0.43%	2.3%	11.7%
DEAE+PZ	This work	0.18%	0.51%	0.17%	0.59%	2.1%	12.8%
	Jiafei et al. ¹⁷	0.26%	0.71%	0.31%	0.99%	1.4%	5.6%
	Fu et al. ¹⁶	-	-	-	-	7.6%	16.1%



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