

Viscosity Measurement and Correlation of Unloaded and CO₂-Loaded Aqueous Solutions of *N*-Methyldiethanolamine + 2-Amino-2-methyl-1-propanol

Nithin B. Kummamuru, Dag A. Eimer, and Zulkifli Idris*

Cite This: *J. Chem. Eng. Data* 2020, 65, 3072–3078

Read Online

ACCESS |



Metrics & More

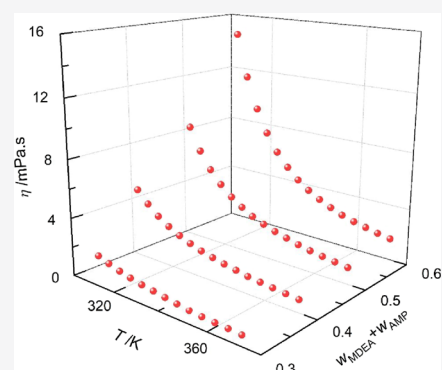


Article Recommendations



Supporting Information

ABSTRACT: This work contributes to new and complementary experimental viscosity data for blended amine mixtures of aqueous *N*-methyldiethanolamine + 2-amino-2-methyl-1-propanol (MDEA + AMP) solutions with and without CO₂ at different temperatures and mass fractions. For the unloaded MDEA + AMP solutions, measurements were conducted with total amine mass fractions ranging from 0.30 to 0.60. In the case of CO₂-loaded aqueous MDEA + AMP solutions, experiments were performed at CO₂ loadings ranging from 0.11 to 0.80. Proposed correlations were used to represent viscosity at the unloaded and CO₂-loaded solutions within experimental uncertainty.



1. INTRODUCTION

The chemical absorption process, also known as amine scrubbing using aqueous alkanolamines, is a well-known technology and is applied in removing CO₂ and/or H₂S from gas. The conventionally used primary, secondary, and tertiary amines in industry are monoethanolamine (MEA), diethanolamine (DEA), and *N*-methyldiethanolamine (MDEA), respectively.^{1,2}

The concept of blending alkanolamines has received attention as they provide better CO₂ capture efficiency than a single absorbent.^{2–5} A number of amine blends have been proposed and investigated.^{6–9} It was also reported by Idem et al.¹⁰ and Aaron and Tsouris¹¹ that the heat duty for the blended system was much lower compared to that of the single amine system, inferring that the use of blended amine system in industry for CO₂ capture is economically appealing.

The tertiary amine MDEA is considered to be an important amine for removal of CO₂ from gas streams because of its properties such as less energy requirement for amine regenerating due to low heat of reaction with CO₂, good resistance to chemical and thermal degradation, and high capture capacity (1 mol of CO₂/1 mol of amine).^{1,12–14} Sterically hindered amine such as 2-amino-2-methyl-1-propanol (AMP), which also approaches the CO₂ loading value equal to that of MDEA, is also considered to be a potential and attractive solvent for acid gas removal due to its resistance to degradation and formation of unstable carbamates.^{15–19}

Considering the advantages of MDEA and AMP in obtaining lower absorption enthalpy, faster absorption kinetics,

lower solvent regeneration requirement, and higher loading capacity, this work reports the viscosity data of blended aqueous solutions of MDEA and AMP, which can be considered as an attractive solvent for the removal of CO₂ coupled with satisfactory stripping characteristics.

Good and reliable physicochemical data of unloaded and CO₂-loaded aqueous alkanolamine solution are important in designing CO₂ capture equipment and developing kinetic and equilibrium models. Previously reported viscosity data of AMP and MDEA + AMP solutions are tabulated in Table 1.^{20–34} However, from the past works, it is found that the viscosity values of CO₂-loaded MDEA + AMP solutions are scarce and viscosity values of aqueous MDEA + AMP solutions are only available at limited compositions and temperatures. The present paper reports new viscosity data for aqueous MDEA + AMP in unloaded and CO₂-loaded systems together with the available literature data.

It is essential to have reliable correlations for accurate representation of experimental data. In this work, viscosities of unloaded aqueous MDEA + AMP solution were correlated as a function of temperature.³⁵ Furthermore, viscosities of CO₂-

Received: January 23, 2020

Accepted: May 7, 2020

Published: May 18, 2020

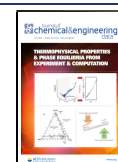


Table 1. Details of Previous Viscosity Work on AMP and MDEA + AMP Solutions Reported in the Literature

source	instrument used	concentration covered (mass %)	temperature covered (K)	CO ₂ -loaded
Pure and aqueous 2-amino-2-methyl-1-propanol (AMP)				
Khan et al. ²⁰	Ostwald viscometer	10 to 30	298.15 to 313.15	no
Li and Lie ²¹	Cannon-Fenske viscometer	20, 30, and 100	303.15 to 353.15	no
Mandal et al. ²²	Ostwald viscometer	30 and 100	293.15 to 353.15	no
Xu et al. ²³	Cannon-Fenske viscometer	17 and 26	296.75 to 349.85	no
Samanta and Bandyopadhyay ²⁴	Ostwald viscometer	30	298 to 333	no
Samanta and Bandyopadhyay ²⁵	Ostwald viscometer	30	313	no
Xiao et al. ²⁶	Cannon-Fenske viscometer	13 and 15	303.15 to 313.15	no
Mandal et al. ²⁷	Ostwald viscometer	30	313	no
Dash et al. ²⁸	Ostwald viscometer	40	303 to 323	no
Henni et al. ²⁹	Cannon-Ubbelohde viscometers	20 to 100	298.15 to 343.15	no
Álvarez et al. ³⁰	Schott-Geräte AVS 350 automatic Ubbelohde viscometer	50 and 100	298.15 to 323.15	no
Welsh and Davis ³¹	Cannon-Fenske viscometer	50	283.15 to 333.15	no
Ghulam et al. ³⁴	Ubbelohde viscometer	5 to 100	303.15 to 333.15	no
Aqueous <i>N</i> -methyl-diethanolamine (MDEA-W ₁) + 2-amino-2-methyl-1-propanol (AMP-W ₂)				
Shokouhi et al. ¹⁴	falling weight viscometer	W ₁ , 40; W ₂ , 5	303.15 to 363.15	yes
Huang et al. ³²	Cannon-Fenske viscometer	W ₁ , 10; W ₂ , 0.08 to 2	303.15 to 313.15	no
Welsh and Davis ³¹	Cannon-Fenske viscometer	W ₁ , 5 to 25; W ₂ , 5 to 25	283.15 to 333.15	no
Haratipour et al. ³³	falling weight viscometer	W ₁ , 40; W ₂ , 5	303.15 to 363.15	yes

Table 2. Chemicals Used in This Work^a

chemical name	CAS number	mole fraction purity ^a	source	purification
<i>N</i> -methyl-diethanolamine (MDEA)	105-59-9	≥0.99	Sigma Aldrich	degas
2-amino-2-methyl-1-propanol (AMP)	124-68-5	≥0.99	Sigma Aldrich	degas
carbon dioxide (CO ₂)	124-38-9	0.99999	AGA Norge AS	no
nitrogen (N ₂)	7727-37-9	0.99999	AGA Norge AS	no
sodium hydroxide (NaOH)	1310-73-2	N/A	Merck KGaA	no
hydrochloric acid (HCl)	7647-01-0	N/A	Merck KGaA	no
barium chloride dihydrate (BaCl ₂ ·2H ₂ O)	10326-27-9	≥0.99	Merck KGaA	no

^aAs stated by the supplier. N/A: not available.

loaded aqueous MDEA + AMP solutions were correlated using a modified version of the Setchenow equation.^{36,37}

2. MATERIALS AND METHODS

Table 2 reports the chemicals used in this work. All the chemicals in this work are of analytical grade and were used as they came without any further purification. Degassed MDEA, AMP, and Milli-Q water (resistivity, 18.2 MΩ·m) were used for sample preparation, and all the samples were prepared gravimetrically. High-purity CO₂ gas at a flow rate of 0.15 L·min⁻¹ was bubbled into the unloaded amine solutions to prepare CO₂-loaded amine solutions. For each of the unloaded aqueous MDEA + AMP solution studied in this work, a set of CO₂-loaded samples with different loading values (mole of CO₂/mole of MDEA + AMP) were prepared by varying the CO₂ bubbling time. An acid–base titration method as explained in previous publications was used to determine the actual values of CO₂ loading in the solutions.^{38,39}

Viscosities of unloaded and CO₂-loaded MDEA + AMP aqueous solutions were measured at temperatures between 298.15 and 373.15 K, as these cover the typical temperature range for CO₂ absorption and desorption processes in industry,^{1,40} using an Anton-Paar Physica MCR 101 rheometer (part number 16101) with a double-gap pressure cell XL (DG35.12/PR, measuring cell serial number 80462200).

Our earlier publication provides a thorough description on the viscosity measurement in our laboratory.⁴¹ Prior to actual

experiments, routine air checks and motor adjustments were carried out as suggested by Anton-Paar. The rheometer was also calibrated using a standard viscosity solution S3S from Paragon Scientific Ltd. at temperatures between 273.15 and 373.15 K. The reported experimental values in this work were corrected against the calibrated values. The combined standard experimental uncertainties for the unloaded and CO₂-loaded systems are calculated as 0.10 and 0.15 mPa·s, respectively, corresponding with our recent publication.⁴²

3. RESULTS AND DISCUSSION

This section is organized into three parts. The first part reports viscosity of aqueous AMP solutions to validate the measurement system. In the second part, experimental viscosities of unloaded aqueous MDEA + AMP solutions are reported together with one model to correlate the data. Subsequently, viscosities of CO₂-loaded aqueous MDEA + AMP solutions and one correlation method are discussed in Section 3.3. The average absolute deviation (Ω) values, as defined in eq 1, were calculated to estimate the performance of the models used for representing data,

$$\Omega \text{ (mPa}\cdot\text{s)} = \frac{1}{N} \sum_{i=1}^N |\eta_i^E - \eta_i^C| \quad (1)$$

where N , η_i^E , and η_i^C refer to number of data, experimental, and calculated viscosities, respectively.

3.1. Viscosity of Aqueous AMP Solution. The viscosity of aqueous AMP solutions from this work (Table S1) and the literature at five different concentrations is shown in Figure 1.^{21,22,24,30,31} Table 3 shows the Ω between experimental data

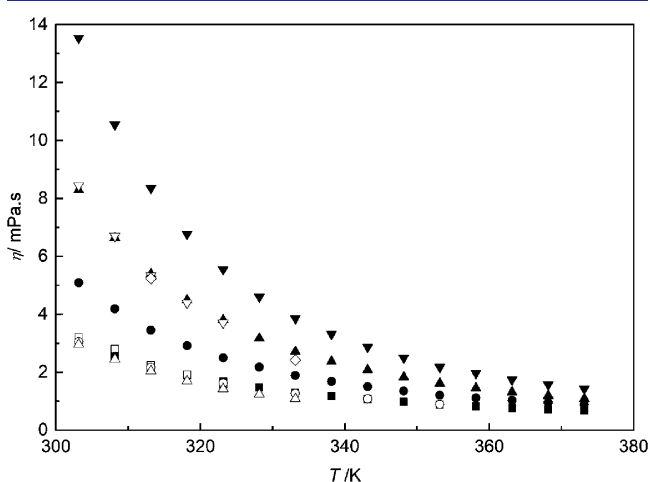


Figure 1. Viscosities of aqueous AMP solutions at temperatures 303.15 to 373.15 K. Symbols: this work, x_{AMP} , 0.079 (solid square); this work, x_{AMP} , 0.118 (solid circle); this work, x_{AMP} , 0.168 (solid up-pointing triangle); this work, x_{AMP} , 0.232 (solid down-pointing triangle); Mandal et al.,²² x_{AMP} , 0.079 (open square); Li and Lie,²¹ x_{AMP} , 0.079 (open circle); Samanta et al.,²⁴ x_{AMP} , 0.079 (open up-pointing triangle); Álvarez et al.,³⁰ x_{AMP} , 0.168 (open down-pointing triangle); Welsh et al.,³¹ x_{AMP} , 0.168 (open diamond).

Table 3. Average Absolute Deviation (Ω) of Viscosity between This Work and the Literature for Aqueous AMP Solutions

aqueous AMP solution		Ω (mPa·s)
reference		
	$w_{\text{AMP}}/x_{\text{AMP}}$, 0.3/0.079	
Mandal et al. ²²		0.11
Li and Lie ²¹		0.02
Samanta and Bandyopadhyay ²⁴		0.18
	$w_{\text{AMP}}/x_{\text{AMP}}$, 0.5/0.168	
Álvarez et al. ³⁰		0.09
Welsh and Davis ³¹		0.22

from this work and that of the literature for aqueous AMP solutions. As can be seen, the small Ω differences between this work and the corresponding values from the literature indicate that our instrument is functioning well and would be expected to give reliable experimental data. The minimal deviations between experimental and literature values could be due to several factors such as purity of chemicals and different equipment used for measurement.

3.2. Viscosity of Unloaded Aqueous MDEA + AMP Solutions. The viscosity of unloaded aqueous MDEA + AMP solutions was measured at different mass fractions of MDEA + AMP at temperatures ranging from 303.15 to 373.15 K. The experimental results for unloaded aqueous MDEA + AMP solutions are shown in Table 4. Figure 2 shows the temperature dependence of the viscosities of unloaded aqueous MDEA + AMP solutions. The viscosity of the blended amine system increased with an increase in amine concentration. However, at a given mass fraction of MDEA and AMP ($w_{\text{MDEA}} + w_{\text{AMP}}$), viscosity decreased with an increase in temperature,

Table 4. Viscosity η of Unloaded Aqueous MDEA + AMP Solutions at Different Mole Fractions of x_{MDEA} and x_{AMP} at Temperatures Ranging from 303.15 to 373.15 K^{a,b}

T (K)	η (mPa·s)			
	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.15/0.15;	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.20/0.20;	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.25/0.25;	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.30/0.30;
	$x_{\text{MDEA}}/x_{\text{AMP}}$, 0.030/0.040	$x_{\text{MDEA}}/x_{\text{AMP}}$, 0.045/0.060	$x_{\text{MDEA}}/x_{\text{AMP}}$, 0.064/0.086	$x_{\text{MDEA}}/x_{\text{AMP}}$, 0.089/0.012
303.15	2.88	4.78	8.27	14.53
308.15	2.46	3.96	6.65	11.38
313.15	2.11	3.30	5.41	9.04
318.15	1.85	2.81	4.50	7.32
323.15	1.63	2.40	3.77	5.98
328.15	1.46	2.10	3.22	4.98
333.15	1.30	1.84	2.77	4.18
338.15	1.19	1.65	2.43	3.58
343.15	1.08	1.48	2.15	3.09
348.15	0.98	1.33	1.91	2.69
353.15	0.88	1.19	1.69	2.34
358.15	0.82	1.10	1.53	2.09
363.15	0.79	1.02	1.39	1.89
368.15	0.76	0.94	1.27	1.70
373.15	0.71	0.87	1.16	1.54

^aThe operating pressure was maintained by N_2 gas ($p = 400$ kPa) throughout the temperature range. ^bStandard uncertainties u are $u(w_{\text{MDEA}+\text{AMP}}) = 0.01$, $u(T) = 0.03$ K, and $u(P) = 0.2$ kPa. The combined standard uncertainty for viscosity measurement $u_c(\eta)$ is 0.10 mPa·s.

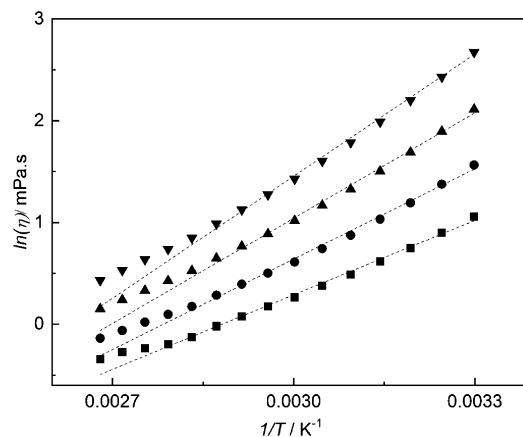


Figure 2. Experimental viscosities of unloaded aqueous MDEA + AMP solutions compared with calculated values from eq 2. Symbols: $w_{\text{MDEA}}/w_{\text{AMP}}$, 0.15/0.15 (solid square); $w_{\text{MDEA}}/w_{\text{AMP}}$, 0.20/0.20 (solid circle); $w_{\text{MDEA}}/w_{\text{AMP}}$, 0.25/0.25 (solid up-pointing triangle); $w_{\text{MDEA}}/w_{\text{AMP}}$, 0.30/0.30 (solid up-pointing triangle). Dashed lines refer to calculated viscosity values from using eq 2.

inferring that the force of attraction between MDEA, AMP, and water molecules decreases with an increase in kinetic energy.

Besides the experiments, a model that can correlate the experimental viscosities is also important. As the viscosity values for unloaded aqueous MDEA + AMP solutions at all the concentrations of amine concentrations decreased nonlinearly with increasing temperature, a logarithmic function model as shown in eq 2 can be used to fit the viscosity data^{35,43}

$$\ln(\eta_{\text{mix}}/\text{mPa}\cdot\text{s}) = A + B/T \quad (2)$$

where η_{mix} refers to the viscosity of the unloaded mixture, T is the temperature in K, and A and B are empirical parameters with the values presented in Table 5. The calculated viscosities

Table 5. Fitting Parameters A and B for Correlation Model, eq 2, Used for Unloaded Aqueous MDEA + AMP Solutions

$w_{\text{MDEA}}/w_{\text{AMP}}$	$x_{\text{MDEA}}/x_{\text{AMP}}$	parameter	
		A	B
0.15/0.15	0.030/0.040	-7.04	2442.86
0.20/0.20	0.045/0.060	-8.26	2966.36
0.25/0.25	0.064/0.086	-9.31	3450.81
0.30/0.30	0.089/0.012	-10.56	4005.05

of unloaded aqueous MDEA + AMP solutions are shown as dashed lines in Figure 2. As can be seen, this model showed good agreement with experimental viscosities. The corresponding Ω between experimental and calculated viscosities is calculated to be 0.07 mPa·s, which is less than the experimental uncertainty for unloaded MDEA + AMP aqueous solutions. The parity plots between experimental and calculated data from eq 2 are illustrated in Figure 3. This low deviation value conveys that this model is able to correlate the viscosity of unloaded aqueous MDEA + AMP solutions satisfactorily.

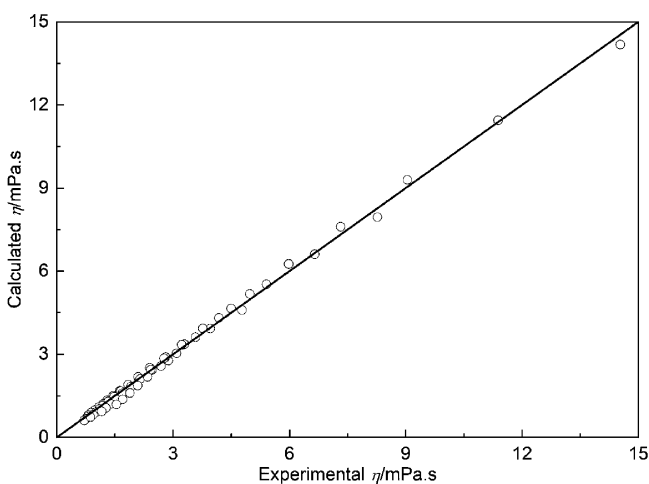


Figure 3. Comparison between experimental and calculated viscosity data for unloaded aqueous MDEA + AMP solutions.

3.3. Viscosity of CO₂-Loaded Aqueous MDEA + AMP Solutions. Viscosities of CO₂-loaded aqueous MDEA + AMP solutions at temperatures 308.15 to 373.15 K are presented in Tables 6–9 for different CO₂ loadings and amine mass fractions. Figures S1 and S2 provide an insight into the influence of CO₂ loading on viscosities of MDEA + AMP solutions. As can be seen from the figures, the viscosity of solution increases with an increase in CO₂ loading value due to the increase in intermolecular forces between aqueous amine molecules, CO₂, and reaction products. Viscosity is also dependent on the overall amine concentration as expected.

In this work, a modified Setchenow equation, as shown in eq 3, was used to correlate viscosities of CO₂-loaded solutions.³⁷ This equation has been used earlier with good results to represent the physical properties of aqueous amine solutions.^{14,44–46}

Table 6. Viscosity η of CO₂-Loaded MDEA + AMP Aqueous Solutions at $w_{\text{MDEA}}/w_{\text{AMP}}$ of 0.15/0.15 Mass Fraction at Different Temperatures T , CO₂ Loading Values α , and CO₂ Mole Fraction x_4 ^{a,b}

parameter	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.15/0.15			
	α (mol CO ₂ /mol amine)	0.40	0.58	0.80
x_4	0.027	0.039	0.053	
T (K)	η (mPa·s)			
313.15	2.59	2.82	2.85	
318.15	2.24	2.48	2.51	
323.15	1.94	2.15	2.21	
328.15	1.71	1.91	2.00	
333.15	1.51	1.69	1.79	
338.15	1.36	1.53	1.64	
343.15	1.23	1.39	1.53	
348.15	1.11	1.26	1.39	
353.15	0.99	1.14	1.26	
358.15	0.92	1.07	1.17	
363.15	0.85	1.00	1.09	
368.15	0.78	0.93	1.01	
373.15	0.72	0.87	0.92	

^aThe CO₂ loading values α are defined as the mole of CO₂ per total moles of MDEA and AMP. The pressure during experiments was maintained by N₂ gas ($p = 400$ kPa). ^bStandard uncertainties u are $u(w_{\text{MDEA+AMP}}) = 0.01$, $u(T) = 0.03$ K, and $u(P) = 0.2$ kPa. The combined standard uncertainty for viscosity measurement $u_c(\eta)$ is 0.15 mPa·s.

Table 7. Viscosity η of CO₂-Loaded MDEA + AMP Aqueous Solutions at $w_{\text{MDEA}}/w_{\text{AMP}}$ of 0.20/0.20 Mass Fraction at Different Temperatures T , CO₂ Loading Values α , and CO₂ Mole Fraction x_4 ^{a,b}

parameter	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.20/0.20				
	α (mol CO ₂ /mol amine)	0.37	0.46	0.56	0.75
x_4	0.037	0.046	0.056	0.073	
T (K)	η (mPa·s)				
308.15	5.99	6.44	6.76	7.24	
313.15	4.97	5.33	5.55	6.15	
318.15	4.20	4.50	4.67	5.23	
323.15	3.57	3.81	3.96	4.50	
328.15	3.09	3.28	3.39	3.93	
333.15	2.69	2.84	2.93	3.44	
338.15	2.39	2.51	2.58	3.07	
343.15	2.14	2.23	2.28	2.75	
348.15	1.92	1.99	2.02	2.47	
353.15	1.73	1.79	1.79	2.21	
358.15	1.58	1.62	1.63	1.94	
363.15	1.46	1.47	1.48	1.74	
368.15	1.33	1.33	1.34	1.56	
373.15	1.22	1.23	1.23	1.38	

^aThe CO₂ loading values α are defined as the mole of CO₂ per total moles of MDEA and AMP. The pressure during experiments was maintained by N₂ gas ($p = 400$ kPa). ^bStandard uncertainties u are $u(w_{\text{MDEA+AMP}}) = 0.01$, $u(T) = 0.03$ K, and $u(P) = 0.2$ kPa. The combined standard uncertainty for viscosity measurement $u_c(\eta)$ is 0.15 mPa·s.

$$\ln\left(\frac{\eta}{\eta_r}\right) = \sum_{j=1}^n k_j \times \alpha^j \quad (3)$$

Table 8. Viscosity η of CO₂-Loaded MDEA + AMP Aqueous Solutions at $w_{\text{MDEA}}/w_{\text{AMP}}$ of 0.25/0.25 Mass Fraction at Different Temperatures T , CO₂ Loading Values α , and CO₂ Mole Fraction x_4 ^{a,b}

parameter	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.25/0.25			
α (mol CO ₂ /mol amine)	0.23	0.29	0.34	0.47
x_4	0.033	0.042	0.049	0.066
T (K)	η (mPa·s)			
308.15	10.03	11.42	12.19	14.22
313.15	8.04	9.17	9.74	11.29
318.15	6.60	7.55	7.96	9.20
323.15	5.48	6.24	6.57	7.57
328.15	4.63	5.29	5.52	6.37
333.15	3.94	4.49	4.68	5.40
338.15	3.42	3.91	4.05	4.66
343.15	2.99	3.41	3.53	4.06
348.15	2.63	3.00	3.10	3.56
353.15	2.32	2.64	2.74	3.14
358.15	2.09	2.38	2.47	2.81
363.15	1.88	2.12	2.21	2.51
368.15	1.70	1.92	2.00	2.26
373.15	1.55	1.74	1.81	2.04

^aThe CO₂ loading values α are defined as the mole of CO₂ per total moles of MDEA and AMP. The pressure during experiments was maintained by N₂ gas ($p = 400$ kPa). ^bStandard uncertainties u are $u(w_{\text{MDEA}+\text{AMP}}) = 0.01$, $u(T) = 0.03$ K, and $u(P) = 0.2$ kPa. The combined standard uncertainty for viscosity measurement $u_c(\eta)$ is 0.15 mPa·s.

Table 9. Viscosity η of CO₂-Loaded MDEA + AMP Aqueous Solutions at $w_{\text{MDEA}}/w_{\text{AMP}}$ of 0.30/0.30 Mass Fraction at Different Temperatures T , CO₂ Loading Values α , and CO₂ Mole Fraction x_4 ^{a,b}

parameter	$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.30/0.30	
α (mol CO ₂ /mol amine)	0.11	0.22
x_4	0.022	0.044
T (K)	η (mPa·s)	
313.15	12.22	18.71
318.15	9.84	12.91
323.15	8.05	10.18
328.15	6.68	8.38
333.15	5.56	6.96
338.15	4.79	5.93
343.15	4.10	5.04
348.15	3.61	4.35
353.15	3.11	3.78
358.15	2.77	3.34
363.15	2.47	2.95
368.15	2.21	2.63
373.15	1.96	2.36

^aThe CO₂ loading values α are defined as the mole of CO₂ per total moles of MDEA and AMP. The pressure during experiments was maintained by N₂ gas ($p = 400$ kPa). ^bStandard uncertainties u are $u(w_{\text{MDEA}+\text{AMP}}) = 0.01$, $u(T) = 0.03$ K, and $u(P) = 0.2$ kPa. The combined standard uncertainty for viscosity measurement $u_c(\eta)$ is 0.15 mPa·s.

In eq 3, η/η_r is the ratio between viscosities of CO₂-loaded and unloaded aqueous MDEA + AMP solutions at same temperatures and MDEA + AMP concentrations, k_j is the Setchenow coefficient, which is temperature-dependent, and α

represents moles of CO₂ dissolved per mole of amine mixture (MDEA + AMP). A second-order modified Setchenow equation was used to correlate viscosity of CO₂-loaded aqueous MDEA + AMP solutions, and the explicit form of eq 3 is presented as in eq 4.

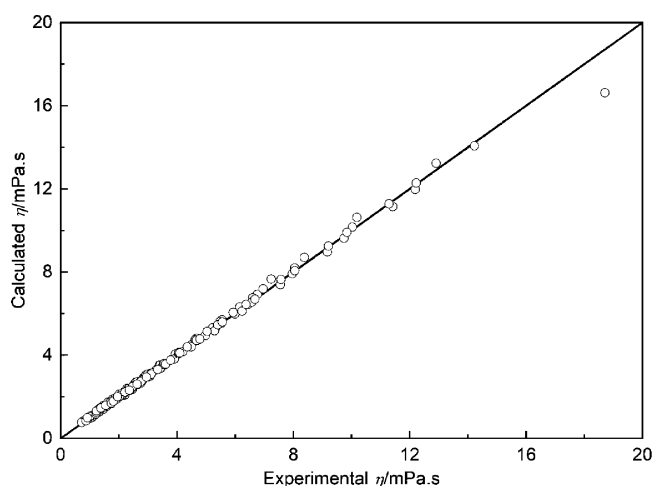
$$\ln\left(\frac{\eta}{\eta_r}\right) = (k_{1,0} + k_{1,1} \times T) \times \alpha + (k_{2,0} + k_{2,1} \times T) \times \alpha^2 \quad (4)$$

The numerical values of the Setchenow temperature-dependent coefficients at different MDEA + AMP concentrations are shown in Table 10 together with the Ω values. A

Table 10. Setchenow Temperature-Dependent Coefficients k and Ω Values Based on Modified Setchenow eq 4 for CO₂-Loaded MDEA + AMP Solutions for Different Mass Fractions of MDEA + AMP ($w_{\text{MDEA}}/w_{\text{AMP}}$)

$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.15/0.15; $x_{\text{MDEA}}/x_{\text{AMP}}$, 0.030/0.040			$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.20/0.20; $x_{\text{MDEA}}/x_{\text{AMP}}$, 0.045/0.060		
$k_{1,0} = 4.453$	$k_{1,1} = -0.012$	$\Omega = 0.035$ mPa·s	$k_{1,0} = 3.406$	$k_{1,1} = -0.0067$	$\Omega = 0.067$ mPa·s
$k_{2,0} = -5.123$	$k_{2,1} = 0.0152$		$k_{2,0} = -2.157$	$k_{2,1} = -0.005$	
$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.25/0.25; $x_{\text{MDEA}}/x_{\text{AMP}}$, 0.064/0.086			$w_{\text{MDEA}}/w_{\text{AMP}}$, 0.30/0.30; $x_{\text{MDEA}}/x_{\text{AMP}}$, 0.089/0.12		
$k_{1,0} = 5.009$	$k_{1,1} = -0.0095$	$\Omega = 0.070$ mPa·s	$k_{1,0} = 2.299$	$k_{1,1} = 0.0016$	$\Omega = 0.161$ mPa·s
$k_{2,0} = -3.288$	$k_{2,1} = 0.0073$		$k_{2,0} = 23.745$	$k_{2,1} = -0.0763$	

parity plot comparing experimental and calculated viscosity data for CO₂-loaded MDEA + AMP solutions studied in this work is shown in Figure 4. As can be seen from Figure 4, the

**Figure 4.** Comparison between experimental and calculated viscosity data for CO₂-loaded aqueous MDEA + AMP solutions.

modified Setchenow equation shows good agreement with our experimental viscosities with corresponding Ω being 0.08 mPa·s for all the systems, which is less than the combined standard experimental uncertainty value of 0.15 mPa·s for CO₂-loaded MDEA + AMP aqueous solutions.

4. CONCLUSIONS

This work presents new experimental data for viscosities of both unloaded and CO₂-loaded aqueous MDEA + AMP

solutions at different temperatures (298.15 to 373.15 K) and concentrations. At all temperatures and concentrations studied, the viscosity of unloaded and CO₂-loaded aqueous MDEA + AMP solutions decreased with an increase in temperature. The calculated viscosities from the regressed model fit the experimental viscosity data of unloaded aqueous MDEA + AMP solutions satisfactorily with an average absolute deviation value less than the combined standard uncertainty for unloaded aqueous MDEA + AMP solutions. The viscosities of the CO₂-loaded aqueous MDEA + AMP solutions were measured at different CO₂ loadings for each blended amine concentration. These viscosity values increased with an increase in CO₂ loading and amine concentration and were found to be higher than those of the unloaded solutions. The experimental viscosity data of CO₂-loaded aqueous MDEA + AMP solutions were correlated using a modified Setchenow equation. The model was able to represent the viscosities of CO₂-loaded MDEA + AMP solutions with an average absolute deviation value less than the combined standard experimental uncertainty value for CO₂-loaded aqueous MDEA + AMP solutions. The reported viscosity data of both unloaded and CO₂-loaded aqueous MDEA + AMP solutions are complementary to the other available amine viscosity data.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.jced.0c00088>.

Density of selected MDEA + AMP samples before and after viscosity measurement, viscosity of unloaded AMP solutions, and plots of viscosity of CO₂-loaded MDEA + AMP solutions (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Zulkifli Idris – Faculty of Technology, Natural Science and Maritime Sciences, University of South-Eastern Norway, Porsgrunn 3918, Norway; orcid.org/0000-0001-7905-9686; Phone: +47 3100 8000; Email: Zulkifli.Idris@usn.no

Authors

Nithin B. Kummamuru – Faculty of Technology, Natural Science and Maritime Sciences, University of South-Eastern Norway, Porsgrunn 3918, Norway; Department of Chemical Engineering, Birla Institute of Technology and Science, Pilani, Hyderabad Campus, Hyderabad, Telangana 500078, India
Dag A. Eimer – Faculty of Technology, Natural Science and Maritime Sciences, University of South-Eastern Norway, Porsgrunn 3918, Norway

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acs.jced.0c00088>

Notes

The authors declare no competing financial interest.

■ REFERENCES

(1) Kohl, A.; Nielsen, R. *Gas Purification*; 5th ed.; Gulf Publishing Company: Houston, Texas, 1997.
(2) Cuccia, L.; Dugay, J.; Bontemps, D.; Louis-Louis, M.; Morand, T.; Kanniche, M.; Bellosta, V.; Vial, J. Monitoring of the blend monoethanolamine/ methyl-diethanolamine/water for post-combustion CO₂ capture. *Int. J. Greenhouse Gas Control* **2019**, *80*, 43–53.

(3) Adewale, A.; Nabil, H.; Earl, G.; Mohammad, Z. Absorption of CO₂ by amine blends solution: an experimental evaluation. *Int. J. Eng. Sci.* **2013**, *3*, 12–23.

(4) Abdelbaki, B.; Mohammed, A.-M. Reactive Absorption of CO₂ into Aqueous Mixtures of Methyl-diethanolamine and Diethanolamine. *Int. J. Chem. Eng. Appl.* **2014**, *5*, 291–297.

(5) Kim, S.; Shi, H.; Lee, J. Y. CO₂ absorption mechanism in amine solvents and enhancement of CO₂ capture capability in blended amine solvent. *Int. J. Greenhouse Gas Control* **2016**, *45*, 181–188.

(6) Bishnoi, S.; Rochelle, G. T. Absorption of carbon dioxide into aqueous piperazine: reaction kinetics, mass transfer and solubility. *Chem. Eng. Sci.* **2000**, *55*, 5531–5543.

(7) Xu, G.-W.; Zhang, C.-F.; Qin, S.-J.; Gao, W.-H.; Liu, H.-B. Gas-liquid equilibrium in a CO₂-MDEA-H₂O system and the effect of piperazine on it. *Ind. Eng. Chem. Res.* **1998**, *37*, 1473–1477.

(8) Chakravarty, T.; Phukan, U.; Weiland, R. Reaction of acid gases with mixtures of amines. *Chem. Eng. Prog.* **1985**, *81*, 32–36.

(9) Adeosun, A.; Abu-Zahra, M. R. M. Evaluation of amine-blend solvent systems for CO₂ post-combustion capture applications. *Energy Procedia* **2013**, *37*, 211–218.

(10) Idem, R.; Wilson, M.; Tontiwachwuthikul, P.; Chakma, A.; Veawab, A.; Aroonwilas, A.; Gelowitz, D. Pilot Plant Studies of the CO₂ Capture Performance of Aqueous MEA and Mixed MEA/MDEA Solvents at the University of Regina CO₂ Capture Technology Development Plant and the Boundary Dam CO₂ Capture Demonstration Plant. *Ind. Eng. Chem. Res.* **2006**, *45*, 2414–2420.

(11) Aaron, D.; Tsouris, C. Separation of CO₂ from flue gas: a review. *Sep. Sci. Technol.* **2005**, *40*, 321–348.

(12) Paul, S.; Mandal, B. Density and Viscosity of Aqueous Solutions of (N-Methyl-diethanolamine + Piperazine) and (2-Amino-2-methyl-1-propanol + Piperazine) from (288 to 333) K. *J. Chem. Eng. Data* **2006**, *51*, 1808–1810.

(13) Muhammad, A.; Mutalib, M. I. A.; Wilfred, C. D.; Murugesan, T.; Shafeeq, A. Viscosity, Refractive Index, Surface Tension, and Thermal Decomposition of Aqueous N-Methyl-diethanolamine Solutions from (298.15 to 338.15) K. *J. Chem. Eng. Data* **2008**, *53*, 2226–2229.

(14) Shokouhi, M.; Jalili, A. H.; Samani, F.; Hosseini-Jenab, M. Experimental investigation of the density and viscosity of CO₂-loaded aqueous alkanolamine solutions. *Fluid Phase Equilib.* **2015**, *404*, 96–108.

(15) Sartori, G.; Savage, D. W. Sterically Hindered Amines for Carbon Dioxide Removal from Gases. *Ind. Eng. Chem. Fundam.* **1983**, *22*, 239–249.

(16) Chakraborty, A. K.; Astarita, G.; Bischoff, K. B. CO₂ absorption in aqueous solutions of hindered amines. *Chem. Eng. Sci.* **1986**, *41*, 997–1003.

(17) al-Masabi, F. H.; Castier, M. Simulation of carbon dioxide recovery from flue gases in aqueous 2-amino-2-methyl-1-propanol solutions. *Int. J. Greenhouse Gas Control* **2011**, *5*, 1478–1488.

(18) Xu, S.; Wang, Y.-W.; Otto, F. D.; Mather, A. E. Kinetics of the reaction of carbon dioxide with 2-amino-2-methyl-1-propanol solutions. *Chem. Eng. Sci.* **1996**, *51*, 841–850.

(19) Zhang, P.; Shi, Y.; Wei, J.; Zhao, W.; Ye, Q. Regeneration of 2-amino-2-methyl-1-propanol used for carbon dioxide absorption. *J. Environ. Sci.* **2008**, *20*, 39–44.

(20) Khan, A. A.; Halder, G. N.; Saha, A. K. Comparing CO₂ removal characteristics of aqueous solutions of monoethanolamine, 2-amino-2-methyl-1-propanol, methyl-diethanolamine and piperazine through absorption process. *Int. J. Greenhouse Gas Control* **2016**, *50*, 179–189.

(21) Li, M.-H.; Lie, Y.-C. Densities and Viscosities of Solutions of Monoethanolamine + N-methyl-diethanolamine + Water and Monoethanolamine + 2-Amino-2-methyl-1-propanol + Water. *J. Chem. Eng. Data* **1994**, *39*, 444–447.

(22) Mandal, B. P.; Kundu, M.; Bandyopadhyay, S. S. Density and Viscosity of Aqueous Solutions of (N-Methyl-diethanolamine + Monoethanolamine), (N-Methyl-diethanolamine + Diethanolamine), (2-Amino-2-methyl-1-propanol + Monoethanolamine), and (2-

Amino-2-methyl-1-propanol + Diethanolamine). *J. Chem. Eng. Data* **2003**, *48*, 703–707.

(23) Xu, S.; Otto, F. D.; Mather, A. E. Physical properties of aqueous AMP solutions. *J. Chem. Eng. Data* **1991**, *36*, 71–75.

(24) Samanta, A.; Bandyopadhyay, S. S. Density and Viscosity of Aqueous Solutions of Piperazine and (2-Amino-2-methyl-1-propanol + Piperazine) from 298 to 333 K. *J. Chem. Eng. Data* **2006**, *51*, 467–470.

(25) Samanta, A.; Bandyopadhyay, S. S. Absorption of carbon dioxide into aqueous solutions of piperazine activated 2-amino-2-methyl-1-propanol. *Chem. Eng. Sci.* **2009**, *64*, 1185–1194.

(26) Xiao, J.; Li, C.-W.; Li, M.-H. Kinetics of absorption of carbon dioxide into aqueous solutions of 2-amino-2-methyl-1-propanol + monoethanolamine. *Chem. Eng. Sci.* **2000**, *55*, 161–175.

(27) Mandal, B. P.; Biswas, A. K.; Bandyopadhyay, S. S. Absorption of carbon dioxide into aqueous blends of 2-amino-2-methyl-1-propanol and diethanolamine. *Chem. Eng. Sci.* **2003**, *58*, 4137–4144.

(28) Dash, S. K.; Samanta, A.; Samanta, A. N.; Bandyopadhyay, S. S. Absorption of carbon dioxide in piperazine activated concentrated aqueous 2-amino-2-methyl-1-propanol solvent. *Chem. Eng. Sci.* **2011**, *66*, 3223–3233.

(29) Henni, A.; Hromek, J. J.; Tontiwachwuthikul, P.; Chakma, A. Volumetric Properties and Viscosities for Aqueous AMP Solutions from 25 °C to 70 °C. *J. Chem. Eng. Data* **2003**, *48*, 551–556.

(30) Álvarez, E.; Gómez-Díaz, D.; La Rubia, M. D.; Navaza, J. M. Densities and Viscosities of Aqueous Ternary Mixtures of 2-(Methylamino)ethanol and 2-(Ethylamino)ethanol with Diethanolamine, Triethanolamine, N-Methyldiethanolamine, or 2-Amino-1-methyl-1-propanol from 298.15 to 323.15 K. *J. Chem. Eng. Data* **2006**, *51*, 955–962.

(31) Welsh, L. M.; Davis, R. A. Density and Viscosity of Aqueous Blends of N-Methyldiethanolamine and 2-Amino-2-methyl-1-propanol. *J. Chem. Eng. Data* **1995**, *40*, 257–259.

(32) Huang, Y.-M.; Soriano, A. N.; Caparanga, A. R.; Li, M.-H. Kinetics of absorption of carbon dioxide in 2-amino-2-methyl-1-propanol + N-methyldiethanolamine + water. *J. Taiwan Inst. Chem. Eng.* **2011**, *42*, 76–85.

(33) Haratipour, P.; Baghban, A.; Mohammadi, A. H.; Nazhad, S. H. H.; Bahadori, A. On the estimation of viscosities and densities of CO₂-loaded MDEA, MDEA+AMP, MDEA+DIPA, MDEA+MEA, and MDEA+DEA aqueous solutions. *J. Mol. Liq.* **2017**, *242*, 146–159.

(34) Ghulam, M.; Mohd, S. A.; Azmi, B. M.; Faizan, A. Volumetric Properties, Viscosities and Refractive Indices of Aqueous Solutions of 2-Amino-2-methyl-1-propanol (AMP). *Res. J. Chem. Environ.* **2013**, *17*, 22–31.

(35) Murshid, G.; Shariff, A. M.; Lau, K. K.; Bustam, M. A.; Ahmad, F. Physical Properties of Piperazine (PZ) Activated Aqueous Solutions of 2-Amino-2-hydroxymethyl-1,3-propanediol (AHPD + PZ). *J. Chem. Eng. Data* **2012**, *57*, 133–136.

(36) Setschenow, J. Über die konstitution der salzlösungen auf grund ihres verhaltens zu kohlenäure. *Z. Phys. Chem.* **1889**, *4U*, 117–125.

(37) Prausnitz, J.; Lichtenthaler, R.; Gomes de Azevedo, E. *Molecular Thermodynamics of Fluid-Phase Equilibria*; 3rd ed.; Prentice-Hall, Inc.: New Jersey, 1999.

(38) Jayarathna, S. A.; Jayarathna, C. K.; Kottage, D. A.; Dayarathna, S.; Eimer, D. A.; Melaaen, M. C. Density and Surface Tension Measurements of Partially Carbonated Aqueous Monoethanolamine Solutions. *J. Chem. Eng. Data* **2013**, *58*, 343–348.

(39) Amundsen, T. G.; Øi, L. E.; Eimer, D. A. Density and Viscosity of Monoethanolamine + Water + Carbon Dioxide from (25 to 80) °C. *J. Chem. Eng. Data* **2009**, *54*, 3096–3100.

(40) Oyekan, B. A.; Rochelle, G. T. Energy Performance of Stripper Configurations for CO₂ Capture by Aqueous Amines. *Ind. Eng. Chem. Res.* **2006**, *45*, 2457–2464.

(41) Idris, Z.; Kummamuru, N. B.; Eimer, D. A. Viscosity measurement of unloaded and CO₂-loaded aqueous monoethanolamine at higher concentrations. *J. Mol. Liq.* **2017**, *243*, 638–645.

(42) Kummamuru, N. B.; Idris, Z.; Eimer, D. A. Viscosity measurement and correlation of unloaded and CO₂-loaded aqueous

solutions of N-methyldiethanolamine-piperazine. *J. Chem. Eng. Data* **2019**, *64*, 4692–4700.

(43) Rebolledo-Libreros, M. E.; Trejo, A. Density and Viscosity of Aqueous Blends of Three Alkanolamines: N-Methyldiethanolamine, Diethanolamine, and 2-Amino-2-methyl-1-propanol in the Range of (303 to 343) K. *J. Chem. Eng. Data* **2006**, *51*, 702–707.

(44) Shokouhi, M.; Jalili, A. H.; Mohammadian, A. H.; Hosseini-Jenab, M.; Nouri, S. S. Heat capacity, thermal conductivity and thermal diffusivity of aqueous sulfolane solutions. *Thermochim. Acta* **2013**, *560*, 63–70.

(45) Kelayeh, S. A.; Jalili, A. H.; Ghotbi, C.; Hosseini-Jenab, M.; Taghikhani, V. Densities, Viscosities, and Surface Tensions of Aqueous Mixtures of Sulfolane + Triethanolamine and Sulfolane + Diisopropanolamine. *J. Chem. Eng. Data* **2011**, *56*, 4317–4324.

(46) Idris, Z.; Kummamuru, N. B.; Eimer, D. A. Viscosity Measurement and Correlation of Unloaded and CO₂-Loaded 3-Amino-1-propanol Solution. *J. Chem. Eng. Data* **2018**, *63*, 1454–1459.