Review Article

Physical Properties of MEA + Water + CO$_2$ Mixtures in Postcombustion CO$_2$ Capture: A Review of Correlations and Experimental Studies

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The knowledge of physicochemical properties of a mixture of amine, water, and CO$_2$ is beneficial in evaluating the postcombustion CO$_2$ capture process and process equipment design. This study reviews the literature of density, viscosity, and surface tension measurements with the evaluated measurement uncertainties and proposed correlations for monoethanol amine (MEA), water, and CO$_2$ mixtures. Adequate research has been performed to measure and develop correlations for pure MEA and aqueous MEA mixtures, but further studies are required for CO$_2$-loaded aqueous MEA mixtures. The correlations fit measured properties with an acceptable accuracy, and they are recommended to use in process equipment design, mathematical modelling, and simulations of absorption and desorption.

1. Introduction

Knowledge of physical properties of solvents is important for chemical engineering work like process modelling and simulations, pilot plant operation, and the design of commercial plants [1]. An amine-based CO$_2$ capture process contains equipment like absorption columns, desorption columns, pumps, and heat exchangers. The design of such equipment is depended on physical properties like density, viscosity, and surface tension. As the benchmark solvent, physicochemical properties of MEA (monoethanol amine) in a wide range of concentrations, temperatures, and pressures are essential to examine and compare other potential solvents in postcombustion CO$_2$ capture. The measured physical properties of density, viscosity, surface tension, and thermal expansion coefficient of pure and aqueous MEA mixtures are available in the literature. There is a lack of data on the measured physical properties of CO$_2$-loaded aqueous MEA [2]. Recent studies have extended the range of data available for the CO$_2$-loaded MEA, and correlations have been proposed to fit the data [1, 3, 4].

The main objective of this review is to gather the literature of measured physical properties and semiempirical and empirical correlations of density, viscosity surface tension, and thermal expansion coefficient. The areas with a lack of measured data were identified and challenges were discussed in the experimental studies. The performance of proposed correlations was compared considering the accuracies of the data fit and applicability in the aspects such as mathematical modelling and simulations. The reported measurement uncertainties of pure MEA, aqueous MEA, and CO$_2$-loaded aqueous MEA solutions were tabulated and compared.

2. Density, Viscosity, and Surface Tension Measurements

Density, viscosity, and surface tension are used in the mass transfer and interfacial area correlations that were developed for both random and structured packings. The measured data of MEA + H$_2$O mixtures and MEA + H$_2$O + CO$_2$ mixtures have been published in various sources in which the

measurements were performed under different MEA concentrations, temperatures, and CO_2 loadings. Various advanced instruments have been used to acquire a high accuracy of measurements. In the analysis of CO_2 capture processes, it is essential to measure physical properties that cover all the conditions of the process.

2.1. Density of MEA and H₂O Mixtures.

Table 1 summarizes the performed studies on density measurements of pure MEA under different temperatures in which most of the previous density measurements were limited to the temperature range of 293.15 to 353.15 K [2, 4–24]. DiGuilio et al. [9] studied densities of various ethanolamines and MEA in the temperature range of 294.4 to 431.3 K. There is a lack of information about densities of MEA at high pressures. Sobrino et al. [25] were able to measure densities of aqueous MEA mixtures at both high temperatures and pressures. The study was performed within the temperature range of 293.15 to 393.15 K and the pressure range from 0.1 MPa to 120 MPa. The density of aqueous MEA has been measured extensively under a wide range of MEA concentrations as shown in Table 2. The data are highly valuable because of their usability in the calculation of other important physiochemical parameters in the process. In the process, the absorption column operates with CO_2-loaded aqueous MEA solution. Typical operating conditions for the absorption process with CO_2 loading are from generally 0.2 to 0.5 mol CO_2/mol MEA [30]. The studies performed on density measurement of CO_2-loaded aqueous MEA are listed in Table 3. Several challenges were noticed in density measurement of aqueous MEA and CO_2-loaded aqueous MEA solutions. There is a high probability to evaporate MEA from the mixtures at high temperatures. Further, a desorption of CO_2 is also present in CO_2-loaded aqueous mixtures at high temperatures and high CO_2 loadings. Accordingly, care must be given to minimize occurrence of such phenomena through visual observations to get accurate density measurements. CO_2 or MEA evaporation is observed as bubble formation inside the U-tube in oscillating density meters, which leads to an error in density measurement.

2.2. Viscosity of MEA and Water Mixtures.

Viscosity measurements of MEA and water mixtures are equally important as density measurements in the postcombustion absorption process. Viscosity has a high impact on the mass transfer coefficient of gas into a liquid in a packed bed absorber [32]. The viscosity of MEA varies with the amount of water and CO_2 present in the solution and decreases as the solution temperature increases. The available literature for the viscosity measurements of pure MEA is shown in Table 4 [2, 9, 13–17, 19, 23, 24, 28, 33]. Previous studies have attempted to cover the viscosity data in the range of 0–100 mass% MEA [14, 17, 33]. Measurements at a temperature above 373.15 K are reported in [33, 36]. For CO_2-loaded aqueous MEA, most of the reported studies presented the viscosity of 30 mass% MEA solutions within the CO_2-loading range of 0–0.5 mol CO_2/mol MEA. Idris et al. [34] discussed the viscosity measurements at higher (>50 mass%)

MEA concentrations. The study performed by Arachchige et al. [37] presented data at higher temperatures (>373.15 K). Tables 5 and 6 list studies performed on viscosity measurements of aqueous MEA and CO_2-loaded aqueous MEA, respectively. The evaporation and desorption of MEA and CO_2 from aqueous MEA and CO_2-loaded aqueous MEA solutions cause errors in the viscosity measurements. Idris et al. [34] adopted a method to suppress the CO_2-loaded aqueous MEA mixture using N_2 gas with 4 bar pressure to avoid the escape of CO_2 from the system. Further, Idris et al. [34] claim that the applied pressure would not influence the outcome of the experiments.

2.3. Surface Tension of MEA and Water Mixtures.

Surface tension has a high influence on the effective interfacial area of the packing material [32] and ultimately influences the overall mass transfer rate. Accurate and reliable surface tension data can enhance the confidence in process simulations, which will reduce the cost and safety margins [40]. The surface tension measurement of MEA solutions also can be performed for pure MEA, aqueous MEA, and CO_2-loaded aqueous MEA. Literature is available for the measured surface tension of MEA for all kinds of solutions. Vazquez et al. [41] measured both pure MEA and aqueous MEA at different temperatures from 298.15 to 333.15 K using a Traube stalagmometer and a Prolabo tensiometer based on the Wilhemy plate method. Idris et al. [40] and Han et al. [4] measured the surface tension of pure MEA and aqueous MEA at different temperatures using Rame-Hart advanced goniometer model 500. For CO_2-loaded aqueous MEA, Jayarathna et al. [31] measured aqueous solutions of 20–70 mass% MEA with CO_2 loading 0–0.5 mol CO_2/mol MEA at temperatures from 303.15 to 333.15 K and also 80 mass% MEA with CO_2 loading 0–0.5 mol CO_2/mole MEA at temperatures from 313.15 to 343.15 K.

2.4. Uncertainty of Density, Viscosity, and Surface Tension Measurements.

Analysis of the measurement uncertainty provides a quantitative indication of the quality of the measurement result [42]. Subsequently, it gives information about the confidence in any decision based on its use. Effective identification of uncertainty sources is vital and (combined) standard uncertainty is calculated by combining the respective uncertainty components of all important uncertainty sources. Guide to the Expression of Uncertainty in Measurement (GUM) published by ISO facilitates a guidance to evaluate the uncertainty in the output of a measurement system [43].

The functional relationship between measured quantity $x = \{x_i\}$ (the input) and the measurement result y (the output) is shown as

$$y = f(x), \qquad (1)$$

$$u^2(y) = \sum_{i=1}^{n} \left(\frac{\partial f}{\partial x_i}\right)^2 u^2(x_i) + 2 \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} u(x_i, x_j). \qquad (2)$$

TABLE 1: Previous measurements of density of pure MEA.

Source	T (K)		No of points	Method
	Low	High		
Touhara et al. [5]	298.15		1	Pycnometer
Yang et al. [6]	293.15	343.15	6	Anton Paar (DMA 5000M)
Li and Shen [7]	303.15	353.15	8	Pycnometer
Wang et al. [8]	293.45	360.65	5	Pycnometer
DiGuilio et al. [9]	294.4	431.3	8	Pycnometer
Page et al. [10]	283.15	313.15	3	Flow densimeter
Maham et al. [11]	298.15	353.15	5	Anton Paar (DMA 45)
Guevara and Rodriguez [12]	298.15	333.15	8	Sodev 03D vibrating densimeter
Li and Lie [13]	303.15	353.15	6	Pycnometer
Lee and Lin [14]	303.15	323.15	3	Pycnometer
Song et al. [15]	303.15	343.15	5	Pycnometer
Kapadi et al. [16]	303.15	318.15	4	Anton Paar (DMA 5000)
Islam et al. [17]	293.15		1	Pycnometer
Valtz et al. [18]	281.15	353.15	37	Anton Paar (DMA 5000)
Geng et al. [19]	288.15	323.15	8	Pycnometer
Pouryousefi and Idem [20]	295.15	333.15	4	Anton Paar (DMA 4500/DMA 5000)
Amundsen et al. [2]	298.15	353.15	5	Anton Paar (DMA 4500)
Taib and Murugesan [21]	303.15	353.15	6	Anton Paar (DMA 5000)
Taib and Murugesan [22]	293.15	353.15	16	Anton Paar (DMA 5000M)
Han et al. [4]	298.15	423.15	20	Anton Paar (DMA 4500/DMA HP)
Abuin et al. [23]	298.15		1	Anton Paar (DSA 5000)
Yang et al. [6]	293.15	343.15	6	Anton Paar (DMA 5000M)
Xu et al. [24]	293.15	333.15	5	Anton Paar (DMA 5000)
Ma et al. [27]	293.15	333.15	5	Anton Paar (DMA 4500M)

TABLE 2: Sources of reported density measurements of aqueous MEA.

Source	Concentration: MEA (x_1)		T (K)		No of points	Method
	Low	High	Low	High		
Weiland et al. [26]	0.0317	0.1643	298.15		4	Hydrometer
Amundsen et al. [2]	0.0687	0.7264	298.15	353.15	30	Anton Paar (DMA 4500)
Han et al. [4]	0.1122	0.7264	298.15	423.15	140	Anton Paar (DMA4500/DMA HP)
Hartono et al. [1]	0.0191	0.1122	293.15	353.15	15	Anton Paar (DMA 4500M)
Page et al. [10]	0.00118	0.99695	283.15	313.15	62	Flow densimeter
Maham et al. [11]	0.0054	0.9660	298.15	353.15	100	Anton Paar (DMA 45)
Lee and Lin [14]	0.1000	0.9000	303.15	323.15	27	Pycnometer
Kapadi et al. [16]	0.1122	0.8486	303.15	383.15	32	Anton Paar (DMA 5000)
Pouryousefi and Idem [20]	0.0155	0.9192	295.15	333.15	80	Anton Paar (DMA 4500/DMA 5000)
Ma et al. [27]	0.1000	0.8995	293.15	333.15	45	Anton Paar (DMA 4500M)
Mandal et al. [28]	0.1122		293.15	323.15	7	Pycnometer
Li and Lie [13]	0.0687		303.15	353.15	6	Pycnometer
Zhang et al. [29]	0.1122		298.15	353.15	9	Anton Paar (DMA 5000M)

TABLE 3: Sources of reported density measurements of CO_2-loaded aqueous MEA.

Source	Concentration: mass% MEA in (MEA + water) solutions		CO_2 loading: α (mol CO_2/ mol MEA)		T (K)	No. of points	Method
	Low	High	Low	High			
Weiland et al. [26]	10	40	0.05	0.5	298.15	40	Hydrometer
Amundsen et al. [2]	20	40	0.1	0.5	298.15–353.15	68	Anton Paar (DMA 4500)
Han et al. [4]	30	60	0.1	0.56	298.15–423.15	240	Anton Paar (DMA 4500/DMA HP)
Jayarathna et al. [31]	20	70	0.1	0.5	303.15–333.15	144	Anton Paar (DMA 4500M)
Jayarathna et al. [3]	80		0.07	0.51	313.15–343.15	64	Anton Paar (DMA 4500M)
Hartono et al. [1]	6.2	30	0.1	0.5	293.15–353.15	68	Anton Paar (DMA 4500M)
Zhang et al. [29]	30		0.14	0.49	298.15–353.15	33	Anton Paar (DMA 5000M)

TABLE 4: Sources of reported viscosity measurements of pure MEA.

Source	T (K) Low	High	No of points	Method
DiGuilio et al. [9]	303.6	423.7	8	Cannon-Ubbelohde capillary viscometer
Li and Lie [13]	303.15	353.15	6	Cannon-Fenske routine viscometer
Lee and Lin [14]	303.15	323.15	3	Haake falling-ball viscometer
Song et al. [15]	303.15	343.15	5	Ubbelohde viscometer
Kapadi et al. [16]	303.15	318.15	4	Ubbelohde viscometer
Islam et al. [17]	293.15	323.15	6	U-tube Ostwald viscometer
Geng et al. [19]	288.15	323.15	8	Ubbelohde viscometer
Amundsen et al. [2]	298.15	353.15	5	ZIDIN viscometer
Abuin et al. [23]	298.15		1	Ubbelohde viscometer
Arachchige et al. [33]	293.15	423.15	15	Anton Paar MCR 101 with a double gap measuring cell
Xu et al. [24]	293.15	333.15	5	Anton PaarAMVn
Ma et al. [27]	293.15	333.15	5	LUNDA iVisc capillary viscometer
Idris et al. [34]	298.15	373.15	16	Anton Paar MCR 101 with a double gap measuring cell
Maham et al. [35]	298.15	353.15	5	Ubbelohde viscometer/capillary viscometer

TABLE 5: Sources of reported viscosity measurements of aqueous MEA.

Source	Concentration: (x_1) MEA Low	High	T (K) Low	High	No of points	Method
Weiland et al. [26]	0.0317	0.1643	298.15		4	Cannon-Fenske viscometer
Amundsen et al. [2]	0.0687	0.7264	298.15	353.15	30	ZIDIN viscometer
Arachchige et al. [33]	0.0317	0.7264	293.15	353.15	72	Anton Paar MCR 101 with a double gap measuring cell
Hartono et al. [1]	0.0191	0.1122	293.15	353.15	26	Anton Paar MCR 100 with a double gap measuring cell
Arachchige et al. [38]	0.0317	0.7264	363.15	423.15	63	Anton Paar MCR 101 with a double gap measuring cell
Idris et al. [34]	0.2278	0.7264	298.15	373.15	128	Anton Paar MCR 101 with a double gap measuring cell
Lee and Lin [14]	0.1000	0.9000	303.15	323.15	27	Haake falling-ball viscometer
Kapadi et al. [16]	0.1122	0.8486	303.15	318.15	32	Ubbelohde viscometer
Islam et al. [17]	0.0322	0.7296	303.15	323.15	45	U-tube Ostwald viscometer
Ma et al. [27]	0.1000	0.8995	293.15	333.15	45	LUNDA iVisc capillary viscometer
Maham et al. [35]	0.0313	0.8446	298.15	353.15	60	Ubbelohde viscometer/capillary viscometer
Li and Lie [13]	0.0687	0.1122	303.15	353.15	6	Cannon-Fenske routine viscometer
Zhang et al. [29]	0.1122		298.15	353.15	7	U-tube capillary viscometer
Mandal et al. [28]	0.1122		298.15	323.15	7	Ostwald viscometer

TABLE 6: Sources of reported viscosity measurements of CO_2-loaded aqueous MEA.

Source	Concentration: mass% MEA Low	High	CO_2 loading: α (mol CO_2/mol MEA) Low	High	T (K) Low	High	No of points	Method
Weiland et al. [26]	10	40	0.05	0.5	298.15		20	Cannon-Fenske viscometer
Amundsen et al. [2]	20	40	0.1	0.5	298.15	353.15	75	ZIDIN viscometer
Fu et al. [39]	20	40	0.1	0.5	298.15		15	NDJ-1 rotational viscometer
Hartono et al. [1]	6.2	30	0.11	0.5	293.15	353.15	100	Anton Paar MCR 100 with a double gap measuring cell
Idris et al. [34]	50	80	0.08	0.52	298.15	373.15	320	Anton Paar MCR 101 with a double gap measuring cell
Arachchige et al. [37]	10	50	0.1	0.5	293.15	423.15	375	Anton Paar MCR 101 with a double gap measuring cell
Zhang et al. [29]	30		0.14	0.49	298.15	353.15	23	U-tube capillary viscometer

Equation (2) describes the propagation of uncertainty based on a first-order Taylor series expansion in which $u^2(y)$, $\partial f/\partial x_i$, $u^2(x_i)$, and $u(x_i, x_j)$ are variance of the measuring result, partial derivative, variance of the input quantity x_i and covariance between x_i and x_j [44].

Literature studies on density measurements of MEA reveal that many studies have given great attention to the uncertainty analysis to calculate the standard uncertainty for the measurements. Several density measurement data rely only on the uncertainty or accuracy of the measuring instrument, and it was not given a significant interest to calculate the combined uncertainty. Many factors contribute to the uncertainty of density measurements. Typical uncertainty sources in density and viscosity measurements are the purity of the material, weight measurements in sample preparation, and temperature variation in the measuring instrument. The calculated $u(y)$ depends on the number of uncertainty sources considered in the evaluation. If the solutions are loaded with CO_2, then the uncertainty of the CO_2 concentration in solution is important and cannot be neglected. Reported uncertainties on previous studies are shown in Table 7.

The uncertainty of CO_2 loading $u(\alpha)$ is challenging to evaluate. Many uncertainty sources are involved, and Jayarathna et al. [45] performed a detailed analysis on $u(\alpha)$ based on the titration method using $BaCl_2$ and NaOH. Amundsen et al. [2] has reported $u(\alpha)$ as 2% which is higher than what Jayarathna et al. [45] reported as 1.3%. In density measurements, although the same instrument was used Hartono et al. [1] stated a lower value for $U_c(\rho)$ compared to Jayarathna et al. [3]. For viscosity measurements, the $U_c(\eta)$ reported by Arachchige et al. [33] was higher than what was calculated by Hartono et al. [1]. Calculated $U_c(\eta)$ by Amundsen et al. [2] for CO_2-loaded aqueous MEA is greater than what Hartono et al. [1] described. In surface tension measurements, the effect of $u(\alpha)$ act similarly on $u(\sigma)$ as in density and viscosity measurement uncertainty.

3. Correlations for Density, Viscosity, Surface Tension, and Thermal Expansion Coefficient

It is useful to fit the measured physical property data in semiempirical and empirical correlations in which they can be used in mathematical modelling and simulations of absorption and desorption process. Several statistical parameters were used by different authors to evaluate the accuracy of the data fit into the correlations as given in equations (3)–(5). This section summarizes recent development in the field and correlations derived for different physical properties.

Average Absolute Deviation

$$\text{AAD} = \frac{1}{N} \sum_{i=1}^{N} \left| Y_i^E - Y_i^C \right|. \tag{3}$$

Average Absolute Relative Deviation

$$\text{AARD} = \frac{100\%}{N} \sum_{i=1}^{N} \left| \frac{Y_i^E - Y_i^C}{Y_i^E} \right|. \tag{4}$$

Maximum Deviation

$$\text{MD} = \text{MAX} \left| Y_i^E - Y_i^C \right|. \tag{5}$$

3.1. Density Correlations. This section discusses the empirical correlations developed for different types of MEA solutions. It also highlights the theoretical background of those correlations especially the excess volume of MEA and water mixtures. Table 8 summarizes the various published correlations for the density of pure, aqueous, and CO_2-loaded aqueous MEA mixtures. The density of pure liquids at different temperatures was fitted into a second-order polynomial as shown in equations (6), and coefficients were found through a regression [3, 6, 9, 12, 49]. Table 9 lists the parameters found for the polynomial correlation. Valtz et al. [18] used the correlation presented in Reid et al. [50] as given in equation (7) to predict the density of pure MEA at different temperatures. The parameters are given in Table 10.

In binary mixtures, excess molar volume V^E as given in equations (8) and (9) arises due to the different shape and size of the component molecules, physical interactions, and specific or chemical interactions among the component molecules [51–53]. Mathematically, it is defined as the difference of molar volumes between real and ideal mixtures. The theory of Prigogine–Flory–Patterson [54, 55] discusses V^E as a summation of interactional contribution, a free volume contribution, and a pressure contribution [56].

Redlich and Kister [57] illustrate an algebraic representation to adopt the excess thermodynamic properties of nonelectrolyte solutions. Therefore, the excess molar volume is presented in a power series with temperature-dependent parameters. This approach has been adopted to correlate excess molar volumes of the MEA and water binary mixture. The effect of temperature on excess volume is figured by introducing a second-order polynomial for parameters in the Redlich–Kister type correlation as shown in equations (10) and (11).

Amundsen et al. [2] and Lee and Lin [14] calculated the coefficients (A_i) for different temperatures while Hsu and Li [49] presented (A_i) for the entire temperature range of (303.15–353.15) K. A similar work was performed by Han et al. [4] in which the temperature dependence was correlated as a linear relation with respect to temperature. Hartono et al. [1] and Yang et al. [6] also developed a simplified Redlich–Kister type algebraic representation to fit the measured data as given in equations (12) and (13), respectively. The influence of pressure on the density of

TABLE 7: Literature of measurement uncertainty.

Property	Source	Instrument	Uncertainty	Comment
Density	Jayarathna et al. [31]	Anton Paar DMA HP	$U_c(\rho) = \pm 4.42 \, \text{kg·m}^{-3}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded aqueous MEA (20–70 mass%)
	Jayarathna et al. [3]	Anton Paar DMA 4500	$U_c(\rho) = \pm 6.34 \, \text{kg·m}^{-3}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded aqueous MEA (80 mass%)
	Han et al. [4]	Anton Paar DMA 4500 at $T < 373.15 \, \text{K}$ DMA HP at $T \geq 373.15 \, \text{K}$	$U_c(\rho) = \pm 0.68 \, \text{kg·m}^{-3}$ at $T < 373.15 \, \text{K}$ Level of confidence = 0.95, where $k = 2$	Aqueous MEA
			$U_c(\rho) = \pm 0.70 \, \text{kg·m}^{-3}$ $2.6 \, \text{kg·m}^{-3}$ at $T \geq 373.15 \, \text{K}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded aqueous MEA
	Abuin et al. [23]	Anton Paar DSA 5000	$U_c(\rho) = \pm 2 \times 10^{-4} \, \text{g·cm}^{-3}$ Level of confidence = 0.95, where $k = 2$	Pure MEA
	Xu et al. [24]	Anton Paar DSA 5000	$u(\rho) = \pm 0.0001 \, \text{gcm}^{-3}$ Standard uncertainty	Pure MEA
	Yang et al. [6]	Anton Paar DMA 5000 M	$u(\rho) = \pm 5 \times 10^{-6} \, \text{gcm}^{-3}$ Standard uncertainty	Pure MEA
	Amundsen et al. [2]	Anton Paar DMA 4500 M	$u(\rho) = \pm 5 \times 10^{-5} \, \text{gcm}^{-3}$ Standard uncertainty	CO_2-loaded and unloaded aqueous MEA
	Hartono et al. [1]	Anton Paar DMA 4500 M	$U_c(\rho) = \pm 0.02 \, \text{kg·m}^{-3}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded and unloaded aqueous MEA
Viscosity	Xu et al. [24]	Anton PaarAMVn	$u(\eta) = \pm 2\%$ Standard uncertainty	Pure MEA
	Amundsen et al. [2]	ZIDIN viscometer	$u(\eta) = \pm 0.01 \, \text{MPa·s}$ Standard uncertainty	CO_2-loaded and unloaded aqueous MEA
	Hartono et al. [1]	Anton Paar MCR 100	$U_c(\eta) = \pm 0.007 \, \text{MPa·s}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded and unloaded aqueous MEA
	Arachchige et al. [33]	Anton Paar MCR 101	$U_c(\eta) = \pm 0.015 \, \text{MPa·s}$ Level of confidence = 0.95, where $k = 2$	Aqueous MEA
Surface tension	Jayarathna et al. [31]	Rame-Hart advanced goniometer model 500	$U_c(\eta) = \pm 0.0004 \, \text{N·m}^{-1}$ Level of confidence = 0.95, where $k = 2$	Aqueous MEA
			$0.0012 \, \text{N·m}^{-1}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded aqueous MEA (20–70 mass%)
	Jayarathna et al. [3]	Rame-Hart advanced goniometer model 500	$0.0018 \, \text{N·m}^{-1}$ Level of confidence = 0.95, where $k = 2$	CO_2-loaded aqueous MEA (80 mass%)
	Han et al. [4]	Rame-Hart advanced goniometer model 500	$0.0004 \, \text{N·m}^{-1}$ Level of confidence = 0.95, where $k = 2$	Aqueous MEA

aqueous MEA was studied by Sobrino et al. [25]. The measured densities from 0.1 MPa up to 120 MPa under different temperatures (293.15–393.15) K and MEA compositions (10–40 mass%) were fitted to a modified Tammann–Tait equation as given in equation (14). Cheng et al. [47] developed a correlation as illustrated in equation (15) based on densities of pure liquids and mass fraction of MEA in the mixture. The correlation is capable of representing densities at different temperatures.

The construction of a proper correlation to fit the density of CO_2-loaded aqueous MEA solutions is challenging as the CO_2 dissolve and react with MEA forming various ions including carbamate, bicarbonate, and protonated MEA. The solution becomes an electrolyte and molecular interactions are more dominant than a MEA and water mixture without CO_2. Various attempts have been taken to build an effective correlation that can be easily used in process design and simulations. Licht and Weiland [48] proposed a

TABLE 8: Correlations for the density of MEA + H$_2$O + CO$_2$ mixtures.

Correlation		T (K)	P (MPa)	Data sources
$\rho = a_1 + a_2T + a_3T^2$	(6)	293.15–343.15	0.097	[6]
		294.4–431.3	0.1013	[9]
		303.15–333.15	0.8	[31]
$\rho = (A/(B^{[1+(1-(T/T_C))^C]})) \cdot (M_i/1000)$	(7)	281.15–317.15	0.1013	[18]
$V^E = V_m - x_1V_1^0 - x_2V_2^0$	(8)			
$V^E = ([x_1M_1 + x_2M_2]/\rho) - ((x_1M_1)/\rho_1) - ((x_2M_2)/\rho_2)$	(9)	298.15–423.15	0.1 from (298.15–363.15)	[4]
$V^E = x_2(1-x_2)\sum_{i=0}^{i=n}A_i(1-2x_2)^i$	(10)		0.7 from (363.15–423.15)	
$A_i = a + b(T - 273.15)$	(11)			
$V^E = x_1x_2 \cdot (A_0 + A_1t + A_2x_1 + A_3x_1^2)$	(12)	293.15–353.15	0.1013	[46]
$V^E = x_1x_2 \cdot (A_0 + A_1T + A_2x_1)$	(13)	283.15–333.15	0.097	[6]
$\rho(T,p) = (A_0 + A_1T + A_2T^2)/(1 - C\ln((B_0 + B_1T + B_2T^2 + p)/(B_0 + B_1T + B_2T^2 + 0.1\,\text{MPa})))$	(14)	293.15–393.15	0.1–120	[25]
$\rho = (1 - wf_1)\rho_2 + wf_1\rho_1 + wf_1(1 - wf_1)(a + bt + cwf_1t^d)$	(15)	283.15–373.15	0.1013	[47]
$1/\rho = u_wV_w^oe^{[\beta_w(T-T_o)]} + u_{A_1}V_{A_1}^oe^{[\beta_{A_1}(T-T_o)]} + u_{A_2}V_{A_2}^oe^{[\beta_{A_2}(T-T_o)]} + w_{CO_2}V_{CO_2}^oe^{[\beta_{CO_2}(T-T_o)]}$	(16)			[48]
$\rho = (x_1M_1 + x_2M_2 + x_3M_3)/\bar{V}$	(17)			
$V = x_1V_1 + x_2V_2 + x_3V_3 + x_1x_2V^* + x_1x_3V^{**}$	(18)	298.15	0.1013	[26]
$V_1 = M_1/(aT^2 + bT + c)$	(19)			
$V^{**} = d + ex_1$	(20)			
$\rho_{loaded} = \rho_{unloaded}/(1 - w_{CO_2added}(1 - \Phi^3))$	(21)			
$w_{CO_2added} = (\alpha x_1M_3)/(x_1M_1 + (1 - x_1 - \alpha x_1)M_2 + \alpha x_1M_3)$	(22)	293.15–353.15	0.1013	[1]
$\Phi = (a_1x_1\alpha + a_2x_1)/(a_3 + x_1)$	(23)			

Density measurements performed under atmospheric conditions are mentioned with 0.1013 MPa pressure.

TABLE 9: Parameters of density correlation for pure MEA.

Sources	Density parameters		
Density (kg/m^3)	$a_1 (kg/m^3)$	a_2 ($kg/m^3 \cdot K$)	$a_3 (kg/m^3 \cdot K^2)$
DiGuillo et al. [9]	1181.9	−0.38724	$−6.1668 \times 10^4$
Jayarathna et al. [3]	1195	−0.4566	$−5.327 \times 10^4$
Density (g/cm^3)	$a_1 (g/cm^3)$	$a_2 (g/cm^3 \cdot K)$	$a_3 (g/cm^3 \cdot K^2)$
Hsu and Li [49]	1.190	$−4.29990 \times 10^{-4}$	$−5.66040 \times 10^{-7}$
Yang et al. [6]	1.2213	$−6.1156 \times 10^{-4}$	$−2.9982 \times 10^{-7}$
Guevara and Rodriguez [12]	1.03297	$−8.0498 \times 10^{-4}$	$−3.595 \times 10^{-7}$

TABLE 10: Parameters of Valtz et al. [18] density correlation for pure MEA.

T_c (K)	A (kmol·m^{-3})	B	C
678.20	1.0002	0.2244	0.2238

correlation to predict the density of CO_2-loaded aqueous amines including MEA as described in equation (16). Weiland et al. [26] proposed a new correlation as from equations (17) to (20) for several amines, and it is extensively used in various studies related to MEA. The correlation shown from equations (21) to (23) was developed by Hartono et al. [1] for CO_2-loaded mixtures. The correlation requires the density of unloaded mixtures to represent the density data of CO_2-loaded mixtures. Literature can be found related to the verification and parameter estimation of Weiland's correlation for various MEA concentrations and temperatures. Weiland's correlation was used to fit measured density under different MEA concentrations (10–40 mass%) and CO_2 loading 0.05–0.25 mol CO_2/mol MEA at 298.15 K. Amundsen et al. [2] extended the temperature range of density measurement from 298.15 K to 353.15 K and used the same parameter values as given by Weiland et al. [26] to validate the correlation. The maximum deviation between the measurement and the correlation obtained by Amundsen et al. [2] is 1.6% at 353.15 K. Jayarathna et al. [31] extended the measured MEA concentration up to 70 mass% of aqueous MEA and CO_2 loading 0.1–0.5 mol CO_2/mol MEA in the temperature range of 303.15–333.15 K. The parameters of Weiland's correlation were estimated within that range and accuracy of the data fit was reported as 2.03 kg·m^{-3} of AAD. Han et al. [4] also used Weiland's correlation for the density prediction in an extended temperature range up to 413.15 K of the CO_2-loaded solutions. It introduced a nonlinear temperature dependence for the correlation parameters and gained a deviation between measured and correlated as 3.8 kg·m^{-3} of AAD. The main difference between Hartono's correlation and Weiland's correlation is that Hartono's correlation needs the density of unloaded density to calculate the density of loaded solutions.

A study was performed to investigate the accuracies of correlations proposed for aqueous MEA and CO_2-loaded aqueous MEA mixtures. The calculated AARD and AMD for different density correlations of aqueous MEA are listed in Table 11. Hartono's correlation for density of aqueous MEA used density data from Maham et al. [11] while Han et al.'s correlation used data from their own experiments [4]. The

highest AARD of 0.16% was observed for Han's correlation for the density data published by Amundsen et al. [2] while a maximum deviation of 4.07 kg·m^{-3} at $x_1 = 0.1$ and $T = 293.15$ K for the presented data by Ma et al. [27]. For Hartono's correlation, a maximum AARD of 0.05% and a maximum deviation of 1.79 kg·m^{-3} at $x_1 = 0.1$ and $T = 293.15$ K were found for measured viscosities given by Ma et al. [27].

Table 12 lists the calculated AARD and AMD of correlations proposed for density of CO_2-loaded aqueous MEA mixtures. Therefore, Hartono's correlation and Weiland's correlation, which was modified by Han et al. [4] for CO_2-loaded aqueous MEA, were studied with different literature for density data of 30 mass% CO_2-loaded mixtures. Correlations were able to represent literature data with less than 1% AARD. Weiland's correlation showed a higher deviation for data presented by Amundsen et al. [2] and Zhang et al. [29] compared to Hartono et al. [1] and Han et al. [4]. Hartono's correlation showed a good agreement with data given by Zhang et al. [29]. The maximum deviation is beyond the expanded combined uncertainties reported in data sources, and calculated AARD shows that the agreement between correlated and experimental densities is satisfactory.

3.2. Viscosity Correlations. The nature of the model depends on the solution characteristics. Generally, the liquid viscosity decreases with the increase of temperature, and it increases with the increase of pressure. For pure MEA, an exponential model was frequently used to correlate the temperature dependence of viscosity. Table 13 summarizes the various published correlations for the viscosity of different MEA solutions. The relation between the viscosity of pure MEA with temperature can be represented by the Arrhenius equation shown in equation (30) and Teng et al. [59] calculated the activation energy for viscous flow from the data presented in DiGuilio et al. [9]. DiGuilio et al. [9] used a modified Andrade from (1934) viscosity model [60] by Vogel [61] as shown in equation (31).

Unlike ideal density, several mathematical relations have been proposed to determine ideal viscosity in a liquid mixture in the literature.

Kendall and Monroe [62]:

$$\ln (\eta_{\text{ideal}}) = \sum_{i}^{n} x_i \ln (\eta_i). \tag{24}$$

TABLE 11: Comparison of density correlations with different literature data for aqueous MEA mixtures.

Data source	Ma et al. [27]	Amundsen et al. [2]	Maham et al. [11]
T (K)	293.15–333.15	293.15–353.15	298.15–353.15
x_1	0.1000–0.8995	0.0687–0.7264	0.0054–0.9660
Han's correlation			
AARD (%)	0.11	0.16	0.10
MD (kg·m^{-3})	4.07	3.52	3.64
Hartono's correlation			
AARD (%)	0.05	0.03	0.03
MD (kg·m^{-3})	1.79	1.09	1.02

TABLE 12: Comparison of density correlations with different literature data for CO_2-loaded aqueous MEA mixtures.

Data source	Hartono et al. [1]	Amundsen et al. [2]	Zhang et al. [29]	Han et al. [4]
T (K)	293.15–353.15	293.15–353.15	298.15–353.15	298.15–413.15
α (mol CO_2/mol MEA)	0.11–0.5	0.1–0.5	0.14–0.49	0.1–0.56
Weiland's correlation				
AARD (%)	0.25	0.50	0.58	0.26
MD (kg·m^{-3})	6.46	16.08	13.86	10.81
Hartono's correlation				
AARD (%)	0.67	0.37	0.09	0.57
MD (kg·m^{-3})	8.13	7.05	3.54	19.53

Bingham [63]:

$$\frac{1}{\eta_{\text{ideal}}} = \sum_i^n x_i \frac{1}{\eta_i}. \tag{25}$$

Cronauer et al. [64] for ideal kinematic viscosity:

$$\ln(\nu_{\text{ideal}}) = \sum_i^n x_i \ln(\nu_i). \tag{26}$$

And the following expression is frequently used in recent publications [65].

$$\eta_{\text{ideal}} = \sum_i^n x_i \eta_i. \tag{27}$$

The viscosity of aqueous MEA deviates from the ideal mixture viscosity. This deviation of excess viscosity has been studied to make correlations fit the measured viscosity of the mixture. Accordingly, correlations are built to fit the viscosity deviation that is the viscosity difference between a real solution and an ideal solution.

$$\Delta\eta = \eta - \sum_{i=1}^2 x_i \eta_i, \tag{28}$$

$$\ln(\Delta\eta) = \ln(\eta) - \sum_{i=1}^2 x_i \ln(\eta_i). \tag{29}$$

McAllister presented a model to calculate kinematic viscosity in a binary mixture [2, 66, 67]. It is a semiempirical model, which is based on Eyring's absolute rate theory [68]. This model is given in two forms as the McAllister Three-Body Model and Four-Body Model considering different intermolecular interactions with neighbouring molecules.

Lee and Lin [14] and Amundsen et al. [2] adopted the Three-Body model as shown in equation (32) to fit viscosity data of aqueous MEA at different temperatures. Arachchige et al. [33] used a correlation suggested by Teng et al. [59] given in equation (33) to correlate measured viscosity of aqueous MEA at different temperatures. This correlation uses the viscosity of pure water and a polynomial to fit the viscosity of the binary mixture. The polynomial coefficients were found through a regression analysis at different temperatures. A Redlich–Kister type correlation as illustrated in equation (34) was proposed by Islam et al. [17] to determine $\Delta\eta$ (excess viscosity), and the parameters were found for different temperatures through a regression. A similar work was performed by Hartono et al. [1]. Then a Redlich–Kister type model was proposed to fit $\ln(\Delta\eta)$ given in equation (35). The main advantage of Hartono's aqueous MEA viscosity correlation is that it comprises the temperature dependence of viscosity that is not considered in Islam's correlation. Idris et al. [34] discussed the applicability of correlations based on the work by Heric-Brewer [69], Jouyban-Acree [70], Herráez et al. [71], and Redlich-Kister [57] as given in equations (36)–(39) respectively. The fitting parameters are in the form of a second-order polynomial of temperature to correlate temperature dependency of the viscosity as given in equation (40).

Limited attempts have been made to build correlations for the viscosity data of CO_2-loaded aqueous MEA mixtures. Accordingly, more measurements are still required to validate the existing data and correlations. Weiland et al. [26] developed a correlation for CO_2-loaded aqueous MEA for viscosity under different CO_2 loadings, MEA concentrations, and temperatures as described by equation (41). It is applicable for viscosities up to 40 mass% of MEA aqueous solutions at CO_2 loading of 0.6 mol CO_2/mol MEA to a

TABLE 13: Correlations for the Viscosity of MEA + H₂O + CO₂ mixtures.

Correlation		T (K)	P (MPa)	Data sources
$\eta = Ae^{E/RT}$	(30)	303.6–423.7	0.1013	[9]
$\ln(\eta) = A + (B/(T+C))$	(31)	303.6–423.7	0.1013	[9]
$\ln(v) = x_1^3 \cdot \ln(v_1) + 3x_1^2 x_2 \cdot \ln(v_{12}) + 3x_1 x_2^2 \cdot \ln(v_{21}) + x_2^3 \cdot \ln(v_2) - \ln(x_1 + x_2 \cdot [M_2/M_1]) + 3x_1^2 x_2 \cdot \ln([2$ $+M_2/M_1]/3) + 3x_1 x_2^2 \cdot \ln([1 + 2M_2/M_1]/3) + x_2^3 \cdot \ln(M_2/M_1)$	(32)	298.15–353.15	0.1013	[2]
$\ln(\eta) = \ln(\eta_0) + \sum_{i=1}^{i=n} a_i x_1^i$	(33)	293.15–353.15	0.1013	[33]
$\Delta\eta = x_1 x_2 \sum_{i=0}^{n} B_i (2x_1 - 1)^i$	(34)	303.15–323.15	0.1013	[17]
$\ln(\Delta\eta) = x_1 x_2 (b_1 + b_2 t + b_3 t^2 + b_4 x_1)$	(35)	293.15–353.15	0.1013	[1]
$\ln(\eta) = x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_1 \ln(M_1) + x_2 \ln(M_2) - \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 \sum_{i=0}^{n} [A_i (x_1 - x_2)^i]$	(36)			
$\ln(\eta) = x_1 \ln(\eta_1) + x_2 \ln(\eta_2) + x_1 x_2 \sum_{i=0}^{n} {}^{(A_i x_2^i)}$	(37)			
$\eta = \eta_1 + (\eta_2 - \eta_1) x_2^{\sum_{i=0}^{n} (A_i x_2^i)}$	(38)	298.15–373.15	0.4	[34]
$\eta = x_1 \eta_1 + x_2 \eta_2 + x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i$	(39)			
$A_{ij} = A_{i0} + A_{i1} T + A_{i2} T^2$	(40)			
$\eta/\eta_{H_2O} = \exp(\{[(a \cdot w_{MEA} + b)T + (c \cdot w_{MEA} + d)][\alpha(e \cdot w_{MEA} + f \cdot T + g) + 1] w_{MEA}\}/T^2)$	(41)	298.15	0.1013	[26]
$\ln(\eta_{loaded}) = x_3 \ln(\eta_l^*) + (1 - x_3) \ln(\eta_{unloaded})$	(42)	293.15–353.15	0.1013	[1]
$\ln(\eta_l^*) = (a_1 x_1 + a_2 \alpha x_1^2)/(a_3 + x_1)$	(43)			
$\ln(\eta_{loaded}/\eta_{unloaded}) = \sum_{i=1}^{2} A_i \alpha^i$	(44)	293.15–373.15	0.4	[34]
$A_i = a_{i,0} + a_{i,1} T$	(45)			
$\eta = (hN_A/V)\exp(\Delta F^{E*}/RT)$	(46)	313.15–343.15	0.1013	[58]
$\eta/\eta_{ideal} = (V_{ideal}/V)\exp(\Delta F^{E*}/RT)$	(47)			
$\ln(\eta V) = \ln(\eta V)_{ideal} + (\Delta F^{E*}/RT)$	(48)			
$\ln(\eta V) = \sum_i x_i \cdot \ln(\eta_i V_i^0) + (\Delta F^{E*}/RT)$	(49)			

Viscosity measurements performed under atmospheric conditions are mentioned with 0.1013 MPa pressure.

maximum temperature of 298.15 K. Amundsen et al. [2] adopted Weiland's correlation to fit the measured viscosities at different amine concentrations, CO_2 loadings, and temperatures. Hartono et al. [1] developed a correlation for different CO_2 loadings and temperatures by making a relation between viscosities of CO_2-loaded and unloaded aqueous MEA solutions as given in equation (42). The correlation was fit for CO_2-loaded viscosities of 30 and 40 mass% MEA and claimed 3.9% maximum AARD. Idris et al. [34] adopted a modified Setschenow-type [72] correlation as shown in equations (44) and (45) to fit CO_2-loaded aqueous MEA data at high MEA concentrations. This approach has been tested for the physical properties of amine solutions by Shokouhi et al. [73, 74]. A new approach was taken by Matin et al. [58] using Eyring's absolute rate theory [68] as illustrated in equations (46)–(49). Therefore, assuming the equivalence between the Gibbs free energy of activation for viscous flow and the equilibrium Gibbs free energy of mixing, the concepts of classical thermodynamics can be extended to the viscous flow behaviour of liquid mixtures [65]. The electrolyte-NRTL model is used to calculate the excess Gibbs free energy. Having tested for different terms, Matin et al. [58] revealed that the Gibbs free energy of mixing is the appropriate thermodynamic quantity to substitute for the excess free energy of activation for viscous flow for CO_2-loaded aqueous MEA mixture. The absolute rate theory with a reliable thermodynamic model is applicable for viscosity estimation of strong electrolyte systems, such as CO_2-loaded alkanolamine solutions.

The proposed correlations for viscosity of aqueous MEA were examined for accuracies compared to literature viscosity data. Table 14 lists the calculated AARD and maximum deviation for the McAllister model based on fitted parameters by Amundsen et al. [2] and Hartono's correlation for the considered three data sources. It was observed that the AARD for viscosity correlations are greater than the AARD for density correlations for aqueous MEA. For Amundsen's correlation, the highest AARD of 5.66% was reported for data presented by Ma et al. [27] and a maximum deviation was observed as 0.871 MPa·s at $x_1 = 0.6220$ and $T = 298.15$ K for data given by Maham et al. [35]. Hartono's correlation showed a highest AARD of 4.35% for the viscosity data presented by Ma et al. [27] and a maximum deviation of 0.854 MPa·s for data presented by Maham et al. [35] at $x_1 = 0.8446$ and $T = 303.15$ K.

The accuracies for the correlations proposed for viscosity of CO_2-loaded MEA were in the same order as with viscosity correlations for aqueous MEA. Weiland's correlation and Hartono's correlation were studied for their accuracies of the data predictions compared to the measured viscosities of 30 mass% CO_2-loaded aqueous MEA mixtures at different temperatures, and calculated AARD and maximum deviation are shown in Table 15. Weiland's correlation showed a highest AARD of 4% and a maximum deviation of 0.176 MPa·s at the CO_2 loading of 0.5 mol CO_2/mol MEA and $T = 298.15$ K for viscosities published by Amundsen et al. [2]. This could be due to the uncertainties related to the

TABLE 14: Comparison of viscosity correlations with different literature data for aqueous MEA mixtures.

Data source	Ma et al. [27]	Amundsen et al. [2]	Maham et al. [35]
T (K)	293.15–333.15	293.15–353.15	293.15–353.15
x_1	0.1000–0.8995	0.0687–0.7264	0.0313–0.8446
McAllister model			
AARD (%)	5.66	3.30	2.15
MD (mPa·s)	0.773	0.105	0.871
Hartono's correlation			
AARD (%)	4.35	2.38	2.39
MD (mPa·s)	0.825	0.774	0.854

experiments. Hartono's correlation showed a highest AARD of 3.80% for work done by Amundsen et al. [2] and a maximum deviation of 0.195 MPa·s at the CO_2 loading of 0.38 mol CO_2/mol MEA and $T = 303.15$ K for data presented by Zhang et al. [29].

3.3. Surface Tension Correlations.

Table 16 lists the relevant correlations for the surface tension. The behaviour of surface tension of pure and aqueous MEA is claimed to be linear with the temperature [4, 41], and data were fitted according to the correlation proposed for pure components as given in equation (50) [75]. The nonlinearity of surface tension with MEA concentration at a given temperature was correlated as illustrated in equation (51) [31, 76] by Vazquez et al. [41] and Han et al. [4] over a range of MEA concentrations and temperatures.

Surface tension measurements performed under atmospheric conditions are mentioned with 0.1013 MPa pressure.

For the surface tension of CO_2-loaded aqueous MEA, several attempts have been made by Jayarathna et al. [31] to build an appropriate correlation and a polynomial function was proposed including the CO_2 loading and temperature as independent variables as in equation (52). In this correlation, there is no variable defined to represent MEA concentration in the solution. The parameters of the polynomial were found through a regression analysis using measured surface tension data of 20–70 mass% MEA with CO_2 loading 0–0.5 mol CO_2/mol MEA at temperatures from 303.15 K to 333.15 K. Another correlation was introduced as given in equation (53) for the experiments with 80 mass% MEA with CO_2 loading 0–0.5 mol CO_2/mol MEA at temperatures from 313.15 K to 343.15 K in which the coefficients of the polynomial were found under different temperatures [3]. The applicability of the Connors and Wright model was discussed. The surface tension of liquid CO_2 was considered as a fitting parameter in equation (55) since it does not exist under such conditions [31].

3.4. Thermal Expansion Coefficient Correlations.

The thermal expansion coefficient describes the volume variation

TABLE 15: Comparison of viscosity correlations with different literature data for CO_2-loaded aqueous MEA mixtures.

Data source	Hartono et al. [1]	Amundsen et al. [2]	Zhang et al. [29]
T (K)	293.15–353.15	293.15–353.15	298.15–353.15
α (mol CO_2/mol MEA)	0.11–0.5	0.1–0.5	0.14–0.49
Weiland's correlation			
AARD (%)	1.57	4.00	2.80
MD (mPa·s)	0.077	0.176	0.119
Hartono's correlation			
AARD (%)	3.47	3.80	3.69
MD (mPa·s)	0.154	0.188	0.195

against temperature and is defined as in equation (56) for a liquid:

$$\beta = -\rho^{-1}\left(\frac{\partial \rho}{\partial T}\right). \tag{56}$$

For a mixture, equation (57) can be derived in terms of excess volume of the mixture and thermal expansions of the pure liquids [6]:

$$\beta = V^{-1}\left[\left(\frac{\partial V^E}{\partial T}\right) + \sum_i^n (x_i \beta_i V_i)\right]. \tag{57}$$

Only a few studies have been performed on MEA solutions and Yang et al. [6] present data of thermal expansion coefficient for pure MEA.

4. Discussion

The correlations found for the density of all types of studied mixtures including pure, aqueous MEA, and CO_2-loaded aqueous MEA were able to fit measured densities with acceptable accuracies to perform engineering calculations. Correlations for the density of aqueous mixtures based on the excess volume of aqueous MEA provide some theoretical insight to analyse the mixtures for molecular size and shape of the components. The need for a higher number of parameters to enhance accuracy is a drawback in this type of correlations. Available correlations are empirical and measured data are required to estimate the correlation parameters. The reported accuracies for density correlations of nonloaded solutions are as expected to be better than for correlations of CO_2-loaded solutions.

The comparison of the correlations with available density data in literature showed that Hartono's density correlation for aqueous MEA gave a minimum AARD of 0.03%. Han's correlation and Hartono's density correlation for CO_2-loaded aqueous MEA showed a minimum AARD of 0.09%.

For engineering purposes, all the density correlations are satisfactory. Most of the used methods are empirical. For scientific evaluations, theoretical models are more attractive to evaluate reasonable dependencies of different parameters. One promising example is to make use of Prigogine–Flory–Patterson's [54, 55] approach for aqueous MEA mixtures and extend to the CO_2-loaded aqueous MEA mixtures.

For the viscosities, correlations are available for viscosities of all types of studied mixtures. The reported accuracies of data fitting are satisfactory to perform calculations in design, mathematical modelling, and simulation. Correlations reported by Heric-Brewer [69], Jouyban-Acree [70], and McAllister [67] demonstrate a theoretical background in the model structure. Lack of theoretical insight is a drawback in viscosity correlations related to the CO_2-loaded aqueous MEA solutions. The method proposed by Matin et al. [58] to use Gibbs free energy of mixing from the electrolyte-NRTL model for the excess free energy of activation for viscous flow in the Eyring [68] viscosity model to predict viscosities of CO_2-loaded solutions has benefits and drawbacks. The electrolyte-NRTL is a complex model with many parameters involved. Commercial process simulation packages such as ASPEN Plus have the electrolyte-NRTL model with relevant interaction parameters for CO_2-loaded aqueous MEA systems that make it easy to adopt the model to perform viscosity calculations.

The performed correlation comparison for viscosity with available viscosity data in the literature showed that Hartono's correlation for viscosity of aqueous MEA and Weiland's correlation for viscosity of CO_2-loaded aqueous MEA were reasonably good for considered viscosity data. For today's use, the correlations proposed by Weiland et al. [26] and Hartono et al. [1] are attractive in the aspects of accuracy, easy understanding, and implementation. In the future, it is recommended to work towards use of more theoretically based models. One example with potential is Eyring's viscosity model combined with NRTL model for viscosity predictions.

5. Conclusion

This study summarizes measured data and correlations developed for the density, viscosity, surface tension, and thermal expansion coefficient of pure, aqueous MEA, and CO_2-loaded aqueous MEA solutions. For the density, an adequate amount of data is available for pure and aqueous MEA mixtures under different concentrations and temperatures. There is a lack of density data of CO_2-loaded aqueous MEA especially at higher concentrations of MEA. The correlations available in literature for all studied types of solutions are in good agreement with measured densities.

For viscosities, data are available for pure MEA and aqueous MEA to cover mole fractions from 0 to 1 up to temperature 423.15 K. The available data for CO_2-loaded aqueous MEA mixtures are limited to some special MEA concentrations and CO_2 loadings. Recent studies have

TABLE 16: Correlations for the surface tension of MEA + H_2O + CO_2 mixtures.

Correlation		T (K)	P (MPa)	Data sources
$\sigma = K_1 - K_2 t$	(50)	298.15–333.15	0.1013	[4, 41]
$\sigma_{mix} = \sigma_1 + \sum_{i\geq 2} (1 + (a_i x_i/(1 - b_i)(1 + \sum_{j\geq 2} b_j/(1 - b_j) x_j))) x_i (\sigma_i - \sigma_1)$	(51)	303.15–333.15	0.1013	[4]
$\sigma_{mix} = a_1 + a_2 \cdot \alpha + a_3 \cdot T + a_4 \cdot \alpha^2 + a_5 \cdot \alpha \cdot T + a_6 \cdot T^2 + a_7 \cdot \alpha^3 + a_8 \cdot \alpha^2 \cdot T + a_9 \cdot \alpha^4 + a_{10} \cdot \alpha^3 \cdot T + a_4 \cdot \alpha^2 \cdot T^2$	(52)	303.15–333.15	0.1013	[31]
$\sigma_{mix} = a \cdot \alpha^2 + b \cdot \alpha + c$	(53)	313.15–343.15	0.1013	[3]
$\sigma_{mix} = \sigma_2 + \sum_{i=1,3} (1 + (a_i x_i/(1 - b_i)(1 + \sum_{j=1,3} (b_j/(1 - b_j) x_j))) x_i (\sigma_i - \sigma_2)$	(54)	303.15–333.15	0.1013	[31]
$\sigma_{CO_2} = a_1 + a_2 T$	(55)			

measured viscosities of CO_2-loaded mixtures at high MEA concentrations. Further studies are required to fill the gaps and validate the existing data. The correlations and semi-empirical models used for pure and aqueous MEA are capable of fitting data with acceptable accuracy. The developed correlations for CO_2-loaded mixtures need improvements to fit measured data in a wide range of MEA concentrations and temperatures. For engineering purposes, it is recommended to make use of more theoretically based models.

Surface tension data for pure and aqueous MEA mixtures are available in the literature. The data have been correlated to different types of correlations with acceptable accuracy. It is recommended to perform further studies to measure the surface tension of CO_2-loaded aqueous MEA mixtures to fill the gaps and check the validity of the existing data. For the thermal expansion coefficient, studies are needed to determine the thermal expansion coefficient for CO_2-loaded aqueous MEA mixtures.

Nomenclature

E: Activation energy of viscous flow, $J \cdot mol^{-1}$
ΔF^*:   Free energy of activation for viscous flow, $J \cdot mol^{-1}$
ΔF^{E*}:   Excess free energy of activation for viscous flow, $J \cdot mol^{-1}$
h: Planck's constant, $m^2 \cdot kg \cdot s^{-1}$
K: Regression parameter
M_1:   Molecular weight of MEA, $g \cdot mol^{-1}$
M_2:   Molecular weight of water, $g \cdot mol^{-1}$
N: Number of data
N_A: Avogadro number, mol^{-1}
R: Universal gas constant, $J \cdot mol^{-1} \cdot K^{-1}$
t: Temperature, $°C$
T: Temperature, K
T_c: Critical temperature, K
T_0:   Reference temperature 308 K
u: Uncertainty
U: Expanded uncertainty
v: Kinematic viscosity, $m^2 \cdot s^{-1}$
v_{ideal}:   Kinematic viscosity of an ideal mixture, $m^2 \cdot s^{-1}$
V_i^0:   Molar volume of ith pure component, $m^3 \cdot mol^{-1}$
V_1^0:   Molar volume of MEA, $m^3 \cdot mol^{-1}$
V_2^0:   Molar volume of water, $m^3 \cdot mol^{-1}$
V^E: Excess molar volume, $m^3 \cdot mol^{-1}$
V_m: Molar volume of the mixture, $m^3 \cdot mol^{-1}$
w_{MEA}:   Mass percent MEA
x_i: Mole fraction of ith component
x_1:   Mole fraction of MEA
x_2:   Mole fraction of water
Y_i^E: Measured property
Y_i^C: Calculated property

Greek letters

ρ: Density, $kg \cdot m^{-3}$
ρ_1:   Density of pure MEA, $kg \cdot m^{-3}$
ρ_2:   Density of pure water, $kg \cdot m^{-3}$
η: Viscosity, $Pa \cdot s$
$\Delta \eta$:   Viscosity difference, $Pa \cdot s$
η_{ideal}:   Viscosity of an ideal mixture, $Pa \cdot s$
η_{H_2O}:   Viscosity of water, $Pa \cdot s$
η_γ: Viscosity deviation, $Pa \cdot s$
$\eta_{unloaded}$:   Viscosity of unloaded solution, $Pa \cdot s$
σ: Surface tension, $N \cdot m^{-1}$
σ_{mix}:   Surface tension of a mixture, $N \cdot m^{-1}$
β: Bulk thermal expansively, K^{-1}
β: Thermal expansion coefficient, K^{-1}
α: CO_2 loading, mol CO_2/mol MEA.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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