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Experimental data and modeling for density and viscosity of carbon dioxide (CO_2)-loaded and -unloaded aqueous blend of 2-(ethylamino) ethanol (EAE) and aminoethylethanolamine (AEEA) for post-combustion CO_2 capture

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ABSTRACT

Amine based chemical absorption is the most developed technique for post-combustion CO_2 capture from flue gases of low CO_2 partial pressure. Density and viscosity data of CO_2 – loaded and –unloaded absorbent are important in kinetics study and design the absorption column. Density and viscosity of CO_2 loaded and –unloaded aqueous blend of 2-(ethylamino)ethanol (EAE) + aminoethylethanolamine (AEEA) were obtained experimentally in the temperature range of 293.15 to 323.15 K with 5 K temperature interval at atmospheric pressure. Concentration of aqueous EAE + AEEA blend was 10 wt%, 20 wt%, and 30 wt% with 7/3 weight ratio of EAE/AEEA. Excess volume was calculated by using experimental density data and correlated with Redlich-Kister type equation. Correlations were developed to calculate density and viscosity. For CO_2-loaded and –unloaded aqueous EAE + AEEA blend, correlations predicted data were with average absolute deviation percentage (AAD %) of 0.1286, 0.0208, respectively for density while with AAD % 4.744, 4.433, respectively for viscosity. Wieland model was also correlated to CO_2-loaded viscosity data and AAD % was 2.852 for this model. Moreover, diffusivity of CO_2 into the aqueous EAE + AEEA blend was calculated using modified Stokes-Einstein equation.

1. Introduction

Carbon dioxide (CO_2) is the greenhouse gas and emitted in the atmosphere because of the human activities like coal fired power plant operation, steel and aluminum production, cement industries and natural gas processing. The world may be warmer at least 3 to 4 °C by 2100 due to greenhouse gas emission [1]. Intended nationally determined countries (INDCs) of many countries have assured to reduce greenhouse gas emission to an extent of 9% per capita by 2030 [2]. Post combustion CO_2 capture by amine based chemical absorption is most matured technique and cost effective for implementing it in the existing power plants [3–5]. Liang et al. [6,7] had published excellent reviews on the latest advances and developments in post-combustion CO_2 capture using amine solvents. From their reviews, it can be concluded that there is no single solvent which has all favorable properties for CO_2 capture by absorption-desorption process. To minimize demerits and to utilize advantages of individual amines, recently, several amine blends have been investigated for CO_2 capture [8–13].

The physicochemical properties such as density, viscosity and diffusivity of CO_2 into the absorbent are required to design and optimize absorption column and CO_2 capture process [14]. Densities data are used to determine the physical solubility of CO_2 in the solvent, mass transfer and solvent reaction kinetics. Volumetric properties from density data can be used to explore our knowledge of molecular interaction in the mixture [15]. Viscosity data are required to calculate pressure drop of flow, heat transfer coefficient and mass transfer coefficient. Moreover, viscosity of absorbent is also useful to find out mass diffusivity of CO_2 in amine solvent using Stokes-Einstein Eq. [16].

Chowdhuri et al. [17] published volumetric and viscometric properties of some aqueous monoalkanolamines. The thermodynamic properties and CO_2 solubility of the blend of

monoethanolamine (MEA) and diethylenetriamine (DETA)/ aminoethylethanolamine (AEEA) were measured Moosavi et al. [18]. Volumetric and viscometric properties of aqueous blend of piperazine (PZ) + 2-Amino-2-methyl-1-propanol (AMP) were determined by Liu et al. [19]. The study on density, viscosity and refractive index of aqueous CO_2 loaded and unloaded 2-(ethylamino) ethanol (EAE) for post combustion CO_2 capture was carried out by Gao et al. [20]. Viscosity data of unloaded and CO_2 loaded of aqueous solution of *N*-methyldiethanolamine and AMP was reported by Kummamuru et al. [21].

EAE is a hindered secondary amine and has higher CO_2 loading with lower heat of absorption because it produces unstable carbamate [22,23]. AEEA is an alkanoldiamine and has been shown high CO_2

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Table 1
Details of used chemicals in this work.

Chemical name	CAS number	Source	Initial purity	Purification method
2-(ethylamino)ethanol (EAE)	110–73-6	Sigma Aldrich, St. Louice, USA	≥ 98%[c]	none
Aminoehtylethanolamine (AEEA)	111–41-1	sd Fine chemical limited, Mumbai, India	98%[c]	none
Acetone	67–64-1	sd Fine chemical limited, Mumbai, India	99%[c]	none
Methanol	67–56-1	sd Fine chemical limited, Mumbai, India	99%[c]	none
Hydrochloric acid	7647-01-0	sd Fine chemical limited, Mumbai, India	35–38%[c]	none
Carbon-dioxide gas	124–38-9	Linde India Ltd.	99.99%[d]	none
Nitrogen gas	7727-37-9	Linde India Ltd.	99.99%[d]	none
Water[a]	7732-18-5	Sigma Aldrich, St. Louice, USA	99.99%[c]	none
Water[b]	7732-18-5	Our laboratory	99.99%[c]	Double distillation

[a] De-ionized water for density-meter calibration.
[b] Double-distilled water for making aqueous solution.
[c] Mass fraction.
[d] Volume fraction.

loading (mol CO_2/mol amine), faster reaction kinetics but its high heat of absorption [24,25] makes it not very useful as a single absorbent for CO_2 capture. In the literature, AEEA has been used as an activator in the amine blends to improve solvent performance for CO_2 capture [18,26–28]. In our previous research paper [29], it has been shown that aqueous mixture of EAE + AEEA is a better absorbent in terms of CO_2 loading, cyclic capacity and heat of absorption. But, there is lack of viscosity and density data of aqueous blend of EAE + AEEA in literature. Experimental data of viscosity and density of CO_2 loaded and –unloaded new absorbent (aqueous EAE + AEEA), study of its molecular interaction and development of new empirical models to predict density and viscosity of CO_2 loaded and –unloaded absorbent are novelties of this paper.

In the present work, density and viscosity of CO_2- loaded and -unloaded aqueous blend of EAE + AEEA were measured in the temperature range 293.15–323.15 K at atmospheric pressure. Temperature interval was kept at 5 K. New models were developed to predict density and viscosity of CO_2- loaded and -unloaded aqueous blend of EAE + AEEA. Wieland model [29] was used to calculate viscosity of CO_2 loaded samples. Total concentration of solution was 10 wt%, 20 wt%, and 30 wt % and it was used in term of weight fraction ($w = 0.10$, $w = 0.20$, and $w = 0.30$, respectively) in the calculation with used correlations. Weight fraction of AEEA in the blend was kept constant at 0.30 (AEEA/EAE weight ratio at 3/7). Mass diffusivity of CO_2 into this amine blend was calculated using modified Stokes-Einstein equation.

2. Experimental section

2.1. Chemicals and -unloaded sample preparation

The EAE (98% purity) was purchased from Sigma Aldrich, St. Louice USA. AEEA (98% purity) and hydrochloric acid (HCl, 35–38% purity) was purchased from Sd Fine chemical limited, Mumbai, India. HCl was used for titration of amine samples to measure CO_2 loading. All chemicals were used without further purification. Description of all chemicals which were used in the experimentation was listed in

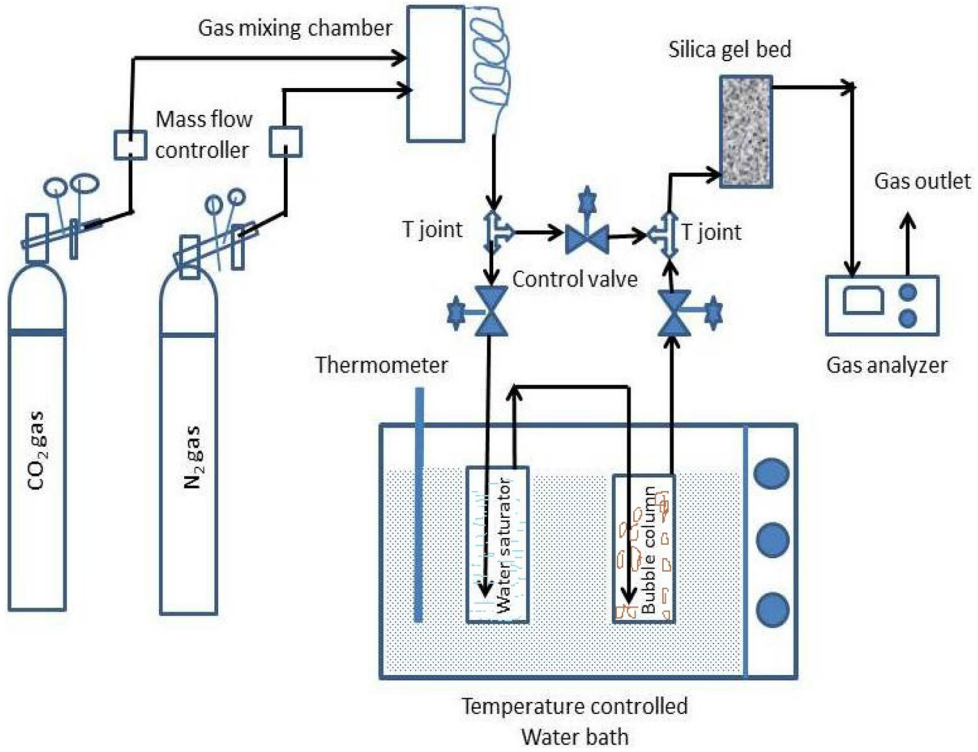

Fig. 1. Experimental set-up for CO_2 gas absorption to prepare CO_2 loaded solutions [29].

Table 2
Experimental data of density and viscosity of EAE and AEEA at T = (293.15–323.15) K and 101.325 kPa pressure and comparison with data available in the literature.*[a]

T/(K)	ρ/(kg.m^{-3})				μ/(mPa.s)			
	EAE		AEEA		EAE		AEEA	
	This work	Literature data	This work	Literature data	This work	Literature data	This work	Literature data
293.15	917.2	916.584[a] 917.79[b]	1029.2	1028.76[d]	17.18	15.281[b]	153.62	
298.15	913.8	912.483[a] 913.39[b]	1025.4	1025.28[e] 1025.29[f]	12.42	12.348[b]	99.27	98.6[e] 98.62[g]
303.15	910.2	908.744[a] 909.82[b] 909.1[c]	1021.7	1021.23[d] 1021.53[e]	9.64	9.590[b] 9.639[c]	71.31	70.5[e] 70.57[g]
308.15	906.3	904.9[c]	1017.9		7.96	7.830[c]	52.94	53.11[g]
313.15	902.0	901.79[b] 900.8[c]	1013.9	1013.68[d] 1014.002[e] 1013.98[f]	6.51	6.406[b] 6.419[c]	37.67	39.4[e]
318.15	897.5	896.8[c]	1009.7		5.32	5.358[c]	28.23	
323.15	892.8	893.68[b] 892.6[c]	1005.4	1006.11[d]	4.28	4.527[b] 4.517[c]	21.55	23.4[e]

* Standard uncertainties u are u(T) = 0.2 K for density measurement, u(T) = 1 K for viscosity measurement, and u(P) = 1 kPa, and expanded uncertainties at 95% confidence level are U(ρ) = 0.003 ρ (kg.m^{-3}) and U(μ) = 0.08 μ (mPa.s).
[a] Reference [31].
[b] Reference [32].
[c] Reference [17].
[d] Reference [15].
[e] Reference [33].
[f] Reference [34].
[g] Reference [18].

Table 1. EAE, AEEA, and distilled water were used for making aqueous blend of EAE + AEEA.

2.2. CO_2-loaded sample preparation

CO_2-loaded samples were prepared by absorption of CO_2 into the –unloaded sample. The absorption process was carried out in a bubble column reactor of 150 ml volume capacity. 120 ml of fresh –unloaded sample of aqueous EAE + AEEA was filled in the bubble column and CO_2 gas was passed into the solution. CO_2 loading process was started after first bubble formation and continued for (3 to 4 h) until the almost saturation was occurred. Experimental set-up for CO_2 absorption was given in Fig. 1 and detailed CO_2 loading analysis and CO_2 absorption mechanism was given elsewhere [29]. Partially CO_2 loaded samples were prepared by mixing CO_2 – loaded solution with –unloaded solution and stored at 293.15 K until it needed for measurement of CO_2 loading, density and viscosity.

2.3. Density measurement

The density of CO_2 – loaded and –unloaded samples were measured using Anton Paar density meter (Model DMA™ 35) in the temperature range 293.15 K to 323.15 K and at atmospheric pressure. The density measurement by density meter was carried out based on the oscillating U – tube principle. The measuring cell, a U shaped borosilicate glass tube was filled with sample. Before each sample injection, the tube of density meter was cleaned using acetone and calibration was done by using de-ionized water at 293.15 K and atmospheric pressure. Sample was placed in a temperature controlled water bath (with an accuracy of ±1 K). When desired temperature attained at constant temperature for more than 10 min, sample was injected into the measuring cell of density meter. Density data with temperature of the sample was shown on the instrument's display. Accuracy of density and temperature measurement of instrument were 0.001 g.cm^{-3} and 0.2 °C, respectively. Density of each sample was measured three times and average value was reported.

2.4. Viscosity measurement

Viscosity of CO_2 – loaded and –unloaded aqueous EAE + AEEA blend were obtained by using Sine-wave Vibro viscometer of A & D Company (model SV-10, with accuracy of 1% repeatability) in the temperature range of 293.15 to 323.15 K and at atmospheric pressure. The

Table 3
Density, excess volume, viscosity, and diffusivity of aqueous EAE + AEEA blend at T = (293.15–323.15) K and 101.325 kPa pressure[a].

T/(K)	ρ/(kg.m^{-3})	$V_E \times 10^6$ / (m^3.mol^{-1})	μ/(mPa.s)	$D_{CO2-(EAE+AEEA+H2O)}$ ×10^9 /(m^{2}s^{-1})
		$w = 0.10$		
293.15	997.8	−0.0986	1.42	1.316
298.15	996.7	−0.1023	1.25	1.497
303.15	995.3	−0.1058	1.12	1.672
308.15	993.6	−0.1092	1.02	1.874
313.15	991.9	−0.1189	0.94	2.059
318.15	990.1	−0.1289	0.87	2.286
323.15	988.2	−0.1410	0.82	2.483
		$w = 0.20$		
293.15	996.4	−0.1899	2.02	1.014
298.15	995.6	−0.2066	1.73	1.177
303.15	994.1	−0.2142	1.55	1.315
308.15	992.2	−0.2195	1.4	1.482
313.15	990.3	−0.2318	1.26	1.658
318.15	988.5	−0.2493	1.16	1.848
323.15	986.6	−0.2692	1.07	2.039
		$w = 0.30$		
293.15	995.2	−0.3067	3.06	0.745
298.15	994	−0.3225	2.51	0.894
303.15	992.6	−0.3397	2.11	1.046
308.15	990.8	−0.3545	1.83	1.216
313.15	988.9	−0.3750	1.62	1.377
318.15	986.8	−0.3942	1.45	1.566
323.15	984.5	−0.4137	1.31	1.755

[a] Standard uncertainties u are u(T) = 1 K for viscosity measurement, u(T) = 0.2 K for density measurement, u(P) = 1 kPa, and u(w_1) = 0.01, and expanded uncertainties at 95% confidence level are U(μ) = 0.08 μ mPa.s and U(ρ) = 0.003 ρ (kg.m^{-3}).

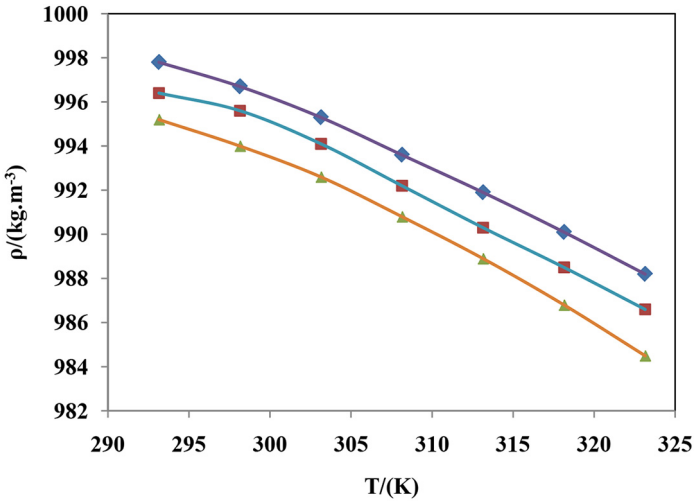

Fig. 2. Density of aqueous EAE + AEEA blend versus temperature for different concentration (in weight fraction) of EAE + AEEA, w: for (♦) 0.10; (■) 0.20; (▲) 0.30; and lines (—) for calculated values with eq. 6.

viscometer was cleaned using methanol and calibrated with double distilled water before each sampling. Temperature of samples was maintained constant using a temperature controlled water bath. Sample at desired temperature was filled in sample cup of viscometer then viscosity and temperature of the sample was measured and displayed at display of viscometer. The process of viscosity measuring of a sample was repeated three times and average value was reported.

In order to ensure the purity of used chemicals and the experimental apparatus accuracy, density and viscosity of pure EAE and AEEA were measured and compared with available literature data [15,17,18,31–34]. Comparison of experimental data of this work with literature data in the temperature range of 293.15 to 323.15 K at atmospheric pressure was given in Table 2. Average absolute deviation % (AAD %) for experimental and literature data of density and viscosity was measured by eq. 1 and reported as 0.063% and 2.695% for density and viscosity, respectively. The lower values of AAD % indicate that the measured density and viscosity data of this work were reliable.

$$AAD\% = \frac{100}{n} \times \sum_{i=1}^{n} \left| \frac{Y_{lit,i} - Y_{exp,i}}{Y_{lit,i}} \right| \quad (1)$$

where, $Y_{exp,\,i}$, $Y_{lit,\,i}$, and n are experimental data (density or viscosity), literature data and number of data point, respectively.

3. Results and discussions

3.1. Density

3.1.1. Density of CO_2 -unloaded aqueous EAE + AEEA

Density of aqueous EAE + AEEA blend was measured in the temperature range 293.15 to 323.15 K in the 5 K temperature steps at atmospheric pressure. Total concentration of solution was 10 wt%, 20 wt%, and 30 wt%. Amount of AEEA in the amine mixture was fixed at 0.30 wt. fraction. It could be shown from Table 3 and Fig. 2 that density of aqueous EAE + AEEA blend was decreased by increasing temperature as well as increasing total concentration of amine blend. Density decreased by increasing sample temperature due to increase of volume of sample with constant mass. Density of pure EAE is much lower than the density of water, that's why density decreased by increasing amine concentration in the blend.

Weiland et al. [30] proposed a correlation (eq. 2) to calculate the density of alkanolamine solution.

Table 4
Regressed parameters (c_0, c_1, and c_2) of Eq. 6 and Eq. 9 at different temperature.

T/(K)	Eq. 6			Eq. 9		
	c_0	c_1	c_2	c_0	c_1	c_2
293.15	1.30	10.30	10	0.1814	−2.7873	8.9008
298.15	0.20	46.20	−25	0.1249	−1.6979	5.2803
303.15	0.40	44.65	−15	−0.0214	0.5453	−1.1248
308.15	0.80	40.42	0	−0.0624	1.4305	−3.6735
313.15	1.40	37.73	10	−0.0147	1.1527	−2.9035
318.15	1.46	46.64	0	−0.0391	1.5865	−3.9635
323.15	1.20	53.02	−25	−0.0006	1.4800	−3.6775

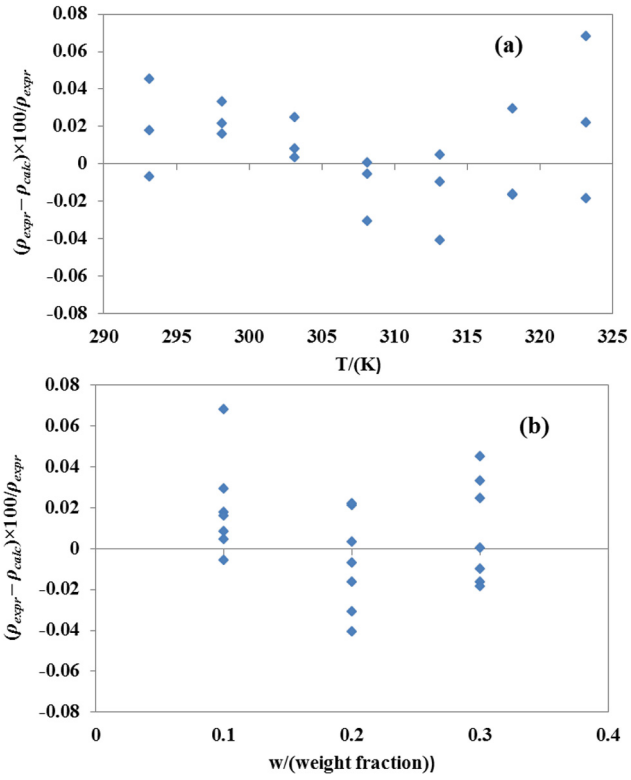

Fig. 3. Relative deviations of experimental and calculated density data of CO_2-unloaded aqueous EAE + AEEA blend from eq. 7 as a function of **(a)** temperature and **(b)** concentration (EAE + AEEA weight fraction).

Table 5
Density and viscosity of CO_2-loaded aqueous EAE + AEEA blend at T = (293.15–323.15) K and 101.325 kPa pressure[a].

α**	T/(K)	ρ/(kg.m^{-3}) $w = 0.10$	μ/(mPa.s)	α**	T/(K)	ρ/(kg.m^{-3}) $w = 0.20$	μ/(mPa.s)
0.270	293.15	1010.5	1.45	0.657	293.15	1047.2	2.26
	298.15	1008.4	1.30		298.15	1045.2	1.97
	303.15	1006.2	1.15		303.15	1043.1	1.78
	308.15	1003.9	1.04		308.15	1040.9	1.59
	313.15	1001.6	0.96		313.15	1038.5	1.46
	318.15	999.2	0.88		318.15	1036.6	1.34
	323.15	996.8	0.83		323.15	1034.8	1.24
0.520	293.15	1026.8	1.46	0.754	293.15	1054.9	2.27
	298.15	1025.7	1.32		298.15	1052.8	1.99
	303.15	1023.9	1.18		303.15	1050.6	1.81
	308.15	1022.1	1.07		308.15	1048.3	1.63
	313.15	1020.2	0.97		313.15	1046	1.50
	318.15	1018.1	0.90		318.15	1043.6	1.38
	323.15	1015.8	0.85		323.15	1041.1	1.27
0.770	293.15	1042.1	1.47			$w = 0.30$	
	298.15	1040	1.34	0.129	293.15	1039.8	3.35
	303.15	1038.2	1.19		298.15	1038.7	2.62
	308.15	1036.6	1.08		303.15	1036.8	2.42
	313.15	1034.7	0.99		308.15	1034.9	2.13
	318.15	1032.3	0.91		313.15	1032.6	1.85
	323.15	1030.6	0.86		318.15	1030.8	1.65
0.981	293.15	1057.6	1.49		323.15	1028.7	1.47
	298.15	1055.5	1.35	0.464	293.15	1062.4	3.96
	303.15	1053.3	1.21		298.15	1060.2	3.34
	308.15	1051.1	1.10		303.15	1057.9	2.95
	313.15	1049.8	1.00		308.15	1054.4	2.66
	318.15	1047.5	0.93		313.15	1051.7	2.38
	323.15	1045.1	0.87		318.15	1048.8	2.12
		$w = 0.20$			323.15	1045.8	1.91
0.155	293.15	1016.7	2.05	0.644	293.15	1071.8	4.20
	298.15	1015.8	1.77		298.15	1070	3.62
	303.15	1013.7	1.57		303.15	1068.1	3.20
	308.15	1011.6	1.42		308.15	1066.7	2.88
	313.15	1009.5	1.27		313.15	1064	2.58
	318.15	1007.2	1.17		318.15	1062.2	2.36
	323.15	1005.8	1.08		323.15	1060.1	2.13
0.502	293.15	1036	2.22	0.760	293.15	1080.9	4.42
	298.15	1034.6	1.95		298.15	1078.6	3.73
	303.15	1032.7	1.75		303.15	1076.2	3.34
	308.15	1030.9	1.57		308.15	1074.7	2.99
	313.15	1028.4	1.43		313.15	1072.8	2.69
	318.15	1026.6	1.31		318.15	1070.3	2.46
	323.15	1024.5	1.20		323.15	1068.4	2.22

[a] Standard uncertainties u are u(T) = 1 K for viscosity measurement, u(T) = 0.2 K for density measurement, u(P) = 1 kPa, u(w) = 0.01, u(α) = 0.01 and expanded uncertainties at 95% confidence level are U(μ) = 0.08 μ mPa.s and U(ρ) = 0.003 ρ (kg.m^{-3}).
**α is CO_2 loading, which was defined as the (mol CO_2/mol amine).

$$\rho_m = \frac{\sum_{i=1}^{n}(x_i M_i)}{V_m} \tag{2}$$

where, ρ_m, V_m, x_i, *and* M_i *are* density of the mixture (kg.m^{-3}), molar volume of the mixture (m^3.mol^{-1}), mole fraction of component *i*, and molecular weight (kg.mol^{-1}), respectively.

In order to analyze dependency of temperature and amine content in the mixture on the interaction of its molecules, excess volume (V^E) could be calculated by eq. 3.

$$V^E = V_m - \sum_{i=1}^{n}(x_i V_i) \tag{3}$$

where, V_i is the molar volume (m^3.mol^{-1}) of component *i*.

Excess volume of aqueous EAE + AEEA blend was calculated by eq. 4 and listed in Table 3.

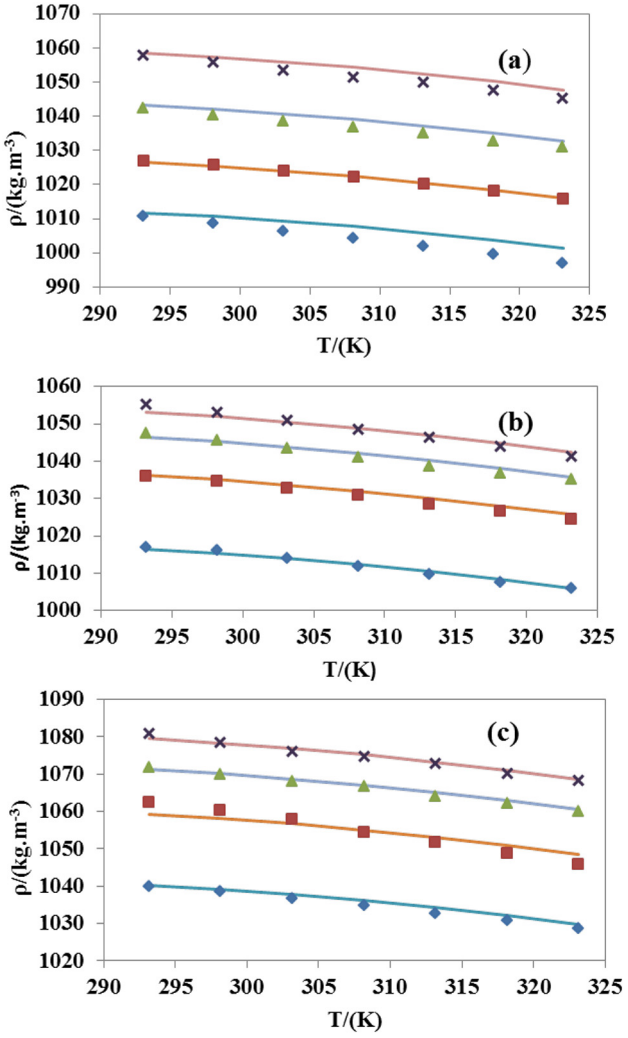

Fig. 4. Experimental and calculated density data of CO_2-loaded aqueous EAE + AEEA blend as a function of temperature for different (EAE + AEEA) concentration and CO_2 loading (α); **(a)** w = 0.10 and α: for (♦) 0.27; (■) 0.52; (▲) 0.77; and (×) 0.981; **(b)** w = 0.20 and α: for (♦) 0.155; (■) 0.502; (▲) 0.657; and (×) 0.754; **(c)** w = 0.10 and α: for (♦) 0.129; (■) 0.464; (▲) 0.644; and (×) 0.76; and lines (—) for calculated values with eq. 8.

$$V^E = \left[\frac{(x_1M_1 + x_2M_2 + x_3M_3)}{\rho_m}\right] - \left[\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} + \frac{x_3M_3}{\rho_3}\right] \tag{4}$$

where, x_1, x_2, and x_3 were mole fraction of EAE, AEEA, and water, respectively. M_1, M_2, and M_3 were molecular weight (kg.mol^{-1}) of EAE, AEEA, and water, respectively. ρ_1, ρ_2, and ρ_3 were density (kg.m^{-3}) of EAE, AEEA, and water, respectively. Excess volume over all temperature and concentration was negative. It revealed formation of hydrogen bond and contracting behavior of aqueous EAE + AEEA solution.

Physical properties (i.e., density and viscosity) of binary system were calculated by using Redlich-Kister [35] equation by some researchers [15,17,20,32] in the literature. However, for the ternary system, use of the Redlich-Kister equation to calculate physical property was very rigorous and time taking method. In order to simply the calculation for ternary system, in this work V^E (m^3.mol^{-1}) was calculated by using eq. 5.

$$V^E = 10^{-6}\sum_{i=0}^{m} c_i w^i \tag{5}$$

where, c_i was the Redlich-Kister type coefficient [35] and called mass interaction factor by Pandey-Mondal [36] in the literature. w was the concentration of ternary mixture in weight fraction and m was an integer

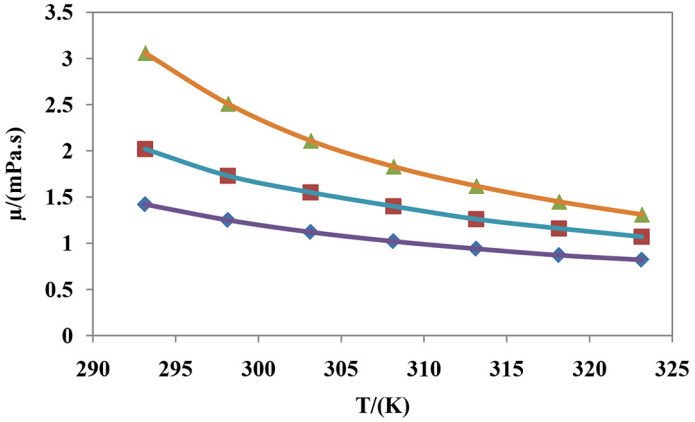

Fig. 5. Viscosity of aqueous EAE + AEEA blend versus temperature for different concentration (in weight fraction) of EAE + AEEA, w: for (♦) 0.10; (■) 0.20; (▲) 0.30; and lines (—) for calculated values with eq. 9.

with the variation from 1 to any number that could be well fitted by the measured experimental data. Values of c_i were calculated by using least-squares fitting. In this work second order polynomial was fitted very well to excess volume data. Regressed coefficients c_0, c_1, and c_2 were given in Table 4. Density of CO_2-unloaded aqueous EAE + AEEA was calculated by eq. 6 with AAD % of 0.00013 and shown in Fig. 1.

$$\rho_{m,calc} = \left[\frac{(x_1M_1 + x_2M_2 + x_3M_3)}{\left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} + \frac{x_3M_3}{\rho_3}\right) + 10^{-6}\sum_{i=0}^{2} c_i w^i} \right] \quad (6)$$

For simplicity of application we used a model (eq. 7) to calculate density ($kg.m^{-3}$) of aqueous EAE + AEEA blend, which was function of temperature and concentration of amine blend (in weight fraction).

$$\rho = \left(a + bT + cT^2\right).(1 + dw^e) \quad (7)$$

where, a, b, c, d, and e were coefficients of model. ρ, T and w were density ($kg.m^{-3}$), temperature (K), and concentration of amine blend (weight fraction), respectively. Value of coefficient was obtained by multiple regressions using Excel solver. Experimental data of density fitted well for $a = -0.00481$, $b = 2.627681$, $c = 641.1562$, $d = -0.02833$, and $e = 1.771689$. Calculated density data by eq. 7 were depicted in Fig. S1.

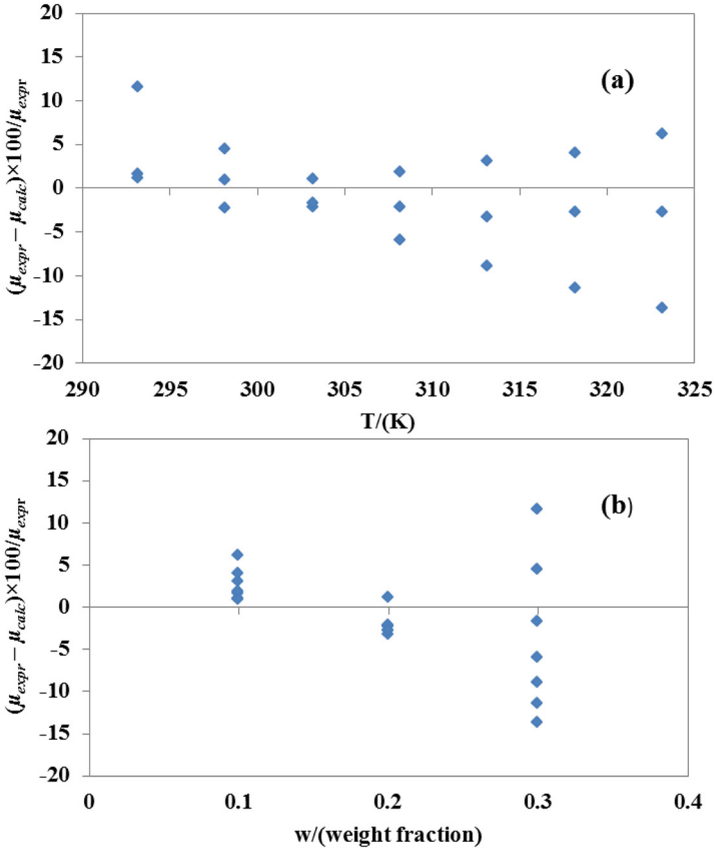

Fig. 6. Relative deviations of experimental and calculated viscosity data of CO_2-unloaded aqueous EAE + AEEA blend from eq. 10 as a function of **(a)** temperature and **(b)** concentration (EAE + AEEA weight fraction).

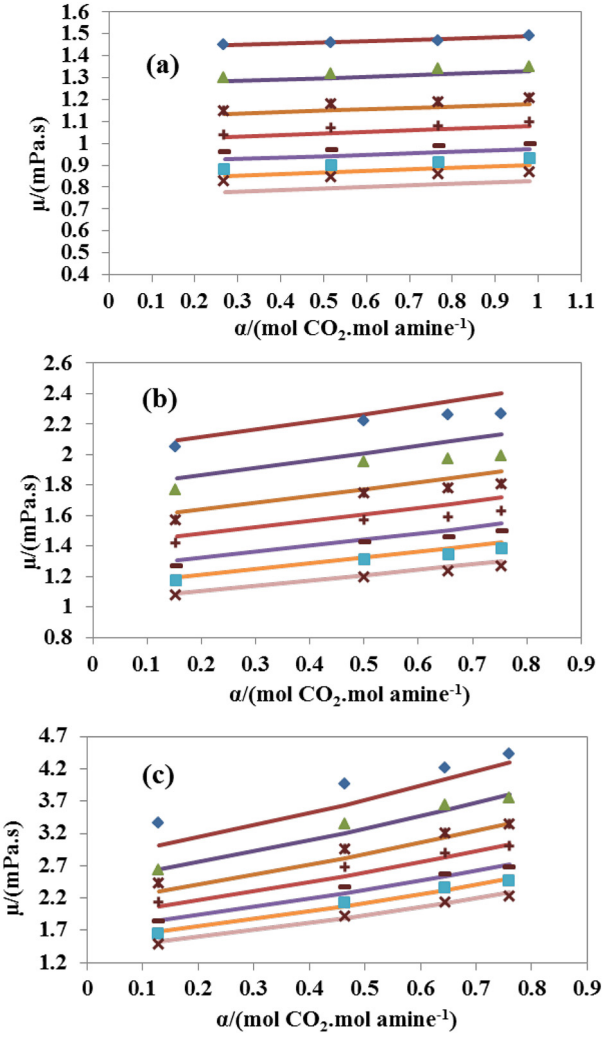

Fig. 7. Experimental and calculated viscosity data of CO_2-loaded aqueous EAE + AEEA blend as a function of CO_2 loading (α) at different temperature; T: for (♦) 293.15 K; (▲) 298.15 K; and (*) 303.15 K; (+) 308.15 K; (−) 313.15 K; (■) 318.15 K; (×) 323.15 K; and lines (—) for calculated values with eq. 11. For different (EAE + AEEA) concentration **(a)** w = 0.10, **(b)** w = 0.20, and **(c)** w = 0.30.

Relative deviation between experimental density data and calculated values were shown in Fig. 3. AAD % for this model was 0.0208 and justified good agreement of predicted data with experimental data.

3.1.2. Density of CO_2-loaded aqueous EAE + AEEA

Experimental data of density of CO_2-loaded aqueous EAE + AEEA were given in Table 5. Density of CO_2-loaded solution increased by increasing CO_2 loading (α) and decreased by increasing temperature. As much as CO_2 loaded in the solution, mass of solution increased and change in volume was negligible that's why density of solution increased by increasing CO_2 loading.

Density of CO_2-loaded solution was correlated by newly proposed model (eq. 8) in this work. This model worked very well in wide range of α (mol CO_2. mol amine^{-1}) and temperature in the range of 293.15 to 323.15 K.

$$\rho = \left(a + bT + cT^2\right).(1 + dw^e).(1 + f\alpha^g) \tag{8}$$

where, w was concentration of amine blend (weight fraction). a, b, c, d, e, f, and g were parameters of equation and were found out by multiple regressions using experimental data of CO_2-loaded aqueous EAE + AEEA. Values of parameters were reported as $a = -0.00481$, $b = 2.627681$, $c = 641.1562$, $d = 0.993036$, $e = 2.724007$, $f = 0.059869$, and $g = 1.243301$. Calculated values of density of CO_2-loaded solution were shown in Fig. 4. AAD % for this model was 0.1286. Relative deviation between experimental data and calculated data of density of CO_2 -loaded solutions were depicted in Fig. S2.

3.2. Viscosity

3.2.1. Viscosity of CO_2-unloaded aqueous EAE + AEEA

Viscosity data were measured for same samples that were used to measure density data. It could be shown in Table 3 and Fig. 5 that viscosity of aqueous EAE + AEEA was decreased by increasing temperature. This could be explained as increasing temperature results in increase in kinetic energy of molecules and adhesive forces between molecules also got weaker at higher temperature that's why viscosity of sample decreased at higher temperature. However, viscosity of solution increased by increasing amine concentration because of pure EAE and AEEA are more viscous than water. Increasing amine content in the solution also favors more hydrogen bonding between amine (EAE and AEEA) molecules and water molecules. Due to more hydrogen bonding, adhesive forces increased and viscosity of solution increased.

Viscosity of CO_2-unloaded aqueous EAE + AEEA blend was correlated using Pandey-Mondal model [36] (eq. 9) for aqueous ternary mixture. This model utilized viscosity of pure individual component in the mixture at same temperature to predict viscosity of aqueous ternary mixture.

$$\mu_{m,T} = exp\left[w_1 \ln\mu_{1,T} + w_2 \ln\mu_{2,T} + w_3 \ln\mu_{3,T}\right] + \sum_{i=0}^{m} c_i w^i \tag{9}$$

where, $\mu_{m,T}$, $\mu_{1,T}$, $\mu_{2,T}$, and $\mu_{3,T}$ were viscosity (mPa.s) of mixture, EAE, AEEA, and water at temperature T(K), respectively. w_1, w_2, and w_3, were weight fraction of EAE, AEEA, and water in the solution, respectively. w was concentration of solution (in weight fraction). c_i was the coefficient of equation and m was an integer for that model equation fitted well with experimental data. In this work, for m = 2 model predicted acceptable viscosity value with very minor deviation with experimental data. Model parameters c_0, c_1, and c_2 were found out by least square method and given in Table 4. ADD % for this model was 0.0051 and indicated that good agreement of model predicted data and experimental values. Comparison of experimental viscosity of aqueous EAE + AEEA blend and calculated data by eq. 9 were depicted in Fig. 5.

It was observed that viscosity of CO_2-free aqueous EAE + AEEA was non-linear function of temperature and concentration of solution. In order to avoid rigorous calculation and viscosity data requirement of pure components with coefficients of eq. 9, a simple correlation (eq. 10) was developed to predict viscosity of aqueous EAE + AEEA. That was in the form of modified Vogel-Tamman-Fulcher (VTF) type Eq. [35] and represented as follows

$$\mu = \exp\left[a + \frac{b}{(T-c)}\right].(1 + dw^e) \tag{10}$$

Where, a, b, c, d, and e were the parameters of the equation. μ denoted the viscosity (mPa.s) and w was the concentration of the amine blend (weight fraction). Parameters of the equation were found out by multiple regression and reported as; $a = -0.84012$, $b = 320.4324$, $c = 180.3574$, $d = 8.415291$, and $e = 1.325809$. AAD % was 4.433 for this correlation. Calculated viscosity data by eq. 10 were presented in Fig. S3. Goodness of model predicted data with experimental values was shown in Fig. 6 in terms of relative deviation.

3.2.2. Viscosity of CO_2-loaded aqueous EAE + AEEA

Viscosity of CO_2 loaded solution was decreased by increasing temperature and slightly increased by increasing CO_2 loading (α). Increment in viscosity may be due to formation of complex substituents by

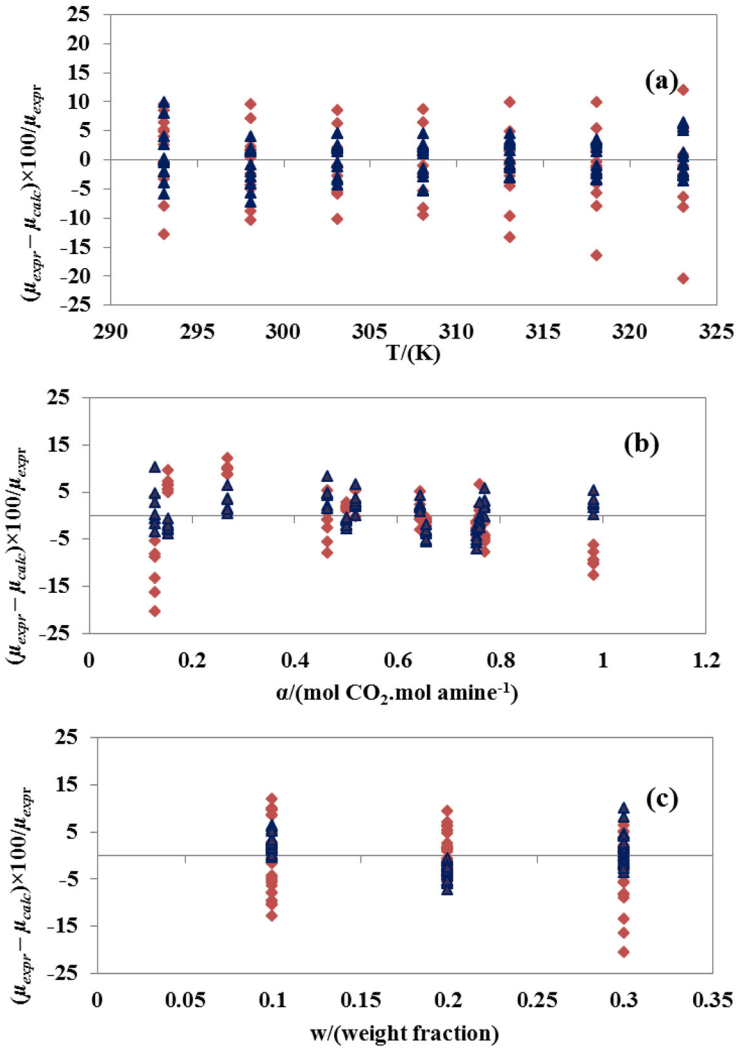

Fig. 8. Comparison of relative deviations of experimental and calculated viscosity data of CO_2-loaded aqueous EAE + AEEA blend from eq. 11 (▲) and eq. 12 (♦) as a function of **(a)** temperature, **(b)** CO_2 loading, and **(c)** concentration (EAE + AEEA weight fraction).

chemical reaction of CO_2 with EAE and AEEA in the solution. Experimental data of CO_2 loaded samples were fitted in the Wieland model [30] (eq. 11) as a function of concentration of solution, CO_2 loading, and temperature.

$$\frac{\mu}{\mu_{H_2O}} = \exp\left[\frac{[(aw+b)T+(cw+d)].[\alpha(ew+fT+g)+1].w}{T^2}\right] \quad (11)$$

Where, a, b, c, d, e, f, and g were model fitting parameters. μ, and μ_{H2O} were viscosity (mPa.s) of CO_2 loaded solution and water, respectively. T was temperature (K) and w was the concentration of the solution (weight fraction). Model parameters were obtained by multiple regressions using Excel solver and reported as; $a = -205.705$, $b = 1069.555$, $c = 6.380324$, $d = 5.336988$, $e = 2.137066$, $f = 0.004624$, and $g = -1.45083$. Agreement between calculated viscosities of CO_2 loaded aqueous EAE + AEEA by eq. 11 and experimentally obtained data were very good and shown in Fig. 7. AAD % for this model fitting was 2.852.

There was need of viscosity data of water with temperature to calculate viscosity of CO2 loaded solution with Wieland model [30]. In order to simplify the calculation, a new model (eq. 12) was developed to calculate viscosity of CO_2 -loaded aqueous EAE + AEEA. This model was also the function of temperature, concentration, and CO_2 -loading, however, there was no requirement of viscosity of water data.

$$\mu = \exp\left[a + \frac{b}{(T-c)}\right].(1+dw^e).(1+f\alpha^g) \quad (12)$$

Where, notation of eq. 12 was similar as eq.11. Model parameters of eq. 12 were found out by multiple regressions using Excel solver and given as; $a = -2.84012$, $b = 230.4324$, $c = 180.3574$, $d = 47.59638$, $e = 2.689817$, $f = 0.538985$, and $g = 0.726287$. Viscosities of CO_2 loaded aqueous EAE + AEEA were calculated by this newly proposed model and relative deviation from experimental data were measured. Comparison of relative deviation for Wieland model [30] calculated data and this newly proposed model calculated data was depicted in Fig. 8. At several points, goodness of fitting for proposed model Eq. 12 was better than Wieland model Eq. 11. However, AAD % for new proposed model was greater than the AAD % for Eq. 11 and reported as 4.744.

3.3. Diffusivity of CO_2 into aqueous EAE + AEEA blend

Viscosity data of CO_2-unloaded solution were utilized to calculate diffusivity of CO_2 into the aqueous amine mixture by using modified Stokes – Einstein Eq. [37] that could be written as follows:

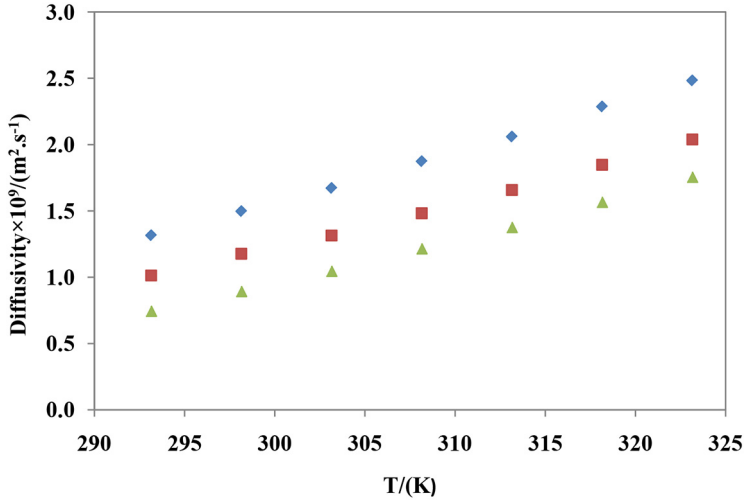

Fig. 9. Diffusivity of CO_2 into the aqueous EAE + AEEA blend versus temperature for different concentration (in weight fraction) of EAE + AEEA, w: for (♦) 0.10; (■) 0.20; and (▲) 0.30.

$$D_{CO_2-H_2O}\cdot\left(\mu_{H_2O}\right)^{0.74} = D_{CO_2-(EAE+AEEA+H_2O)}\cdot\left(\mu_{EAE+AEEA+H_2O}\right)^{0.74} \quad (13)$$

where, $D_{CO2-H2O}$ was diffusivity of CO_2 into water, μ_{H2O} was viscosity (mPa.s) of water, $\mu_{EAE+AEEA+H2O}$ was viscosity of CO_2–unloaded aqueous EAE + AEEA blend and $D_{CO2-(EAE+AEEA+H2O)}$ was diffusivity of CO_2 into the aqueous EAE + AEEA blend.

$D_{CO_2-H_2O}$ was taken from the literature [38] as a function of temperature and denoted by Eq. 14

$$D_{CO_2-H_2O} = 2.35\times10^{-6}\exp\left(\frac{-2119}{T}\right) \quad (14)$$

Calculated data of $D_{CO_2-(EAE+AEEA+H_2O)}$ were given in Table 3 and presented as a function of temperature in Fig. 9. It could be shown that diffusivity of CO_2 into the aqueous amine blend increased by increasing temperature because of viscosity of solution decreased and movement of molecules increased at higher temperature.

4. Conclusions

Density and viscosity of CO_2-unloaded and CO_2-loaded aqueous EAE + AEEA blend were measured in the temperature range of 293.15 to 323.15 K with 5 K interval at atmospheric pressure. Concentration of amine solution was 10 wt%, 20 wt%, and 30 wt% with 7/3 weight ratio of EAE/AEEA. Density of the solution was decreased by increasing temperature as well as concentration of amine. While, density of CO_2 loaded samples was increased by increasing CO_2-loading. Excess volume of mixture was calculated and reported as negative values in the experimental range of this paper. Experimental density data was correlated with newly developed model with AAD % of 0.0208 and 0.1286 for –unloaded and CO_2-loaded aqueous EAE + AEEA, respectively.

Viscosity of solutions were decreased by increasing temperature, however, increased by increasing concentration and CO_2 loading as well. New models were proposed to calculate –unloaded and CO_2 loaded viscosity for aqueous EAE + AEEA and AAD % was 4.433 and 4.744, respectively. CO_2 loaded viscosity data was also correlated with Wieland model and AAD % for this fitting was 2.852. Moreover it, diffusivity of CO_2 into the aqueous EAE + AEEA blend was calculated using modified Stokes- Einstein equation.

Credit author statement

Diwakar Pandey: Data curation, Writing- Original draft preparation, Visualization, Investigation, Validation.

Monoj Kumar Mondal: Supervision, Writing- Reviewing and Editing, Conceptualization, Methodology.

Declaration of Competing Interest

Authors have no Conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.115678.

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