Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq



Viscosity, density, and derived thermodynamic properties of aqueous 2-(ethylamino)ethanol (EAE), aqueous aminoethylethanolamine (AEEA), and its mixture for post-combustion CO₂ capture



Diwakar Pandey, Monoj Kumar Mondal*

Department of Chemical Engineering and Technology, Indian Institute of Technology, Banaras Hindu University, Varanasi 221005, Uttar Pradesh, India

ARTICLE INFO

Article history: Received 24 October 2020 Received in revised form 3 March 2021 Accepted 6 March 2021 Available online 9 March 2021

Keywords: 2-(ethylamino)ethanol Aminoethylethanolamine Excess molar volume Thermal expansion coefficient Activation molar enthalpy Activation molar entropy

ABSTRACT

In the present study, viscosity and density of aqueous 2-(ethylamino)ethanol (EAE), aqueous aminoethylethanolamine (AEEA), and aqueous EAE + AEEA blend were measured in the temperature range of 293.15 K to 333.15 K at atmospheric pressure. Concentration of aqueous amines was used upto 30 wt% that is usually applicable for CO₂ absorption from flue gases in post combustion CO₂ capture process. EAE/AEEA weight ratio was 9/1, 8/2, and 7/3 in the aqueous EAE + AEEA blend. Viscosity of all mixtures increased by increasing amine concentration and decreased by increasing temperature of samples. Experimental viscosity data were correlated to newly proposed models and average absolute deviation (AAD) % was 1.515, 2.027, and 2.889 for viscosity of EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O, respectively. Density of aqueous EAE and aqueous EAE + AEEA blend (at fixed EAE/AEEA weight ratio) decreased by increasing amount of amine in the mixture but aqueous AEEA showed increased density by increasing concentration of AEEA. Excess molar volume (V^E) calculated for all aqueous mixtures and V^E of aqueous EAE and aqueous AEEA was fitted in Redlich-Kister equation. Density of aqueous EAE + AEEA blend was correlated to a new empirical model and AAD % for this model was 0.018. Isobaric thermal expansion coefficient (α_n) of binary and ternary solutions was calculated from density versus temperature data. Activation molar enthalpy (ΔH^*), activation molar entropy (ΔS^*), and activation molar Gibbs free energy (ΔG^*) at 298.15 K for aqueous EAE, aqueous AEEA, and aqueous EAE + AEEA blend were calculated using viscosity and density data on the basis of Eyring theory of liquid viscosity.

© 2021 Elsevier B.V. All rights reserved.

1. Introduction

Carbon dioxide (CO₂) gas contributes a major fraction in greenhouse gases that are main cause of global warming. Due to environmental concern, CO₂ capture took attention of researchers in recent decades. There are several techniques to CO₂ capture i.e. adsorption technique, absorption technique, membrane separation technique, cryogenic technique etc. but absorption technique is most matured and useful of post-combustion low partial pressure CO₂ capture [1,2].

Viscosity and density data of absorbent is used in the design of absorption column and optimization of CO_2 capture process [3]. Viscosity data are required to calculate pressure drop of flow, heat transfer coefficients and mass transfer coefficients. Moreover, viscosity of absorbent is also useful to find out mass diffusivity of CO_2 in amine solvent using Stoke-Einstein eq. [4]. Densities data are used to determine the physical solubility of CO_2 in the solvent, mass transfer and solvent reaction kinetics [5]. The volumetric and thermodynamic properties of solutions

* Corresponding author. E-mail address: mkmondal13@yahoo.com (M.K. Mondal). are used to study the molecular interactions of various components of the mixture [6].

2-(Ethylamino)ethanol (EAE) is a hindered secondary amine and has higher CO_2 loading with lower heat of absorption because it produces unstable carbamate [7,8]. Aminoethylethanolamine (AEEA) is an alkanoldiamine and has been shown high CO_2 loading (mol CO_2 /mol amine), faster reaction kinetics but its heat of absorption is high [9,10]. In the literature AEEA has been used as an activator in the amine blends to improve solvent performance for CO_2 capture [11–15].

Physicochemical properties data of various aqueous alkanolamines mixture has been published in the literature [5,11,16–21]. Gao et al. [19] measured experimental data of viscosity, density and refractive index of aqueous EAE but they had not presented its activation thermodynamic properties for viscous flow. Viscosity and density data of aqueous AEEA has been published in literature [20,21] but there are very less data points in the range of upto 30 wt% (0.0689 mol fraction of amine in aqueous mixture) of concentration of amine. That's why those data are less useful for calculations in $\rm CO_2$ capture process where concentration of amines used upto 30 wt% generally. Moreover, there is lack of viscosity and density data of aqueous blend of EAE + AEEA in the literature.

In this paper viscosity and density of aqueous EAE, aqueous EAE + AEEA, and density of aqueous AEEA were measured in the temperature range of 293.15 K to 333.15 K with intervals of 5 K and at atmospheric pressure. Experimental viscosity data of aqueous AEEA was taken from our published paper [22]. Concentration of EAE and AEEA in the binary aqueous mixture varied from 5 wt% to 30 wt% with intervals of 5 wt%. In the ternary mixture of EAE + AEEA+H2O concentration of amines (EAE + AEEA) was 10 wt%, 20 wt% and 30 wt%. Weight ratio of EAE/AEEA was 9/1, 8/2, and 7/3 so weight fraction of AEEA in the amine (EAE + AEEA) was 0.10, 0.20, and 0.30. Experimental viscosity data was correlated to newly proposed model. Density data of binary mixture correlated with Redlich-Kister model [23] and density data of ternary mixture fitted in new model. Isobaric thermal expansion coefficient was calculated from density versus temperature data. Using viscosity and density data, thermodynamic properties were measured for activation of viscous flow.

2. Experimental section

2.1. Chemicals

The EAE (98% purity) was purchased from Sigma Aldrich, St. Louice USA. AEEA (98% purity) was purchased from Sd Fine chemical limited, Mumbai, India. All chemicals were used without further purification. Description of all chemicals which were used in the experimentation was listed in Table 1. Distilled water was used for making aqueous mixture of EAE, AEEA, and EAE + AEEA.

2.2. Solution preparation method

Solution was prepared by mixing alkanolamines with double-distilled water in the appropriate amount to obtain the desired concentration. Density data was used to convert volume into mass. Density was measured using density meter (Anton Paar density meter, Model: DMATM 35). Volume of components of binary and tertiary mixture was measured carefully with micro-pipette.

2.3. Viscosity measurement

Viscosity of samples was measured by using Sine-wave Vibro viscometer of A & D Company (model SV-10, with accuracy of 1% repeatability and uncertainty of $\pm 3\%$) in the temperature range 293.15 K to 333.15 K and at atmospheric pressure. Temperature of samples was maintained constant using a temperature controlled water bath. Error in temperature measurement was ± 1 K. The viscometer was cleaned using methanol and calibrated with double distilled water (of known viscosity) before each sampling. 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. Viscosity measurement by this viscometer did not require any equation for calculation. The process of viscosity measuring of a sample was repeated three times and average value was reported.

Table 1Details of used chemicals in this study.

Chemical Name CAS number Purification method Source Initial purity 2-(ethylamino)ethanol (EAE) 110-73-6 Sigma Aldrich, St. Louice, USA ≥ 98% none 98%^c Aminoehtylethanolamine (AEEA) sd Fine chemical limited, Mumbai, India 111-41-1 none 99% sd Fine chemical limited, Mumbai, India Acetone 67-64-1 none Methanol 67-56-1 sd Fine chemical limited, Mumbai, India 99%c none Watera 7732-18-5 Sigma Aldrich, St. Louice, USA 99.99% none Waterb 7732-18-5 Our laboratory 99.99% Double distillation

2.4. Density measurement

The density of samples was measured using Anton Paar density meter (Model DMA™ 35) in the temperature range 293.15 K to 333.15 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 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.

3. Results and discussions

3.1. Validation of used apparatus and experimental procedure

In order to ensure the validation of experimental apparatus and experimental procedure, experimental data of density and viscosity were measured and compared with available literature data [5,11,17,19–21,24,25]. Comparison of experimental viscosity and density data with literature data was given in Table 2 for pure EAE and pure AEEA. Viscosity of aqueous EAE and density of aqueous AEEA were compared in Fig. S1 and Fig. S2, respectively, of the supporting information file. 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.

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

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

3.2. Viscosity

3.2.1. Viscosity of aqueous EAE and aqueous AEEA

Viscosity of aqueous EAE and aqueous AEEA was measured at 293.15 K, 298.15 K, 303.15 K, 308.15 K, 313.15 K, 318.15 K, 323.15 K, 328.15 K, and 333.15 K. Concentrations of samples were 5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt%, and 30 wt% so that weight fraction of amine (w_I) in the aqueous mixture was 0.05,0.10,0.15, 0.20, 0.25, and 0.30. All the measurement was done at atmospheric pressure. Experimental data of viscosity was given in Table 3. Viscosity of the aqueous EAE and aqueous AEEA as well was decreased by increasing temperature of samples. Molecules of amines and water are bonded by intermolecular forces and at

^aDe-ionized water for density-meter calibration, ^bdouble-distilled water for making aqueous solution, and ^cmass percent.

Table 2Experimental 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)	$\rho/(kg.m^{-3})$				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
318.15 323.15	897.5 892.8	896.8 ^c 893.68 ^b 892.6 ^c	1009.7 1005.4	1006.11 ^d	5.32 4.28	5.358 ^c 4.527 ^b 4.517 ^c	28.23 21.55	23.4 ^e

astandard 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 $U(\rho) = 0.003 \, \rho$ (kg.m⁻³) and $U(\mu) = 0.08 \, \mu$ (mPa.s). aReference [24], bReference [25], cReference [17], dReference [5], cReference [20], fReference [21], gReference [11].

In the literature [26,27], a logarithmic function model as shown in eq. 2, was used to correlate the viscosity data.

$$ln \mu = A + \frac{B}{T}$$
(2)

Where, A and B are fitting parameter, T in K and μ is the viscosity (mPa.s). Same model was used to correlate viscosity of aqueous EAE and aqueous AEEA for different concentration of amines in the range of 5 wt% to 30 wt%. Model parameters were calculated using experimental data of viscosity by Excel solver and listed in Table 4.

Based on the Arrhenius equation type model [22], a new model was developed for aqueous mixture of amines. In this model, viscosity was described as a function of temperature and concentration of the aqueous amines. Experimental data of viscosity of binary mixture of EAE + H₂O and AEEA + H₂O were correlated with a newly proposed viscosity model presented by eq. 3.

$$\mu = exp \left[w_1 \left(A + \frac{B}{T} \right) + w_2 \left(C + \frac{D}{T - E} \right) \right] \tag{3}$$

Where, A, B, C, D, and E were the parameters of the equation. μ denoted the viscosity (mPa.s) and w_1 was the concentration of the amine (in weight fraction). T was the temperature of samples in K.

Parameters of model were found out by multiple regressions using Excel solver. For aqueous EAE, values of parameters were given as A=-15.9327, B=6054.772, C=-1.4736, D=81.30073, and E=241.3657. Viscosity of aqueous AEEA were fitted for A=-22.1056, B=8103.578, C=-4.70281, D=1230.874, and E=22.13974. AAD %

was 1.515 and 2.027 for aqueous EAE and aqueous AEEA, respectively, which justified the good agreement of experimental data and model predicted data. Comparison between experimental data of viscosity and calculated data by eq. 3 was depicted in Fig. 1 and Fig. 2 for aqueous EAE and aqueous AEEA, respectively.

Performance of this newly proposed model (eq. 3) was compared with the model used in literature (eq. 2) by calculating standard deviation between experimental data and model predicted data. From Table 5, it can be shown that standard deviation for both the models are very less. So it can be concluded that both models are accurate and acceptable but newly proposed model eq. 3 is better because it can predict viscosity at any point within concentration of 5 wt% to 30 wt% and temperature of 293.15 K to 333.15 K using same reported values of model parameter in this work. While, model eq. 2 could predict the data at particular concentration points (5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt%, and 30 wt%) using Table 4. Calculation of viscosity at the other concentration points would require new value of fitting parameters of eq. 2 on that certain concentration.

3.2.2. Viscosity of EAE + AEEA + H₂O blend

Viscosity of aqueous EAE + AEEA blend was measured at 293.15 K, 298.15 K, 303.15 K, 308.15 K, 313.15 K, 318.15 K, 323.15 K, 328.15 K, 333.15 K, and at atmospheric pressure. Total concentration of amines was 10 wt%, 20 wt% and 30 wt%. Weight ratio (w_1/w_2) of EAE/AEEA was 9/1, 8/2, and 7/3. Composition in weight fraction $(w_1/w_2/w_3)$ of components (EAE/AEEA/H₂O) of samples could be written as (0.09/ 0.01/0.90), (0.08/0.02/0.90), (0.07/0.03/0.90), (0.18/0.02/0.80), (0.16/0.02/0.80), (0.08/0.02/0.90)0.04/0.80), (0.14/0.06/0.80), (0.27/0.03/0.70), (0.24/0.06/0.70), and (0.21/0.09/0.70). Experimental data of viscosity of EAE + AEEA + H₂O blend was given in Table 6. It could be shown that viscosity of all samples was decreased by increasing temperature. This behavior of viscosity could be explained by same reason as discussed for aqueous EAE and AEEA. Increasing total concentration of amines in the mixture for fixed EAE/AEEA weight ratio resulted in signified increase in viscosity of EAE + AEEA + H₂O blend. However, decreasing EAE/AEEA weight ratio for constant total concentration resulted slightly increase in viscosity. This revealed that varying weight ratio of EAE/AEEA did not affect in intermolecular bonding of EAE, AEEA, and H₂O molecules in significant amount. But, increasing total concentration of EAE + AEEA in the mixture increased intermolecular attraction of molecules.

Viscosity of aqueous EAE + AEEA blend ternary system was correlated by newly proposed model eq. 4. That was basically extended form of eq. 3 for ternary system.

Table 3 Viscosity (μ), density (ρ), and excess molar volume (V^E), of aqueous EAE and aqueous AEEA at T=(293.15-333.15) K and 101.325 kPa pressure ^a.

EAE + I	H ₂ O			AEEA +	H ₂ O		
T/(K)	μ/	ρ/(kg.	$V^E \times 10^6$ /	T/(K)	μ/	ρ/(kg.	$V^{E} \times 10^{6}$ /
, ,	(mPa.s)	m^{-3})	$(m^3.mol^{-1})$, , ,	(mPa.s)	m^{-3})	$(m^3.mol^{-1})$
$w_1 = 0.$				$w_1 = 0.$			
293.15	1.34	998.9	-0.0980	293.15	1.20	1001.2	-0.0298
298.15	1.17	997.5	-0.0934	298.15	1.11	999.8	-0.0249
303.15 308.15	1.04 0.94	996.0 994.1	-0.0924 -0.0897	303.15 308.15	0.95 0.85	998.3 996.2	-0.0233 -0.0159
313.15	0.85	991.9	-0.0837 -0.0871	313.15	0.85	994.0	-0.0139 -0.0122
318.15	0.78	989.7	-0.0865	318.15	0.69	991.7	-0.0086
323.15	0.71	987.2	-0.0841	323.15	0.64	989.2	-0.0048
328.15	0.65	984.6	-0.0837	328.15	0.59	986.6	-0.0028
333.15	0.60	981.8	-0.0832	333.15	0.55	983.9	-0.0027
$w_1 = 0.$				$w_1 = 0.$			
293.15	1.72	998.2	-0.1751	293.15	1.60	1004.6	-0.0678
298.15 303.15	1.48 1.31	996.8 995.2	-0.1732 -0.1732	298.15 303.15	1.36 1.18	1003.2 1001.6	-0.0654 -0.0642
308.15	1.17	993.2	-0.1732 -0.1713	308.15	1.06	999.4	-0.0567
313.15	1.04	990.8	-0.1678	313.15	0.93	997.1	-0.0531
318.15	0.94	988.4	-0.1666	318.15	0.84	994.7	-0.0496
323.15	0.85	985.8	-0.1654	323.15	0.77	992.1	-0.0459
328.15	0.78	983.0	-0.1644	328.15	0.72	989.4	-0.0440
333.15	0.72	980.0	-0.1635	333.15	0.64	986.5	-0.0420
$w_1 = 0.$		007.4	0.0554	$w_1 = 0.$		10000	0.1150
293.15	2.20	997.4	-0.2571	293.15	1.92	1008.3	-0.1150
298.15 303.15	1.82 1.59	995.8 993.9	-0.2541 -0.2509	298.15 303.15	1.74 1.48	1006.8 1005.1	-0.1132 -0.1125
308.15	1.40	991.8	-0.2309 -0.2499	308.15	1.32	1003.1	-0.1123 -0.1030
313.15	1.25	989.3	-0.2474	313.15	1.19	1002.7	-0.0994
318.15	1.12	986.7	-0.2454	318.15	1.06	997.8	-0.0962
323.15	1.01	983.9	-0.2435	323.15	0.92	995.1	-0.0926
328.15	0.93	980.8	-0.2396	328.15	0.82	992.3	-0.0909
333.15	0.86	977.6	-0.2380	333.15	0.74	989.3	-0.0888
$w_1 = 0.$				$w_1 = 0.$			0.4=40
293.15	2.82	996.3	-0.3403	293.15	2.63	1012.4	-0.1746
298.15 303.15	2.34 2.02	994.6 992.5	-0.3382 -0.3336	298.15 303.15	2.27 1.94	1010.7 1008.8	-0.1717 -0.1693
308.15	1.76	990.2	-0.3330 -0.3315	308.15	1.66	1006.4	-0.1620
313.15	1.55	987.6	-0.3301	313.15	1.44	1003.8	-0.1564
318.15	1.36	984.8	-0.3271	318.15	1.28	1001.2	-0.1534
323.15	1.19	981.7	-0.3221	323.15	1.15	998.4	-0.1501
328.15	1.10	978.5	-0.3195	328.15	1.04	995.3	-0.1442
333.15	1.01	975.1	-0.3170	333.15	0.93	992.1	-0.1400
$w_1 = 0.$		005.3	0.4210	$w_1 = 0.$		10166	0.2420
293.15 298.15	3.54 2.96	995.2 992.6	-0.4319 -0.4124	293.15 298.15	3.47 2.98	1016.6 1014.8	-0.2420 -0.2399
303.15	2.48	990.4	-0.4124 -0.4085	303.15	2.37	1014.6	-0.2333 -0.2337
308.15	2.08	988.0	-0.4072	308.15	2.05	1010.0	-0.2242
313.15	1.82	985.3	-0.4069	313.15	1.74	1007.3	-0.2187
318.15	1.60	982.4	-0.4052	318.15	1.53	1004.4	-0.2116
323.15	1.40	979.3	-0.4036	323.15	1.31	1001.4	-0.2062
328.15	1.26	976.0	-0.4023	328.15	1.19	998.2	-0.2003
333.15	1.16	972.5	-0.4013	333.15	1.02	994.7	-0.1916
$w_1 = 0.$		002.0	0.5127	$w_1 = 0.$		1020.0	0.2102
293.15 298.15	4.38 3.56	993.8 991.1	-0.5137 -0.4942	293.15 298.15	4.73 3.95	1020.9 1018.6	-0.3183 -0.3080
303.15	3.02	988.6	-0.4942 -0.4862	303.15	3.32	1016.3	-0.3080 -0.3021
308.15	2.48	985.8	-0.4802 -0.4785	308.15	2.61	1010.5	-0.3021 -0.2950
313.15	2.12	982.5	-0.4671	313.15	2.19	1010.8	-0.2874
318.15	1.88	979.4	-0.4640	318.15	1.86	1007.4	-0.2710
323.15	1.65	976.0	-0.4587	323.15	1.59	1004.3	-0.2658
328.15	1.44	972.6	-0.4586	328.15	1.35	1000.9	-0.2575
333.15	1.33	968.9	-0.4563	333.15	1.16	997.3	-0.2485
a Standard	uncertain	ties 11 are	$\mu(T) = 1 K \text{ for}$	viscosity	mascurom	ont u(T)	— 0.2 K for den-

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

$$\mu = exp\left[w_1\left(A + \frac{B}{T}\right) + w_2\left(C + \frac{D}{T}\right) + w_3\left(E + \frac{F}{T - G}\right)\right] \tag{4}$$

Table 4 Fitting parameters of eq. 2 in the range of T = (293.15-333.15) K and for different weight fraction of amine (w_1) .

W ₁	$EAE + H_2O$		AEEA+H ₂ O	
	A	В	A	В
0.05	-6.329	1934.376	-6.526	1965.076
0.10	-6.710	2118.358	-6.991	2175.676
0.15	-6.960	2256.212	-7.373	2357.817
0.20	-7.569	2511.300	-7.743	2548.337
0.25	-8.171	2755.063	-8.935	2979.519
0.30	-8.564	2930.718	-10.289	3473.629

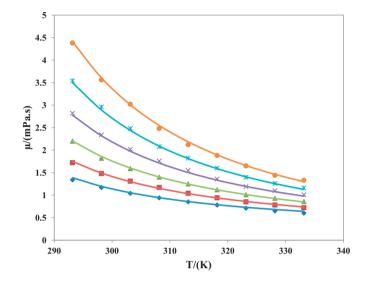


Fig. 1. Viscosity versus temperature of EAE + H₂O for different concentration of EAE in terms of weight fraction (w₁) of EAE, w₁: \spadesuit 0.05; \blacksquare 0.10; \blacktriangle 0.15; \times 0.20; * 0.25; \bullet 0.30; - calculated values with eq. 3.

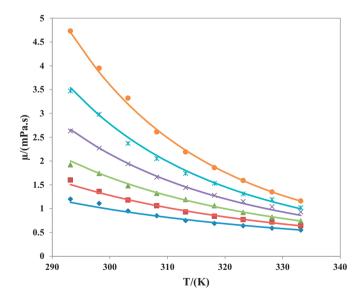


Fig. 2. Viscosity versus temperature of AEEA + H_2O for different concentration of AEEA in terms of weight fraction (w_1) of AEEA, w_1 : ♦ 0.05; ■ 0.10; ▲ 0.15; × 0.20; * 0.25; • 0.30; — calculated values with eq. 3.

Where, A, B, C, D, E, F, and G were model fitting parameters. μ was the viscosity (mPa.s) and T was the temperature of samples in K. w_1 , w_2 , and w_3 were weight fraction of EAE, AEEA, and H_2O , respectively.

Table 5 Standard deviation^a (s.d.) (mPa.s) for experimental data of viscosity of binary mixture and model predicted viscosity data in the range of T=(293.15-333.15) K and for different weight fraction of amine (w_1) .

W_1	$EAE + H_2O$		AEEA+H ₂ O	AEEA+H ₂ O		
	s.d. for model eq. 2	s.d for model eq. 3	s.d for model eq. 2	s.d for model eq. 3		
0.05	0.0139	0.0394	0.0220	0.0581		
0.10	0.0202	0.0225	0.0299	0.0522		
0.15	0.0497	0.0223	0.0227	0.0477		
0.20	0.0493	0.0517	0.0330	0.0571		
0.25	0.0685	0.0478	0.0571	0.0885		
0.30	0.0685	0.0361	0.0529	0.0827		

astandard deviation = $\left[\sum_{i=1}^{n} \frac{(\mu_{\text{operimental}} - \mu_{\text{colculated}})}{n-p}\right]^{0.5}$

where n is the number of experimental points and p is the number of adjustable fitting coefficients.

Value of parameters was calculated by multiple regression using Excel solver and reported as, A=-13.4924, B=4878.7, C=2.184672, D=2.184672, E=-1.43594, E=65.91624, and E=247.4278. Using these values of parameters, viscosity value was predicted at different temperature and composition of blend within the experimental operating conditions of this study. Calculated viscosity data of EAE + AEEA + H₂O blend was shown in Fig. 3. AAD % for this model was 2.889 and low value of AAD indicated that this newly proposed model was accepted for prediction of viscosity of ternary aqueous system.

3.3. Density

3.3.1. Density of aqueous EAE and aqueous AEEA

 increasing temperature of sample. Density of aqueous EAE was decreased and density of aqueous AEEA was increased by increasing amine concentration. That was because of density of pure EAE was less than density of water and density of AEEA was more than density of water at any temperature. Density was decreased by increasing temperature because of increase of volume of sample with constant mass.

In the literature [28], a correlation (eq. 5) was used to calculate the molar volume of the aqueous alkanolamines solution.

$$V_m = \frac{\sum_{i=1}^{n} (x_i M_i)}{\rho_m} \tag{5}$$

Where, ρ_m , V_m , x_i , and M_i were density of the mixture (kg.m⁻³), molar volume of the mixture (m³.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) was calculated by eq. 6.

$$V^{E} = V_{m} - \sum_{i=1}^{n} (x_{i}V_{i})$$
 (6)

Where, V_i was the molar volume (m³.mol⁻¹) of component i.

Excess volume of aqueous EAE + H_2O and AEEA + H_2O blend was calculated by eq. 7 and listed in Table 3. For binary solutions, V^E decreases by increasing temperature. This phenomenon may be due to decrease in self-association and cross association of hydrogen bonds with increasing temperature of binary solutions.

 $\label{eq:table 6} \textbf{Table 6} \\ \textbf{Viscosity } (\mu), \text{ density } (\rho), \text{ and excess molar volume } (\textbf{V}^E), \text{ of EAE} + \text{AEEA} + \text{H}_2\text{O} \text{ blend at } T = (293.15-333.15) \text{ K and } 101.325 \text{ kPa pressure }^a.$

T/(K)	μ/(mPa.s)	$\rho/(\text{kg.m}^{-3})$	$V^E\times 10^6/(m^3.mol^{-1})$	μ/(mPa.s)	$\rho/(kg.m^{-3})$	$V^E\times 10^6/(m^3.mol^{-1})$	μ/(mPa.s)	$\rho/(\text{kg.m}^{-3})$	$V^E \times 10^6 / (m^3.mol^{-1})$
$w_1(EAE)/w_2(AEEA) = 9/1$									
$w_1 + w_2 = 0.10$				$w_1 + w_2 =$	= 0.20		$w_1 + w_2 =$	= 0.30	
293.15	1.36	996.9	-0.1261	1.85	995.3	-0.2675	2.64	993.3	-0.4291
298.15	1.19	995.8	-0.1300	1.58	994.3	-0.2803	2.21	992	-0.4430
303.15	1.05	994.4	-0.1337	1.43	992.8	-0.2884	1.89	990.5	-0.4588
308.15	0.94	992.7	-0.1375	1.30	990.9	-0.2945	1.67	988.9	-0.4797
313.15	0.84	991.0	-0.1476	1.18	988.8	-0.3036	1.51	986.8	-0.4971
318.15	0.78	989.1	-0.1561	1.08	986.7	-0.3155	1.38	984.7	-0.5181
323.15	0.71	987.0	-0.1648	0.93	984.5	-0.3300	1.27	982.4	-0.5396
328.15	0.64	985.2	-0.1837	0.85	982.0	-0.3425	1.17	979.8	-0.5591
333.15	0.57	983.0	-0.1988	0.77	979.6	-0.3621	1.07	977.3	-0.5866
$w_1(EAE)/w_2(AEEA) = 8/2$									
$w_1 + w_2 = 0.10$				$w_1 + w_2 =$	= 0.20		$w_1 + w_2 =$	= 0.30	
293.15	1.39	997.2	-0.1088	1.92	995.9	-0.2298	2.85	994.1	-0.3644
298.15	1.22	996.2	-0.2196	1.64	995.0	-0.2446	2.35	992.9	-0.3804
303.15	1.08	994.8	-0.2293	1.48	993.4	-0.2502	1.98	991.4	-0.3957
308.15	0.97	993.2	-0.2413	1.34	991.4	-0.2538	1.73	989.5	-0.4087
313.15	0.88	991.6	-0.2602	1.21	989.3	-0.2623	1.56	987.6	-0.4300
318.15	0.82	989.5	-0.2712	1.11	987.2	-0.2737	1.41	985.2	-0.4428
323.15	0.76	987.7	-0.2934	0.99	985.0	-0.2875	1.29	983	-0.4657
328.15	0.68	985.8	-0.3180	0.90	982.8	-0.3060	1.19	980.6	-0.4889
333.15	0.60	983.6	-0.3408	0.81	980.5	-0.3270	1.09	978.1	-0.5151
$w_1(EAE)/w_2(AEEA) = 7/3$									
$w_1 + w_2 = 0.10$				$w_1 + w_2 =$	= 0.20		$w_1 + w_2 =$	= 0.30	
293.15	1.42	997.8	-0.0986	2.02	996.4	-0.4935	3.06	995.2	-0.3067
298.15	1.25	996.7	-0.1023	1.73	995.6	-0.5113	2.51	994	-0.3225
303.15	1.12	995.3	-0.1058	1.55	994.1	-0.5200	2.11	992.6	-0.3397
308.15	1.02	993.6	-0.1092	1.4	992.2	-0.5264	1.83	990.8	-0.3545
313.15	0.94	991.9	-0.1189	1.26	990.3	-0.5400	1.62	988.9	-0.3750
318.15	0.87	990.1	-0.1289	1.16	988.5	-0.5587	1.45	986.8	-0.3942
323.15	0.82	988.2	-0.1410	1.07	986.6	-0.5755	1.31	984.5	-0.4136
328.15	0.73	986.2	-0.1553	0.97	984.0	-0.5903	1.22	981.7	-0.4260
333.15	0.65	984.1	-0.1718	0.86	982.6	-0.6076	1.11	979.6	-0.4608

^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_1) = 0.01, u(w_2) = 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).

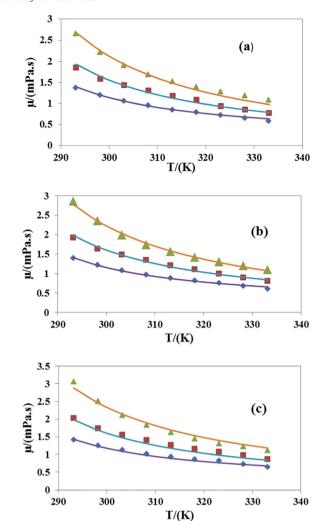


Fig. 3. Viscosity versus temperature of EAE + AEEA + H $_2$ O for concentration in weight fraction (w_1+w_2) of EAE + AEEA, (w_1+w_2) : \spadesuit 0.10; \blacksquare 0.20; \triangleq 0.30; - calculated values with eq. 4 with different weight ratio of EAE/AEEA (w_1/w_2) for **(a)** $(w_1/w_2) = 9/1$, **(b)** $(w_1/w_2) = 8/2$, and **(c)** $(w_1/w_2) = 7/3$.

Table 7Coefficients of Redlich-Kister equation (eq. 8) for calculation of density of aqueous EAE and aqueous AEEA.

T/(K)	A_0	A_1	A_2	A_3	s.d. ^a				
$EAE + H_2$	$EAE + H_2O$								
293.15	1802	6096.9	6850.1	2566.2	0.0035				
298.15	878.66	2991.4	3375.6	1272.9	0.0063				
303.15	804.91	2744.6	3101.5	1171.7	0.0034				
308.15	637.74	2162.2	2427.2	912.12	0.0021				
313.15	859.68	2864.7	3165.1	1169.2	0.0017				
318.15	914.85	3049.3	3370.7	1245.3	0.0017				
323.15	668.69	2218.8	2440.2	897.75	0.0028				
328.15	778.24	2593.2	2863.2	1056.9	0.0050				
333.15	835.15	2783.6	3075.2	1135.4	0.0057				
AEEA + H	I_2O								
293.15	294.28	959.59	1011.8	349.57	0.0345				
298.15	-539.32	-1837.5	-2112.7	-812.52	0.0024				
303.15	-1112.4	-3726.8	-4186.3	-1570.2	0.0052				
308.15	-2005.7	-6695.4	-7473.9	-2783.9	0.0032				
313.15	-2539.4	8471.6	-9442.7	-3511	0.0074				
318.15	-2828.7	-9478.6	-10,606	-3956.8	0.0214				
323.15	-3392.7	-11,355	-12,686	-4725.2	0.0200				
328.15	-3879.4	-12,960	-14,448	-5369.2	0.0254				
333.15	-3889.6	-12,988	-14,471	-5374.5	0.0186				

astandard deviation = $\left[\sum_{i=1}^{n} \frac{\left(V^{E}_{experimental_{i}} - V^{E}_{exhalusted_{i}}\right)^{2}}{n-p}\right]^{0.5}$, where n is the number of experimental points and p is the number of adjustable fitting coefficients.

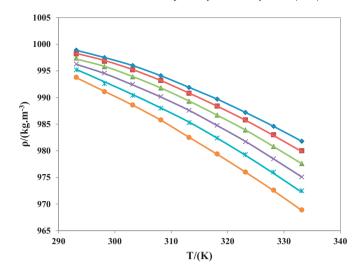


Fig. 4. Density versus temperature of EAE + H₂O for different concentration of EAE in terms of weight fraction (w₁) of EAE, w₁: \spadesuit 0.05; \blacksquare 0.10; \triangle 0.15; \times 0.20; * 0.25; \bullet 0.30; - calculated values with eq. 9.

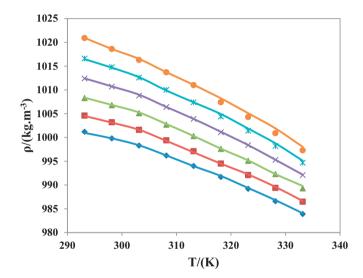


Fig. 5. Density versus temperature of AEEA + H₂O for different concentration of AEEA in terms of weight fraction (w_1) of AEEA, w_1 : \spadesuit 0.05; \blacksquare 0.10; \triangle 0.15; \times 0.20; * 0.25; \bullet 0.30; - calculated values with eq. 9.

 $\label{eq:Table 8} \textbf{Regressed coefficients of eq. 11 to calculate density of EAE} + \text{AEEA} + \text{H}_2\text{O}.$

T/(K)	C ₀	c ₁	c_2	c ₃	s.d.a
293.15	-2.4018	126.31	-500.76	1056.3	0.547
298.15	-3.5201	150.77	-611.48	1210.3	0.482
303.15	-2.5396	130.34	-467.62	933.06	0.403
308.15	-1.9201	121.17	-4.2206	894.56	0.357
313.15	0.0129	89.255	-226.1	535.23	0.248
318.15	-0.7023	111.19	-354.95	777.97	0.244
323.15	-0.1061	109.41	-340.9	753.96	0.254
328.15	1.1238	101.26	-320.96	760.42	0.120
333.15	-0.3296	145.19	-561.01	1164	0.339

 $\label{eq:astandard} \begin{array}{l} ^{a}standard \ deviation = \left[\sum\limits_{i=1}^{n}\frac{\left(\rho_{experimental,i} - \rho_{calculated,i}\right)}{n-p}\right]^{0.5} \end{array}$

where n is the number of experimental points and p is the number of adjustable fitting coefficients.

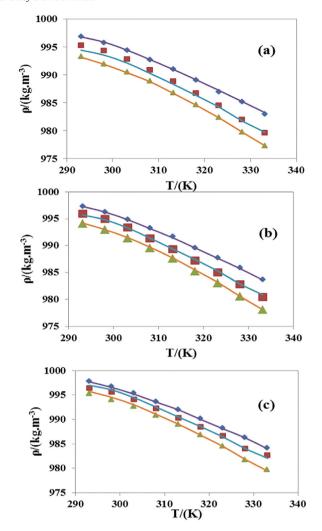


Fig. 6. Density versus temperature of EAE + AEEA + H_2O for concentration in weight fraction $(w_1 + w_2)$ of EAE + AEEA, $(w_1 + w_2)$: • 0.10; • 0.20; • 0.30; - calculated values with eq. 11 with different weight ratio of EAE/AEEA (w_1/w_2) for (a) $(w_1/w_2) = 9/1$, (b) $(w_1/w_2) = 8/2$, and (c) $(w_1/w_2) = 7/3$.

$$V^{E} = \left[\frac{(x_{1}M_{1} + x_{2}M_{2})}{\rho_{m}} \right] - \left[\frac{x_{1}M_{1}}{\rho_{1}} + \frac{x_{2}M_{2}}{\rho_{2}} \right]$$
 (7)

Where, x_1 , and x_2 were mole fraction of EAE or AEEA, and water, respectively. M_1 , and M_2 were molecular weight (kg.mol⁻¹) of EAE or AEEA, and water, respectively. ρ_1 , and ρ_2 were density (kg.m⁻³) of EAE or AEEA, and water, respectively.

Density of binary system were calculated by using the Redlich-Kister [23] equation by some researchers [5,17,19,25] in the literature. In this study, excess volume of binary mixture was fitted to following Redlich-Kister model (eq. 8).

$$V^{E} = 10^{-6} x_{1} x_{2} \sum_{i=0}^{m} A_{i} (x_{1} - x_{2})^{i}$$
 (8)

Where, A_i were coefficients of Redlich-Kister equation. x_1 and x_2 were mole fraction of amine and water, respectively. m was the positive integer for that excess volume could be well fitted by the measured experimental data. Values of A_i were calculated by using least-squares fitting. In this work third order polynomial was fitted very well to excess volume data. Regressed coefficients A_0 , A_1 , A_2 and A_3 were given in

Table 7. Density of aqueous EAE and aqueous AEEA was calculated by eq. 9 and shown by Fig. 4 and Fig. 5, respectively.

$$\rho_m = \left[\frac{(x_1 M_1 + x_2 M_2)}{\left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2}\right) + 10^{-6} x_1 x_2 \sum_{i=0}^3 A_i (x_1 - x_2)^i} \right]$$
(9)

Where, symbols of equation were same as in eq. 5, eq. 7 and eq. 8.

3.3.2. Density of EAE + AEEA + H₂O blend

Density of aqueous EAE + AEEA blend was measured in the same compositions and operating conditions as viscosity was discussed in section 3.2.2 of this paper. Density of all samples was decreased by increasing temperature of samples. Effect of increase in total concentration at constant weight ratio of EAE/AEEA resulted in decrease in density. Varying EAE/AEEA ratio from 9/1 to 7/3 at constant total concentration not affected very much on density of mixture. However, density was slightly increased by increasing amount of AEEA in the mixture. Because of, density of pure AEEA was much more than density of pure EAE.

Excess volume of aqueous EAE + AEEA blend was calculated by eq. 10 and listed in Table 6.

$$V^{E} = \left[\frac{(x_{1}M_{1} + x_{2}M_{2} + x_{3}M_{3})}{\rho_{m}} \right] - \left[\frac{x_{1}M_{1}}{\rho_{1}} + \frac{x_{2}M_{2}}{\rho_{2}} + \frac{x_{3}M_{3}}{\rho_{3}} \right] \tag{10}$$

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 densities (kg.m⁻³) of EAE, AEEA, and water, respectively. Excess volume over all temperature and concentration was negative. It indicated that formation of hydrogen bond and contracting behavior of aqueous EAE + AEEA solution.

In the EAE + AEEA + H₂O system, four different important molecular effects can be considered as, (i) intermolecular dipolar interactions, (ii) geometrical fitting between components, (iii) hydrogen bond rupture, and (iv) dispersive interaction between unlike molecules. The negative contribution of V^E may be due to (i) and (ii) effects while positive contribution of V^E may be due to hydrogen bonds breaking and interaction between molecules of different compounds. Generally, V^E would be decreased by increasing temperature because hydrogen bonds of solution molecules break by the applied energy through solution heating. However, V^E increased by increasing temperature for the ternary solution of EAE + AEEA + H₂O in the studied concentrations range of this work. This may be due to more contributions of (i) and (ii) effects among molecules of EAE, AEEA, and H₂O in comparison to (iii) and (iv) effects, in the studied concentrations and temperature range of EAE + AEEA + H₂O in this paper.

A new correlation was proposed to calculate the density of EAE + AEEA + H₂O blend. This model used density of pure components of mixture to calculate density of mixture at any particular temperature in the range of 293.15 K to 333.15 K.

$$\rho_m = w_1 \rho_1 + w_2 \rho_2 + w_3 \rho_3 + \sum_{i=0}^{i=m} c_i w_1^i$$
(11)

Where, w_1 , w_2 , and w_3 were weight fraction of EAE, AEEA, and H₂O, respectively. ρ_m , ρ_1 , ρ_2 , and ρ_3 were densities (kg.m⁻³) of mixture, EAE, AEEA, and water, respectively. c_i was the Redlich-Kister type coefficient [23] and called mass interaction factor by Pandey-Mondal [22] in the literature. m was an integer number that could be well fitted with the experimental data.

Values of c_i were calculated by least square fitting. In this present work eq. 11 fitted very well for m = 3 to density data of aqueous

Table 9 The values of isobaric thermal expansion coefficient for EAE + H₂O, AEEA+H₂O, and EAE + AEEA + H₂O at T = (293.15–333.15) K and 101.325 kPa pressure.

W_1	$\alpha_p \times 10^4/(\text{K}^{-1})$	$\underline{\alpha_{\rm p}} \times 10^4/({\rm K}^{-1})$								
	T/(K) = 293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	
EAE + H ₂ O										
0.05	2.64	3.06	3.48	3.90	4.32	4.74	5.16	5.58	5.99	
0.10	2.80	3.26	3.72	4.19	4.65	5.11	5.57	6.03	6.49	
0.15	3.12	3.60	4.08	4.56	5.04	5.52	6.00	6.48	6.96	
0.20	3.44	3.94	4.43	4.93	5.43	5.92	6.42	6.91	7.41	
0.25	4.17	4.56	4.95	5.34	5.73	6.12	6.51	6.90	7.29	
0.30	4.97	5.31	5.66	6.00	6.35	6.69	7.04	7.38	7.73	
$AEEA+H_2O$										
0.05	2.85	3.23	3.62	4.01	4.40	4.78	5.17	5.56	5.94	
0.10	2.94	3.35	3.76	4.17	4.58	4.99	5.40	5.82	6.23	
0.15	3.20	3.60	4.00	4.40	4.80	5.20	5.60	5.99	6.39	
0.20	3.42	3.84	4.26	4.68	5.10	5.53	5.95	6.37	6.79	
0.25	3.77	4.20	4.62	5.05	5.47	5.90	6.32	6.75	7.17	
0.30	4.18	4.60	5.02	5.43	5.85	6.27	6.68	7.10	7.52	
$w_1 + w_2$	$EAE + AEEA + H_2O$									
	$w_1(EAE)/w_2(AEEA) =$	= 9/1								
0.10	2.54	2.80	3.05	3.31	3.57	3.83	4.09	4.34	4.60	
0.20	2.57	2.93	3.30	3.67	4.04	4.41	4.77	5.14	5.51	
0.30	2.48	2.88	3.29	3.70	4.11	4.52	4.92	5.33	5.74	
	$w_1(EAE)/w_2(AEEA) =$	= 8/2								
0.10	2.41	2.69	2.96	3.24	3.51	3.79	4.06	4.34	4.61	
0.20	2.85	3.15	3.44	3.74	4.03	4.33	4.62	4.92	5.21	
0.30	2.70	3.06	3.42	3.79	4.15	4.52	4.88	5.24	5.61	
	$w_1(EAE)/w_2(AEEA) =$	= 7/3								
0.10	2.51	2.76	3.01	3.26	3.51	3.75	4.00	4.25	4.50	
0.20	2.71	2.95	3.20	3.44	3.68	3.93	4.17	4.42	4.66	
0.30	2.44	2.84	3.24	3.64	4.04	4.44	4.85	5.25	5.65	

EAE + AEEA mixture. Coefficients c_0 , c_1 , c_2 , and c_3 were given in Table 8. Density of aqueous EAE + AEEA was calculated by eq. 11 with AAD % of 0.018 and shown in Fig. 6.

Table 10 Activation molar enthalpy, entropy, and Gibbs free energy at 298.15 K for activation of viscous flow of EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O blend.

	2 ,	2 '	2
w_1	$\Delta H^*/(kJmol^{-1})$	$\Delta S */(Jmol^{-1} K^{-1})$	$\Delta G^*/(kJmol^{-1})$ at 298.15 K
		$EAE + H_2O$	
0.00	15.18	20.222	9.151
0.05	15.73	19.409	9.943
0.10	17.28	22.140	10.679
0.15	18.35	23.740	11.272
0.20	20.44	28.696	11.884
0.25	22.45	32.808	12.668
0.30	23.85	35.463	13.277
1.00	35.08	50.750	19.949
		$AEEA + H_2O$	
0.00	15.18	20.222	9.151
0.05	15.98	21.046	9.705
0.10	17.72	24.515	10.411
0.15	19.21	27.278	11.077
0.20	20.77	30.308	11.734
0.25	24.33	39.330	12.604
0.30	28.4	50.063	13.474
1.00	49	77.870	25.783
$w_1 + w_2$	EAE + AEEA + H	$_{2}O$ with $w_{1}(EAE)/w_{2}(A$	AEEA) = 9/1
0.10	16.81	22.543	10.089
0.20	17.03	19.931	11.088
0.30	17.37	17.684	12.098
	EAE + AEEA + H	$_{2}O$ with $w_{1}(EAE)/w_{2}(A$	AEEA) = 8/2
0.10	15.94	19.393	10.158
0.20	16.6	18.201	11.173
0.30	18.57	21.241	12.237
	EAE + AEEA + H	$_{2}O$ with $w_{1}(EAE)/w_{2}(A$	AEEA) = 7/3
0.10	14.68	14.91	10.158
0.20	16.19	16.46	11.173
0.30	19.82	24.90	12.237

3.4. Isobaric thermal expansion coefficient

The isobaric thermal expansion coefficient (α_p/K^{-1}) is defined as eq. 12:

$$\alpha_{p} = \left(\frac{1}{V}\right) \left(\frac{\partial V}{\partial T}\right)_{p} \equiv -\left(\frac{\partial ln\rho}{\partial T}\right)_{p} \tag{12}$$

In order to calculate isobaric thermal expansion coefficient, a second order polynomial equation (eq. 13) was fitted to the $ln\rho$ versus T data.

$$ln\rho = a + bT + cT^2 \tag{13}$$

Where, ρ is density (kg.m⁻³) and T is in K. a, b, and c are fitting parameters of the equation. Fitting parameters were calculated and listed in Table S1 in supporting information file. In terms of fitting parameters of eq. 13, isobaric thermal expansion coefficient was defined as eq. 14:

$$\alpha_p = -\left(\frac{\partial ln\rho}{\partial T}\right)_p = -(b + 2cT) \tag{14}$$

Isobaric thermal expansion coefficient of EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O was calculated by eq. 14 and given in Table 9.

3.5. Derived thermodynamic properties for activation of viscous flow

Thermodynamic properties for activation of viscous flow were derived using the viscosity and density data of pure liquids and aqueous mixtures. According to the Eyring and coworkers [29], the viscosity of a liquid strongly depends on the entropy that is dependent on the arrangement of molecules of liquid and its enthalpy. Viscosity of the aqueous mixture of EAE and AEEA could be expressed on the basis of enthalpy and entropy of activation of the system as follow:

$$\mu = \left(\frac{hN_A}{V_m}\right) exp\left(\frac{\Delta H^*}{RT}\right) exp\left(\frac{-\Delta S^*}{R}\right) \tag{15}$$

Where, μ was the viscosity. h, N_A , and R were Planck constant, Avogadro number, and universal gas constant, respectively. ΔH^* , ΔS^* , and T were molar activation enthalpy, molar activation entropy and temperature.

In order to find out ΔH^* and ΔS^* , eq. 11 was rearranged as following eq. 16

$$Rln\left(\frac{\mu V_m}{hN_A}\right) = \frac{\Delta H^*}{T} - \Delta S^* \tag{16}$$

We obtained activation molar enthalpy and activation molar entropy for each liquid mixture with assumption that these parameters were independent of temperature. Plot of $Rln\left(\frac{\mu V_m}{lN_A}\right)$ vs. $\frac{1}{T}$ was used to calculate ΔH^* and ΔS^* . Slope of the plot gave ΔH^* and intercept value was taken as ΔS^* . Using value of activation molar enthalpy and activation molar entropy further, activation molar Gibbs free energy (ΔG^*) was calculated by eq. 17.

$$\Delta G^* = \Delta H^* - T \Delta S^* \tag{17}$$

Activation molar enthalpy, activation molar entropy, and activation molar Gibbs free energy of aqueous EAE, aqueous AEEA, and aqueous EAE \pm AEEA were listed in Table 10.

With increase in the concentration of EAE in EAE + H₂O and AEEA in AEEA + H₂O positive ΔS^* increased that indicated the increase in the degree of disorderness of molecules associated with the activation process for the viscous flow. For EAE + AEEA + H₂O blend increasing total concentration of amines with constant EAE/AEEA ratio at 9/1 resulted in decrease in ΔS^* and that's why decrease in disorderness of associated molecules while for EAE/AEEA ratio 7/3 ΔS^* increased by increasing total concentration. ΔG^* was increased for EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O by increasing concentration of amines in the studied concentration range of this paper. This was the effect of strong association between EAE-H₂O, AEEA-H₂O, and EAE-H₂O-AEEA molecules.

4. Conclusions

Viscosity and density of EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O were measured in the temperature range 293.15 K to 333.15 K at atmospheric pressure. Viscosity of EAE + H_2O , AEEA + H_2O , and EAE +AEEA + H₂O was correlated to newly proposed model and AAD % was 1.515, 2.027, and 2.889, respectively. Excess molar volume (V^{E}) calculated for all aqueous mixtures and V^E of EAE + H₂O and AEEA + H₂O was fitted in Redlich-Kister eq. A new model was used to predict density of aqueous EAE + AEEA blend and AAD % for this model was 0.018. Isobaric thermal expansion coefficient of EAE + H_2O , AEEA + H_2O , and EAE + AEEA + H₂O was determined. Activation molar enthalpy (ΔH^*) , activation molar entropy (ΔS^*) , and activation molar Gibbs free energy (ΔG^*) at 298.15 K for aqueous EAE, aqueous AEEA, and aqueous EAE + AEEA were calculated on the basis of Eyring theory of viscosity of liquids. All the derived thermodynamic properties were positive at every composition of all studied liquid mixture. All liquid mixture studied in this paper (EAE + H₂O, AEEA + H₂O, and EAE + AEEA + H₂O) have industrial importance because they are used for post combustion CO₂ capture by absorption technique.

Declaration of Competing Interest

Authors have no any Conflict of interest.

Acknowledgment

The authors acknowledge the support and financial assistance provided by the Indian Institute of Technology (Banaras Hindu University) Varanasi to carry out the present work.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2021.115873.

References

- [1] M.K. Mondal, H.K. Balsora, P. Varshney, Progress and trends in CO_2 capture/separation technologies: a review, Energy 46 (1) (2012) 431–441.
- [2] C.H. Yu, C.H. Huang, C.S. Tan, A review of CO₂ capture by absorption and adsorption, Aerosol Air Qual. Res. 12 (5) (2012) 745–769.
- [3] A. Hartono, E.O. Mba, H.F. Svendsen, Physical properties of partially CO₂ loaded aqueous monoethanolamine (MEA), J. Chem. Eng. Data 59 (6) (2014) 1808–1816.
- [4] M. Shokouhi, A.H. Jalili, F. Samani, M. Hosseini-Jenab, Experimental investigation of the density and viscosity of CO₂-loaded aqueous alkanolamine solutions, Fluid Phase Equilib. 404 (2015) 96–108.
- [5] X. Wang, J. Chen, X. Wang, Densities and excess molar volumes of the binary system N-methyldiethanolamine+(2-aminoethyl) ethanolamine and its ternary aqueous mixtures from 283.15 to 363.15 K, Phys. Chem. Liq. 54 (4) (2016) 499–506.
- [6] A. Toumi, N. Hafaiedh, M. Bouanz, Thermodynamic properties of triethylamine + water liquid mixture from shear viscosity measurements, Fluid Phase Equilib. 278 (1–2) (2009) 68–75.
- [7] S.J. Hwang, J. Kim, H. Kim, K.S. Lee, Solubility of carbon dioxide in aqueous solutions of three secondary amines: 2-(butylamino) ethanol, 2-(isopropylamino) ethanol, and 2-(ethylamino) ethanol secondary alkanolamine solutions, J. Chem. Eng. Data 62 (2017) 2428–2435.
- [8] I. Folgueira, I. Teijido, A. García-Abuín, D. Gómez-Díaz, A. Rumbo, Carbon dioxide absorption behavior in 2-(ethylamino) ethanol aqueous solutions, Fuel Process. Technol. 131 (2015) 14–20.
- [9] S. Ma'mun, H.F. Svendsen, K.A. Hoff, O. Juliussen, Selection of new absorbents for carbon dioxide capture, Energy Convers. Manag. 48 (2007) 251–258.
- [10] S. Ma'mun, V.Y. Dindore, H.F. Svendsen, Kinetics of the reaction of carbon dioxide with aqueous solutions of 2-((2-aminoethyl) amino) ethanol, Ind. Eng. Chem. Res. 46 (2007) 385–394.
- [11] M. Moosavi, C.J. Sisco, A.A. Rostami, F.M. Vargas, Thermodynamic properties and CO₂ solubility of monoethanolamine + diethylenetriamine /aminoethylethanolamine mixtures: experimental measurements and thermodynamic modeling, Fluid Phase Equilib. 449 (2017) 175-185.
- [12] A. Bajpai, M.K. Mondal, Equilibrium solubility of CO_2 in aqueous mixtures of DEA and AEEA, J. Chem. Eng. Data 58 (2013) 1490–1495.
- [13] S. Kumar, M.K. Mondal, Equilibrium solubility of CO₂ in aqueous blend of 2-(diethylamine) ethanol and 2-(2-aminoethylamine) ethanol, J. Chem. Eng. Data 63 (2018) 1163–1169.
- [14] D. Pandey, M.K. Mondal, Equilibrium CO₂ solubility in the aqueous mixture of MAE and AEEA: experimental study and development of modified thermodynamic model, Fluid Phase Equilib. 522 (2020) 112766.
- [15] H. Kierzkowska-Pawlak, Kinetics of CO₂ absorption in aqueous N, N-diethylethanolamine and its blend with N-(2-aminoethyl) ethanolamine using a stirred cell reactor, Int. J. Greenh. Gas Cont. 37 (2015) 76–84.
- [16] I.M. Lampreia, Â.F. Santos, M.L.C. Moita, L.C. Nobre, Thermodynamic study of aqueous 2-(isopropylamino) ethanol. A sterically hindered new amine absorbent for CO₂ capture, J. Chem. Thermodyn. 81 (2015) 167–176.
- [17] F.I. Chowdhury, S. Akhtar, M.A. Saleh, M.U. Khandaker, Y.M. Amin, A.K. Arof, Volumetric and viscometric properties of aqueous solutions of some monoalkanolamines, J. Mol. Liq. 223 (2016) 299–314.
- [18] X. Liu, C. Zhu, T. Fu, Y. Ma, Volumetric and viscometric properties of ternary solution of (piperazine + 2-Amino-2-methyl-1-propanol+ H₂O) at T=(303.15-343.15) K, J. Mol. Liq. 310 (2020) 113187.
- [19] H. Gao, G. Gao, H. Liu, X. Luo, Z. Liang, R.O. Idem, Density, viscosity, and refractive index of aqueous CO₂-loaded and-unloaded ethylaminoethanol (EAE) solutions from 293.15 to 323.15 K for post combustion CO₂ capture, J. Chem. Eng. Data 62 (12) (2017) 4205–4214.
- [20] M. Mundhwa, R. Alam, A. Henni, Volumetric properties, viscosities, and refractive indices for aqueous 2-((2-aminoethyl)-amino)ethanol solutions from (298.15 to 343.15) K, J. Chem. Eng. Data 51 (2006) 1268–1273.
- [21] M. Stec, A. Tatarczuk, D. Spiewak, A. Wilk, Densities, excess molar volumes, and thermal expansion coefficients of aqueous aminoethylethanolamine solutions at temperatures from 283.15 to 343.15 K, J. Solut. Chem. 43 (2014) 959–971.
- [22] D. Pandey, M.K. Mondal, Experimental data and modeling for viscosity and refractive index of aqueous mixtures with 2-(methylamino)ethanol (MAE) and aminoethylethanolamine (AEEA), J. Chem. Eng. Data 64 (2019) 3346–3355.

- [23] O. Redlich, A.T. Kister, Algebraic representation of thermodynamic properties and
- the classification of solutions, Ind. Eng. Chem. 40 (1948) 345–348.

 [24] I.M.S. Lampreia, F.A. Dias, A.F.S.S. Mendonc, Volumetric properties of 2-ethylaminoethanol in water from 283.15 to 303.15 K, Phys. Chem. Chem. Phys. 5 (2003) 4869–4874.
- [255] A. García-Abuín, D. Gómez-Díaz, J.M. Navaza, A. Rumbo, Ö. Yaşaroılu, Density, speed of sound, viscosity, and excess properties of n -ethyl-2-pyrrolidone + 2-(methylamino)ethanol [or 2-(ethylamino)ethanol] from T = (293.15 to 323.15) K, J. Chem. Eng. Data 60 (2015) 795–800.

 [26] N.B. Kummamuru, D.A. Eimer, Z. Idris, Viscosity measurement and correlation of
- unloaded and CO₂-loaded aqueous solutions of n-methyldiethanolamine+ 2-amino-2-methyl-1-propanol, J. Chem. Eng. Data 65 (2020) 3072–3078.
- [27] G. Murshid, A.M. Shariff, K.K. Lau, M.A. Bustam, F. Ahmad, Physical properties of piperazine (PZ) activated aqueous solutions of 2-Amino-2-hydroxymethyl-1,3propanediol (AHPD + PZ), J. Chem. Eng. Data 57 (2012) 133–136.
- [28] R.H. Weiland, J.C. Dingman, D.B. Cronin, G.J. Browning, Density and viscosity of some partially carbonated aqueous alkanolamine solutions and their blends, J. Chem. Eng. Data 43 (3) (1998) 378-382.
- [29] S. Glasstone, K.J. Laidler, H. Eyring, The Theory of Rate Processes; the Kinetics of Chemical Reactions, Viscosity, Diffusion and Electrochemical Phenomena, McGraw-Hill Book Company, 1941.