

Viscosities of Pure and Aqueous Solutions of Monoethanolamine (MEA), Diethanolamine (DEA) and N-Methyldiethanolamine (MDEA)

Udara S. P. R. Arachchige^{1,a}, Neelakantha Aryal¹, Dag A. Eimer¹, and Morten C. Melaaen¹

¹ Telemark University College, Porsgrunn, 3901, Norway.

ABSTRACT

Viscosities of monoethanolamine, diethanolamine and methyldiethanolamine solutions were examined at a temperature range from (293.15 to 423.15) K for pure amines and (293.15 to 353.15) K for aqueous amines at different compositions. The experimental viscosities measured in this work are in good agreement with those reported in the literature.

INTRODUCTION

Alkanolamines have been used for the acid gas absorption process for a long period. The physical properties of those alkanolamines are important for process design. The main objective of this paper is to provide values for viscosities of different aqueous amines at different temperatures which are needed for the acid gas absorption process. Acid gas absorption process takes place at 313 K temperature. However, different operating temperatures are interested for high performance. Therefore, physical properties like viscosity of amines are important to calculate for different temperature range to perform mathematical calculations. The physical properties such as density, viscosity and solubility data of solvents are important for the acid gas absorption and regeneration process. Different types of amines are available, such as primary amines (MEA, DGA), secondary amines (DEA), tertiary amines (MDEA,

TEA), hindered amines (AMP) and cyclic amines (Piperazine). Important alkanolamines for industrial application are monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA) and N-methyldiethanolamine (MDEA) [1]. Aqueous MEA solutions are widely used for gas treating processes due to high reactivity, low operating cost and ease of reclamation [2]. However, the possibilities of using other solvents like DEA and MDEA have to be analyzed in order to lower the cost of CO₂ capturing. This work has been carried out to determine the effect of temperature on the viscosity of different types of amines at different concentration levels at atmospheric pressure. The experiments were performed to check the viscosities of monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA). The pure alkanolamine viscosity measurements were carried out for the temperature range (293.15 to 423.15) K and alkanolamine solution viscosity were measured in the range of (293.15 to 353.15) K. This represents an extension of previously reported data.

The measured data for the different kind of amines are compared with those available from the literature. The pure viscosity data for MEA, DEA and MDEA solutions are compared with data from Li and Lie1, DiGuilio et al. [3] and Mandal et al. [4] The data given by Amundsen et al. [5], Rinker et

al. [6] and Li and Lie [1] are used to compare the aqueous viscosity data for MEA, DEA and MDEA solutions. Agreement between the measurement data and literature values were found to be satisfactory.

The obtained results were used to produce correlations for amine viscosity at different temperatures. A number of correlations for computation of viscosity of liquids are presented in the literature. Vogel [7] has reported the simple three constant correlation for pure liquid viscosity and Viswanath and Natarajan [8] utilized a similar form for both dynamic and kinematic viscosities.

The equation presented by Vogel was modified by Goletz and Tassios [9] to include the boiling point of the substance. Dutt [10] described a similar idea to Goletz and has derived a correlation using density and boiling point of the component. Pure liquid viscosity can be represented by polynomial type equation mentioned by Girifalco [11] which was extended by Thorpe and Rodger [12].

Digullio et al. [3] have reported a three constant equation which is closer to Vogel's equation for pure amines' viscosity. Among those, Digullio's equation is selected for calculation of pure amines' viscosity due to less deviation when fitting experimental values.

The aqueous amine viscosities measured in this work are used to regress the polynomial for representation of amine viscosities. Teng et al. [13] and Chowdhury et al. [14] have reported the correlations for aqueous amine viscosity variation with molar concentration for specific temperature values. The correlation from Teng et al. [13] is selected for this study because of less deviation when fitted to experimental values.

EXPERIMENTAL SECTION

Dynamic viscosity was measured using MCR 101 Anton Paar double-gap rheometer. The viscometer was calibrated against the petroleum distillate and mineral oil calibration fluid from Paragon Scientific ltd. The calibration factor was decided according to the given temperature for the calibration liquid and experimental viscosity achieved during the calibration. The low-temperature measurements were achieved by applying cooling water supply to the rheometer setup at (293.15, 298.15 and 303.15) K. The MEA was purchased from Merck KGaA, DEA and the MDEA from Merck Schuchardt OHG. The purity of the chemicals in mass basis are 99.5%, 99% and 98% respectively for MEA, DEA and MDEA. The information about chemicals are given in Table 1. Without further purification, all these amines were used for experimental studies. De-gassed distilled water was used for preparation of aqueous amine solutions. All the experiments are performed at 1.01 bar operating pressure.

Table 1. Purity of the amines used in experiments.

Amine type	Mass Purity %	Supplier
MEA	99.5	Merck KGaA
DEA	99	Merck Schuchardt OHG
DEA	98	Merck Schuchardt OHG

RESULTS AND DISCUSSION

The results from the experimental work have been sub-divided into two sections: viscosities of pure amines and viscosities of aqueous amines.

Pure Amine Viscosities

Viscosities of pure MEA, DEA and MDEA from temperature range (293.15 to 423.15) K are tabulated in Tables 2, 3 and 4, respectively. The experimental viscosity results for pure amines, tabulated in Tables 3 to 5, are plotted in Fig. 1 as viscosity vs. temperature. As shown in the Fig. 1, viscosities of pure amines are decreasing with the increase of temperature. The viscosities of pure MEA, DEA and MDEA measured in this work agree well with the literature values taken from Li and Lie¹, DiGuilio et al. [3] and Mandal et al. [4], respectively. The average absolute deviations between the literature values and our data are (0.019, 1.21, 0.39) mPa·s for pure MEA, DEA and MDEA respectively. The deviations are in the range of experimental uncertainties which is given under the each table.

Table 2. Comparison of the Viscosities η of Pure MEA Measured in This Work with Literature Values from Temperature T = (293.15 to 423.15) K

T/K	This work	Li and Lie ¹	DiGuilio et al. ³	Mandal et al. ⁴
$\eta/\text{mPa}\cdot\text{s}$				
293.15	24.085			24.1
298.15	18.924			18.98
303.15	15.151	15.1088	14.86	15.11
313.15	10.006	10.0209	9.89	10.02
323.15	6.962	6.9715		6.972
333.15	5.037	5.0473	4.99	5.047
343.15	3.775	3.7793		3.779
353.15	2.919	2.912	2.90	2.912
363.15	2.334			
373.15	1.914		1.85	
383.15	1.586			
393.15	1.324		1.268	
403.15	1.105			
413.15	0.934		0.918	
423.15	0.806		0.796	

Standard uncertainties u are $u(T) = \pm 0.3$ K and the combined expanded uncertainties U_c are $U_c(\eta) = \pm 0.015$ mPa·s

Table 3. Comparison of the Viscosities η of Pure DEA Measured in This Work with Literature Values from Temperature T = (293.15 to 423.15) K

T/K	This work	DiGuilio et al.3	Mandal et al.4
$\eta/\text{mPa}\cdot\text{s}$			
293.15	889.655	890.5	890.5
298.15	562.315		566.3
303.15	380.18	356	383.9
313.15	192.85	200.9	188.2
323.15	106.936		119.5
333.15	57.304	61.22	57.69
343.15	36.177		35.09
353.15	22.974	23.4	22.43
363.15	15.655		
373.15	10.938	10.53	
383.15	7.702		
393.15	5.708	5.67	
403.15	4.345		
413.15	3.346	3.38	
423.15	2.687	2.7	

Standard uncertainties u are $u(T) = \pm 0.3$ K and the combined expanded uncertainties U_c are $U_c(\eta) = \pm 0.843$ mPa·s

Table 4. Comparison of the Viscosities η of Pure MDEA Measured in This Work with Literature Values from Temperature T = (293.15 to 423.15) K

T/K	This work	Li and Lie [1]	DiGuilio et al. [3]	Mandal et al. [4]
$\eta/\text{mPa}\cdot\text{s}$				
293.15	103.67		102.7	104.5
298.15	77.32			77.19
303.15	57.50	57.859	57.95	57.86
313.15	34.622	34.3085	34.02	34.31
323.15	22.402	21.6716	21.5	21.67
333.15	15.276	14.3856		14.39
343.15	9.965	9.9789		9.987
353.15	7.146	7.0875	7.151	7.088
363.15	5.818			
373.15	4.385		3.987	
383.15	3.362			
393.15	2.656		2.504	

403.15	2.122			
413.15	1.696		1.679	
423.15	1.375		1.406	

Standard uncertainties u are $u(T) = \pm 0.3$ K and the combined expanded uncertainties U_c are $U_c(\eta) = \pm 0.122$ mPa·s

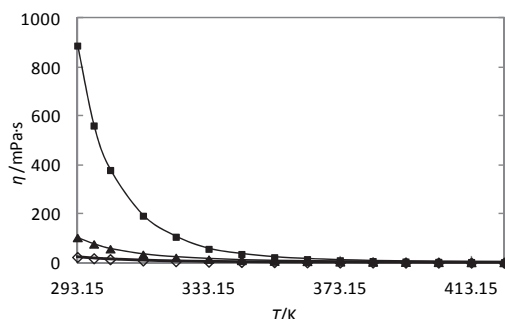


Figure 1. Pure amines' viscosity variation with temperature at (293.15 to 423.15) K: \diamond , MEA; \blacktriangle , MDEA; \blacksquare , DEA.

Aqueous Amine Viscosities

The viscosity data for aqueous MEA, DEA and MDEA solutions are presented in Tables 5, 6 and 7, respectively. The experiments were performed to measure viscosities of amines with concentrations in the range from mass fraction (0.1 to 0.9) for the temperature range (293.15 to 353.15) K. The aqueous MEA, DEA and MDEA viscosity data of Amundsen et al. [5], Rinker et al. [6] and Li and Lie [1] are compared with our experimental data (Figures 2, 3 and 4). The average absolute deviations are (0.08, 0.02, 0.02) mPa·s for aqueous MEA, DEA and MDEA, respectively and the maximum deviation is 0.25 mPa·s when compared against the literature sources. Hence, aqueous amine viscosities measured in this work are in good agreement with those reported by Amundsen et al. [6], Edward et al. [5], Li and Lie [1].

The dynamic viscosities of aqueous MEA, DEA and MDEA are plotted versus temperature in Figures 2, 3 and 4.

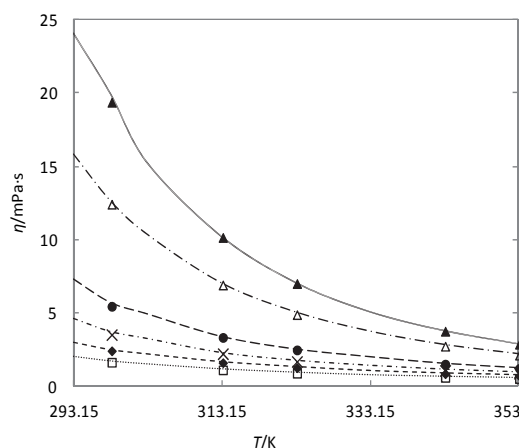


Figure 2. Comparison of aqueous MEA viscosity as a function of temperature. Lines are experimental data: \dots , 20%; $---$, 30%; $- \cdot -$, 40%; $- - -$, 50%; $- \cdot -$, 70%; $---$, 90%. Symbols refer to literature data [5]: \square , 20%; \blacklozenge , 30%; \times , 40%; \bullet , 50%; Δ , 70%; \blacktriangle , 90%.

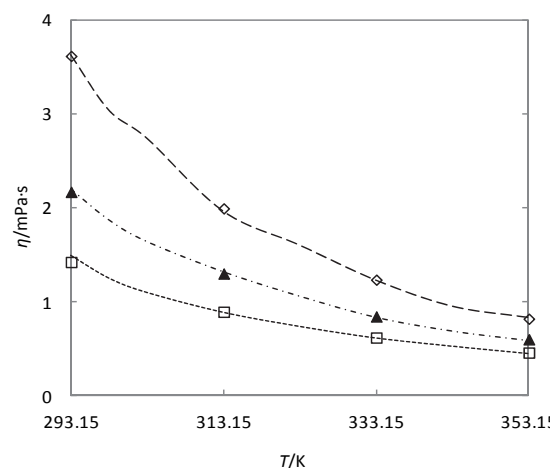


Figure 3. Comparison of aqueous DEA viscosity variation with temperature. Lines are experimental data: \dots , 10%; $---$, 20%; $- - -$, 30%. Symbols refer to literature data: \square , 10%; \blacktriangle , 20%; \blacklozenge , 30%.

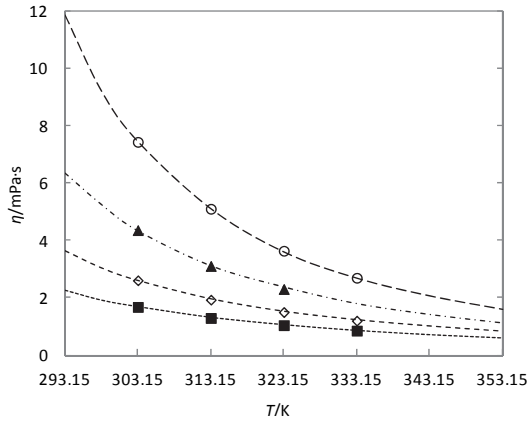


Figure 4. Comparison of aqueous MDEA viscosity variation with temperature: Lines are experimental data: ... , 20%; ---, 30%; - - -, 40%; — —, 50%. Symbols refer to literature data1: ■, 20%; ◇, 30%; ▲, 40%; o, 50%.

Table 5. Viscosities of Aqueous MEA Solutions Measured in This Work from Temperature T = (293.15 to 353.15) K as a function of mass fraction.

Temp	10 %	20 %	30 %	40 %	50 %	60 %	70 %	80 %	90 %
T/K	η/mPa·s								
293.15	1.442	2.005	2.990	4.667	7.345	11.295	15.774	21.003	24.023
298.15	1.299	1.702	2.489	3.765	5.547	9.062	12.602	16.290	19.904
303.15	1.121	1.501	2.195	3.279	4.960	7.417	10.313	13.148	15.200
313.15	0.909	1.169	1.671	2.311	3.423	5.291	7.024	8.950	10.220
323.15	0.715	0.945	1.338	1.782	2.566	3.771	5.055	6.273	7.090
333.15	0.626	0.775	1.065	1.434	2.041	2.973	3.757	4.585	5.110
343.15	0.520	0.671	0.903	1.168	1.594	2.243	2.854	3.449	3.830
353.15	0.449	0.589	0.779	0.977	1.291	1.774	2.226	2.676	2.936

Standard uncertainties u are u(T) = ± 0.3 K and the combined expanded uncertainties U_c are U_c(η) = ± 0.015 mPa·s

Table 6. Viscosities of aqueous DEA solutions measured in this work from T = (293.15 to 353.15) K as a function of mass fraction.

Temp	10 %	20 %	30 %	40 %	50 %	60 %	70 %	80 %	90 %
T/K	η/mPa·s								
293.15	1.499	2.204	3.630	6.515	12.367	25.810	53.677	143.30	353.71
298.15	1.259	1.875	3.040	5.221	9.782	19.237	39.150	97.87	245.43
303.15	1.106	1.644	2.740	4.420	8.008	15.322	31.187	73.47	162.68
313.15	0.896	1.323	1.965	3.145	5.492	10.020	17.889	41.02	88.541
323.15	0.747	1.063	1.604	2.310	3.884	6.501	12.206	24.08	52.487
333.15	0.624	0.838	1.231	1.720	2.869	4.807	7.732	14.67	29.894
343.15	0.537	0.691	0.954	1.341	2.150	3.552	5.595	10.11	19.317
353.15	0.459	0.590	0.833	1.099	1.685	2.659	4.216	07.11	13.035

Standard uncertainties u are u(T) = ± 0.3 K and the combined expanded uncertainties U_c are U_c(η) = ± 0.843 mPa·s

Table 7. Viscosities of aqueous MDEA solutions measured in this work from T = (293.15 to 353.15) K as a function of mass fraction.

Temp	10 %	20 %	30 %	40 %	50 %	60 %	70 %	80 %	90 %
T/K	η/mPa·s								
293.1	1.541	2.268	3.637	6.345	11.838	22.644	37.144	71.950	101.89
298.1	1.316	1.924	3.036	5.245	9.196	17.250	29.030	58.287	81.670

303.1	1.161	1.697	2.594	4.325	7.437	13.374	22.690	42.409	54.659
313.1	0.931	1.325	1.952	3.107	5.101	8.843	14.907	24.450	34.229
323.1	0.773	1.068	1.514	2.367	3.599	6.015	9.618	14.362	19.222
333.1	0.638	0.868	1.218	1.786	2.686	4.084	6.356	10.280	13.716
343.1	0.543	0.719	0.998	1.406	2.070	3.081	4.889	6.875	9.616
353.1	0.468	0.605	0.816	1.109	1.601	2.181	3.664	4.916	6.674

Standard uncertainties u are $u(T) = \pm 0.3$ K and the combined expanded uncertainties U_c are $U_c(\eta) = \pm 0.121$ mPa·s

CORRELATIONS

Correlation for Pure Amine Viscosities

The eq 1 can be used to correlate the pure amines' viscosities as a function of temperature [3]. Table 8 lists the coefficients of Eq 1 obtained by regression and also include the average absolute deviation between the experimental data and regression value.

$$\ln[\eta / (\text{mPa} \cdot \text{s})] = b_1 + \frac{b_2}{T / (\text{K}) - b_3} \quad (1)$$

The b_1 , b_2 and b_3 are constants. Average absolute deviation (AAD), $(\eta - \eta_{reg})$, is 0.05, 2.85 and 0.35 mPa·s respectively for pure MEA, DEA and MDEA.

Table 8. Constants for viscosity correlation (eq. 1).

Substance	b_1	b_2	b_3	AAD
MEA	-3.9303	1021.8	149.1969	0.05
DEA	-5.0559	1601.4	157.6487	2.85
MDEA	-4.7986	1476.9	136.3343	0.35

Correlation for Aqueous Amine Viscosities

Aqueous amine viscosities measured in this work are used to generate the polynomial for representation of amine viscosities using regression. The following polynomial (Eq 2) which is taken from the Teng et al. [13] is used for the regression.

$$\ln[\eta / (\text{mPa} \cdot \text{s})] = \ln \eta_0 + \sum_0^m a_k x^k \quad (2)$$

where η represents the viscosity of the binary solution while η_0 is the viscosity of pure water, and x the mole fraction of the amines. Pure water viscosity is taken from Li and Lie [1].

Calculated polynomial coefficients indicated by a_k are tabulated in Table 9. Deviation of calculated versus measured aqueous amines' viscosities are calculated as AAD and tabulated for different temperature values in the same table. For MEA, AAD values vary from 0.01 to 0.07 and similarly for DEA and MDEA the values vary from 0.01 to 0.08 and 0.002 to 0.05 mPa·s, respectively.

EVALUATION OF EXPERIMENTAL UNCERTAINTIES

Viscosity measurement uncertainties are arising as a combination of the temperature uncertainties, sample preparation uncertainties and instrument uncertainties. The MCR rheometer has itself specified temperature accuracy of ± 0.3 K. The samples were measured using an analytical balance which has accuracy of ± 0.0001 g (0.1mg). The estimated uncertainty in mass fraction of MEA, DEA and MDEA are 0.5%, 0.4% and 0.4% respectively.

Rheometer accuracy is mentioned as ± 0.002 mPa·s for viscosity measurements up to 453K temperature level. Total value of uncertainties for experimental viscosities calculated using combination of all mentioned uncertainties with root sum of

square method. The combined expanded uncertainties for pure amines calculated as ± 0.015 mPa·s, ± 0.843 mPa·s and ± 0.122 mPa·s for MEA, DEA and MDEA respectively. The combined expanded uncertainties for aqueous amines calculated as ± 0.015 mPa·s, ± 0.843 mPa·s and ± 0.121 mPa·s for MEA, DEA and MDEA respectively.

CONCLUSION

The viscosity of MEA, DEA and MDEA were measured at a temperature range from (293.15 to 423.15) K for pure amines and (293.15 to 353.15) K for aqueous amines. Aqueous amine viscosities were measured for a mass fraction range 0.10 to 0.90. As the temperature increased, viscosity of pure and aqueous amine solutions decreased. Moreover, the viscosity of aqueous amine solutions increased as the mass fraction of amine increased for a given temperature.

The measured viscosity data are in good agreement with literature data to the extent available. Two different available

correlations were used to fit the experimental values for pure and aqueous amines. Finally, calculated the deviation for all fitted correlations and correlation which gives lowest deviation is selected for this work. The deviation of regression values and measured values are varied as 0.05, 2.85 and 0.35 mPa·s respectively for pure MEA, DEA and MDEA amines and 0.002 to 0.08 mPa·s for aqueous amines. Deviations are in the range of expanded experimental uncertainties except pure DEA and MDEA deviation. Deviation of the pure DEA and MDEA is slightly higher than the calculated experimental uncertainties.

ACKNOWLEDGMENT

The support of Statoil's research center in Porsgrunn, especially Morten Tande, clarifying possible improvements to obtain more accurate viscosity results and instructions to the equipment used, is gratefully acknowledged.

Table 9. Coefficients of the Polynomial for the binary solutions between MEA, DEA and MDEA and water at different temperatures.

<i>T/K</i>	<i>amine</i>	a_0	a_1	a_2	a_3	a_4	a_5	a_6	<i>AAD</i>
293.15	MEA	0.0508	7.9019	18.0593	-98.1123	126.1164	-19.3461	-39.3815	0.01
	DEA	0.0316	9.8306	10.0557	0.5268	-213.0635	493.5456	-322.356	0.08
	MDEA	0.0112	12.5526	-36.2613	302.8214	-1152.5	1807	-994.287	0.05
298.15	MEA	0.1174	7.4531	7.8148	-30.2603	-26.0201	116.833	-77.2204	0.07
	DEA	-0.0174	11.2239	-5.4608	53.4597	-289.7572	529.0203	-316.2074	0.01
	MDEA	0.102	7.6836	41.9129	-253.5218	647.1595	-803.8854	383.0985	0.03
303.15	MEA	0.0931	6.2491	31.2894	-185.3907	411.4389	-440.4103	186.2332	0.01
	DEA	-0.0853	12.5851	-14.6494	44.9598	-143.5531	227.0852	-130.8371	0.04
	MDEA	0.0064	11.5024	-21.017	145.1402	-561.1386	896.9202	-502.7806	0.02
313.15	MEA	0.0137	10.0756	-38.9883	242.2987	-777.1085	1094.3	-557.2303	0.03
	DEA	-0.1249	15.2392	-76.9155	466.7223	-1433.3	2022.5	-1050.6	0.01
	MDEA	0.0147	10.366	-12.7884	67.2199	-248.1521	372.3297	-196.2842	0.01
323.15	MEA	-0.0887	11.8014	-51.7879	253.5452	-691.4558	883.0652	-421.2013	0.02
	DEA	-0.0719	12.2527	-31.7779	117.2989	-256.8023	276.6759	-115.2711	0.03
	MDEA	0.0477	8.7035	1.3063	-26.4952	44.9838	-46.1525	23.4403	0.02
333.15	MEA	0.0468	7.3836	-16.7796	129.9925	-495.2924	757.0601	-403.1124	0.01
	DEA	-0.0943	13.2504	-74.2482	458.2775	-1390.3	1924.8	-980.5914	0.02
	MDEA	0.029	8.2709	2.7312	-43.5071	80.7989	-47.998	-1.5757	0.002
343.15	MEA	-0.0835	11.4691	-59.8806	305.1176	-838.2553	1081	-521.4676	0.01
	DEA	-0.0176	10.0958	-51.1028	358.8246	-1155.2	1642.2	-847.6918	0.01
	MDEA	0.0542	6.3832	24.7464	-200.711	615.4432	-868.3154	453.2909	0.005
353.15	MEA	-0.1557	13.9979	-90.1129	442.9951	-1150.4	1429.6	-673.9559	0.01
	DEA	-0.067	10.638	-47.4738	263.1139	-742.4101	969.9961	-472.0667	0.01
	MDEA	0.1378	1.6411	86.4756	-594.4721	1794.4	-2487.2	1274.5	0.02

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